Institut für Wärmetechnik

TU Graz
Graz University of Technology

Dipl.-Ing. (FH) Gabriel Reichert M.Sc.

Optimization of Real-Life Performance of Firewood Stoves by Technological and Non-Technological Measures

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Supervisor
Univ.-Prof. Dipl.-Ing. Dr.techn. Christoph Hochenauer

Evaluator
Univ.-Prof. Dr.-Ing. habil. Matthias Gaderer

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For What is a Man Advantaged, if He Gain the Whole World, and Lose Himself, or be Cast Away?

The Bible - Luke 9, 25

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KURZFASSUNG

Für die Bereitstellung von Raumwärme aus erneuerbaren Energien stellen Einzelraumfeuerstätten wie Kaminöfen (EN 13240), Kachelöfen (EN 15544) oder Holzherde (EN 12815) oft verwendete Technologien dar. Obwohl die Nutzung dieser Technologien ein wichtiger Baustein zur Erreichung der europäischen CO_2-Reduktionsziele ist, werden im Praxisbetrieb beträchtliche Mengen an gesundheitsschädlichen Schadstoffen wie Kohlenstoffmonoxid (CO), organische, gasförmige Verbindungen (OGC) und Partikel (PM) emittiert. Die Ursache für erhöhte Emissionen im Praxisbetrieb liegt neben technologischen Aspekten vor allem in den Betriebsbedingungen vor Ort und der Betriebsweise durch den/ die AnwenderIn.

Das Ziel dieser Arbeit war es daher, geeignete technologische und nicht-technologische Optimierungsansätze für einen umweltfreundlicheren Praxisbetrieb zu erarbeiten und diese zu bewerten.

Zu Beginn dieser Arbeit wurde das typische NutzerInnenverhalten auf der Grundlage einer Umfrage ermittelt (Kapitel 2) und daraus kritische Betriebs- und Nutzungsparameter abgeleitet, welche im Praxisbetrieb optimiert werden sollten. Die Umfrageergebnisse zeigten hinsichtlich der Art und der Eigenschaften des verwendeten Brennstoffs, der Anzündmethode und der Luftklappeneinstellungen ein prinzipiell ähnliches NutzerInnenverhalten für die verschiedenen Typen von Scheitholz-Einzelraumfeuerstätten.

Auf der Grundlage der Umfrageergebnisse wurde der Einfluss von zwei relevanten Betriebsparametern, nämlich die Anzündmethode sowie die Zugbedingungen, hinsichtlich ihrer Emissionen und ihres Wirkungsgrads experimentell untersucht (Kapitel 3). Wie die Ergebnisse zeigten, ist das weithin empfohlene Anzünden „von oben“ im Vergleich zum Anzünden „von unten“ nicht für jeden Ofen die beste Wahl. Während ein eindeutiger Zusammenhang zwischen der Höhe des Kaminzugs und dem Wirkungsgrad bestätigt werden konnte, blieb dieser bei den gas- und partikelförmigen Emissionen aus.

Der Einsatz von Katalysatoren stellt eine Möglichkeit dar, gasförmige (CO/ OGC) und staubförmige (PM) Emissionen zu reduzieren. Hierzu wurde in Kapitel 4, 5 und 6 die integrierte Anwendbarkeit zweier verschiedener Oxidationskatalysatoren (keramisch bzw. metallisch: EnviCat®-Long Life Plus) in Scheitholz-Kaminöfen untersucht und das Potential der katalytischen Konvertierungseffizienz mit Hilfe eines speziellen Prüfstands, dem sogenannten DemoCat, ermittelt. Bei CO betrug dieses ~95%, ~60%

bei OGC und ~30% bei PM Emissionen. Ein weiteres Ergebnis der Untersuchung bestand in dem signifikanten Zusammenhang zwischen der Raumgeschwindigkeit sowie der katalytisch beschichteten Trägermaterialfläche und der CO-/ OGC-Konvertierung. Damit wurden beide Parameter neben der Temperatur als Schlüsselparameter für die Entwicklung integrierter Katalysatoren bestätigt. Auf die DemoCat Tests folgte die Validierung der katalytischen Wirkung beider Katalysatoren an fünf verschiedenen handelsüblichen Kaminöfen im realen Anwendungsfall. In diesen Öfen wurde der Einfluss der Katalysatorintegration experimentell analysiert und bewertet. Der aus den veränderten Druck- und Strömungsbedingungen resultierende Primäreffekt des integrierten Trägermaterials beeinflusste die Verbrennungsbedingungen nachteilig und verminderte die katalytische Effizienz. Demzufolge muss der Primäreffekt beim Einbau, d.h. bei der Integration des Katalysators in den jeweiligen Ofen, berücksichtigt werden. Trotz des Primäreffekts ergab sich für die Öfen mit einem integrierten Katalysator ein Netto-Emissionsreduktionspotential von ~90% für CO, ~30% für OGC und ~20% für PM-Emissionen. Demzufolge konnten zukünftige Emissionsgrenzwerte sogar innerhalb eines praxisnahen Prüfbetriebs eingehalten werden. Die experimentelle Bewertung der Anwendbarkeit beider Katalysatoren unter kritischen Betriebszuständen und während des Langzeitbetriebs wurde mit Sicherheits- und Langzeittests vervollständigt. Die Versuchsreihen zeigten ein potentielles Verblockungsrisiko und bestätigten die Notwendigkeit einer regelmäßigen Reinigung der integrierten Katalysatoren.

In Kapitel 7 wurde die offizielle Normprüfmethode für Scheitholz-Kaminöfen (EN 13240) mit der neu entwickelten praxisnahen „*beReal*“-Prüfmethode verglichen. Die Ergebnisse bestätigten für die „*beReal*“-Prüfmethode eine ausreichende Wiederholbarkeit und einen höheren Praxisbezug im Vergleich zum aktuellen Normprüfverfahren.

Die aus den vorhergehenden Untersuchungen gewonnenen Erkenntnisse führen zu der Empfehlung, externe NutzerInnschulungen als nicht-technologische Maßnahmen zur Verbesserung des Praxisbetriebs von Scheitholz-Einzelraumfeuerstätten sowie die Entwicklung von benutzerfreundlichen Kurzanleitungen für einen gerätespezifischen „best-practice“-Betrieb zu entwickeln. Die Kombination aus der Integration von Katalysatoren und einer automatischen Regelung der Verbrennungsluftzufuhr erscheint als vielversprechende technologische Optimierungsmaßnahme. Dieser Entwicklungsansatz einer Kombination primärer und sekundärer Optimierung kann sowohl gas- als auch staubförmige Emissionen im Praxisbetrieb signifikant reduzieren sowie den Wirkungsgrad der eingesetzten Technologien deutlich erhöhen.

Neben allen technologischen Verbesserungen ist die Einführung einer praxisnahen Prüfmethode ein wichtiger Baustein, um technologische Innovationen für einen optimalen Praxisbetrieb zu fördern. Die „*beReal*“-Prüfmethode könnte hierfür entweder als Norm oder als Qualitätslabel mit einer konsequenten Marktüberwachung Anwendung finden und es damit EndkundInnen ermöglichen, die Produktqualität eines Ofens im Praxisbetrieb besser einzuschätzen und zwischen guten und schlechten Produkten zu unterscheiden.

ABSTRACT

Firewood room heating appliances, like firewood roomheaters (EN 13240), tiled stoves (EN 15544) and residential biomass cookers (EN 12815) are commonly used for supplying residential renewable heat. Although they are important for reaching European CO_2 emission reduction targets, they were identified as major source of harmful emissions, like carbon monoxide (CO), organic gaseous compounds (OGC) as well as particulate matter (PM), especially in real-life operation. Beside technology, the operating conditions and the user behavior are essential reasons for increased emissions, especially in real-life operation.

Therefore, this thesis aimed at identification and evaluation of technological and non-technological optimization approaches for a better and environmental friendly real-life performance.

At first, a user survey was performed in order to assess the typical user behavior and to identify critical operational aspects which should be optimized in real-life operation (chapter 2). The results of the survey showed principally similar user behavior of all considered types of appliances regarding most relevant operational characteristics, i.e. kind, properties and amount of used fuels, ignition procedure and air valve settings.

Based on the findings of the survey two critical operational aspects, the ignition technique and the flue gas draught conditions were experimentally evaluated regarding their impact on emissions and thermal efficiency (chapter 3). The combustion experiments comparing top-down and bottom-up ignition technique showed no general advantage of the generally recommended top-down ignition method for all stoves. The evaluation of the effect of draught conditions showed a clear correlation between draught conditions and thermal efficiency. For gaseous emissions positive and negative effects have been observed for different technologies. Therefore, no clear correlation towards gaseous emissions was observed. Regarding PM emissions no effect of increased draught conditions was evident.

In chapter 4, 5, and 6 the applicability of two different types of oxidizing honeycomb catalysts (ceramic and metallic: EnviCat®-Long Life Plus) integrated in firewood stoves was evaluated. This technological measure is regarded as effective to reduce gaseous (CO/ OGC) and particulate matter (PM) emissions. The catalytic efficiency, evaluated in a special tailored test facility, called DemoCat, showed the potential to convert ~95% of CO, ~60% of OGC and ~30% of PM emissions. The conversion rates of CO and OGC emissions correlated with the space velocity and the coated area of honeycomb

carriers which represent together with the temperature key parameters for the integration design. Following the DemoCat tests, integrated catalytic converters were validated in real applications. Therefore, both types of catalysts were integrated in five different firewood stoves and the impact on emissions was experimentally evaluated. The integrated carrier material influenced pressure and flow conditions during the combustion process resulting in a worse combustion performance. This primary effect of the carrier material lowered the catalytic efficiency and should therefore be considered during the development process. However, a net emission reduction potential of ~90% for CO, ~30% for OGC and ~20% for PM emissions was evident. Moreover, future emission limit values were met even in a close-to-real-life test cycle. Finally, the applicability of the integrated catalysts under critical heating operating conditions and under long term operation was evaluated experimentally by a safety and long term test series. These combustion experiments showed the risk of blocking and clarified the need for a regular cleaning of the integrated catalysts.

In chapter 7 comparative evaluations of the official-type-test method (EN 13240) and a new real-life oriented test protocol ("*beReal*") are presented. The results showed that the "*beReal*" test concept has a sufficient repeatability and is capable to evaluate the performance of firewood roomheaters closer to real-life compared to the existing EN test protocol.

Concluding, non-technological optimization measures, like external training arrangements and the development of user friendly manuals focusing on an appliance specific best-practice heating operation, are suggested. The application of integrated catalysts combined with an automatically controlled combustion air supply appears as a promising technological measure. This approach could decrease gaseous and particulate emissions and increase the thermal efficiency in real-life operation significantly. Moreover, primary and secondary optimizations are combined and synergetic effects are used.

Finally, an implementation of a real-life oriented test protocol, e.g. the "*beReal*" test protocol, as a quality label or standard combined with effective market surveillance should be considered as an instrument to push innovation and technological development further and to enable better differentiation of good and poor products for the end-customer.

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ACRONYMS

beReal	Real-life oriented test protocol for firewood roomheaters
cat	Catalyst
cc-rec	Rectangular shaped ceramic catalyst
cc-round	Round shaped ceramic catalyst
CEN	European Committee for Standardization
cpsi	Cells per square inch
CV	Coefficient of variation
d.b.	Dry based
E_a	Activation energy
EC	Elemental carbon
EF	Emission factor
eff	Effective
EDX	Energy dispersive X-ray spectroscopy
ELV	Emission limit values
EN	European norm
ESP	Electrostatic precipitator
FGC	Flue gas composition
FID	Flame ionization detector
IC	Ion chromatography
ICP-MS	Inductively coupled plasma mass spectrometry
max	Maximum
mc-rec	Rectangular shaped metallic catalyst
mc-round	Round shaped metallic catalyst
n	Number of respective parameter (e.g. answers, tests or appliances)
n.d.	Not determined
NDIR	Nondispersive infrared
OC	Organic carbon
OGC	Organic gaseous compounds
ott	Official-type-test
PAC	Polyaromatic compounds
PAH	Polycyclic aromatic hydrocarbons

PM	Particulate matter
$PM_{2.5}$	Particulate emissions with an aerodynamic diameter < 2.5 µm
PM_{10}	Particulate emissions with an aerodynamic diameter < 10 µm
QUG	Quick-User-Guide
R&D	Research and development
SEM	Scanning electron microscope
SPSS	Software for statistical analysis
STP	Standard temperature and pressure (0 °C = 273.15 K / 101,325 Pa)
TC	Total carbon
THC	Total hydrocarbon
TS	Technical specification
VDI	"Verein Deutscher Ingenieure"
w.b.	Wet based
Ø	Average

SYMBOLS

Symbols	**Description**	**Unit**
A	Residues, referred to the mass of the fired test fuel	ma.-%
A	Area	m²
a	Ash content of fired fuel	ma.-% g/kg
B	Combustible constituents in the residues, referred to the mass of residues	ma.-%
C_f	Dry gas meter calibration factor	-
C_{pmd}	Specific heat of dry flue gas in standard conditions	kJ/(K m³)
C_{pmH2O}	Specific heat of water vapor in the flue gas in standard conditions	kJ/(K m³)
C_r	Carbon content of the residue	ma.-%
c	Flue gas velocity	m/s
c_p	Specific heat capacity	kJ/(kg K)
CCR	Catalytic conversion rate	%
CV	Coefficient of variation	%
Δp	Static pressure difference/ flue gas draught	Pa
d	Dimensions (i.e. diameter or depth)	m mm
E	Emission concentration	mg/m³
η	Thermal efficiency	%
H_u	Net calorific value	kJ/kg MJ/kg
L_{min}	Stoichiometric minimum combustion air demand at STP conditions	m³/kg
λ	Lambda value (air/fuel ratio; air excess)	
m	Mass	kg
$\dot{m}$	Mass flow	kg/s
$NERR$	Net emission reduction rate	%
p	Probability value of statistical tests	-
p	Absolute pressure	Pa

PE	Primary effect	%
q	Proportion of flue gas losses	%
Q	(Specific) heat quantity	kJ kJ/kg
$\dot{Q}$	Heat flow	kJ/s
r_P	Pearson's correlation coefficient	-
ρ	Density	kg/m³
σ	Standard deviation	variable
T	Temperature	°C
τ	Time	s
τ_k	Kendall's Tau correlation coefficient	-
V	Volume	m³
$\dot{V}$	Volume flow	m³/s
w	Moisture content of fired fuel	ma.-% kg/kg
$x_{combustibles}$	Combustibles content of residues	kg/kg

CHEMICAL SYMBOLS

Chemical symbol	**Species**
Al_2O_3	Aluminum oxide
C	Carbon
CC	Carbonate carbon
Ca	Calcium
Cl	Chlorine
CH_4	Methane
$C_{10}H_8$	Naphthalene
CO	Carbon monoxide
CO_2	Carbon dioxide
Fe	Iron
K	Potassium
Mg	Magnesium
Na	Sodium
NH_4	Ammonium
Ni	Nickel
NO_3	Nitrate
O_2	Oxygen
P	Phosphorous
Pb	Lead
Pd	Palladium
Pt	Platinum
S	Sulfur
SO_2	Silica
SO_4	Sulfate
Zn	Zinc

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1 INTRODUCTION

This chapter presents an overview about the journal publications which are the basis for this PhD thesis and specifies the author´s own contribution of the total work.

Furthermore, it aims to provide the relevant background information about the utilization of firewood room heating appliances in Europe. An overview about the most important reasons for increased emissions and decreased thermal efficiency in real-life operation is presented and the need for optimization of real-life performance is clarified. Based on the background information the objectives and research questions of this thesis are derived. Finally, the impact of the work for both, scientific community and industry, is presented.

This PhD thesis bases on six published peer-reviewed journal papers summarized in the following list:

- G. REICHERT, C. SCHMIDL, W. HASLINGER, M. SCHWABL, S. AIGENBAUER, M. WÖHLER, C. HOCHENAUER, Investigation of user behavior and assessment of typical operation mode for different types of firewood room heating appliances in Austria, Renew. Energy 93 (2016) 245-254. https://doi.org/10.1016/j.renene.2016.01.092.
- G. REICHERT, H. HARTMANN, W. HASLINGER, H. OEHLER, R. MACK, C. SCHMIDL, C. SCHÖN, M. SCHWABL, H. STRESSLER, R. STURMLECHNER, C. HOCHENAUER, Effect of draught conditions and ignition technique on combustion performance of firewood roomheaters, Renew. Energy 105 (2017) 547-560. https://doi.org/10.1016/j.renene.2016.12.017.
- G. REICHERT, C. SCHMIDL, W. HASLINGER, H. STRESSLER, R. STURMLECHNER, M. SCHWABL, N. KIENZL, M. WÖHLER, C. HOCHENAUER, Long term durability and safety aspects of oxidizing honeycomb catalysts integrated in firewood stoves, Biomass Bioenergy 105 (2017) 428-442. http://dx.doi.org/10.1016/j.biombioe.2017.07.018.
- G. REICHERT, C. SCHMIDL, W. HASLINGER, H. STRESSLER, R. STURMLECHNER, M. SCHWABL, M. WÖHLER, C. HOCHENAUER, Catalytic efficiency of oxidizing honeycomb catalysts integrated in firewood stoves evaluated by a novel measuring

methodology under real-life operating conditions, Renew. Energy 117 (2018) 300-313. https://doi.org/10.1016/j.renene.2017.10.065.

- G. REICHERT, C. SCHMIDL, W. HASLINGER, H. STRESSLER, R. STURMLECHNER, M. SCHWABL, C. HOCHENAUER, Novel method evaluating real-life performance of firewood roomheaters in Europe, Energy Fuels 3 (2), (2018) 1874-1883. https://doi.org/10.1021/acs.energyfuels.7b03673.

- G. REICHERT, C. SCHMIDL, W. HASLINGER, H. STRESSLER, R. STURMLECHNER, M. SCHWABL, M. WÖHLER, C. HOCHENAUER, Impact of oxidizing honeycomb catalysts integrated in firewood stoves on emissions under real-life operating conditions, Fuel Process. Technol. 177 (2018) 109-118. https://doi.org/10.1016/j.fuproc.2018.04.016.

Paper [1] presents a survey which evaluates the user behavior of different types of firewood room heating appliances. The questions of the survey were elaborated by the author with support of Christoph Schmidl, Manuel Schwabl, Stefan Aigenbauer and Marius Wöhler. The survey was conducted as a subcontract by students of the University of Wiener Neustadt. The data evaluation and statistical analysis was conducted by the author.

In paper [2] two different ignition techniques, the top-down ignition and the bottom-up ignition technique were compared. Moreover, the effect of different draught conditions on combustion performance was systematically evaluated at different firewood stoves. The combustion experiments for the ignition tests were designed by the contributions of all authors (except Walter Haslinger and Professor Christoph Hochenauer). The ignition tests of one of the two selected stoves as well as the draught condition tests of one of the three selected stoves and the basic data evaluation were conducted at the TFZ Straubing by Heike Oehler and Robert Mack with support of Hans Hartmann. The ignition tests of the second stove and the tests of the remaining two stoves referring to the draught conditions tests were done by the author with assistance of Harald Stressler. The detailed data evaluation and statistical analysis was carried out by the author with assistance of Rita Sturmlechner.

Paper [3] presents safety and long term tests of two types of oxidizing honeycomb catalysts integrated in three firewood stoves. The test approach was designed by the author with support of Christoph Schmidl and Manuel Schwabl. The combustion experiments, data evaluation and statistical analysis were mainly conducted by the author with assistance of Harald Stressler (experiments) and Rita Sturmlechner (data evaluation). The chemical analysis of deposited material on the catalysts (EC/ OC/ TC) was done by Norbert Kienzl.

In paper [4] the catalytic efficiencies of two types of oxidizing honeycomb catalysts were evaluated in a special tailored test facility (DemoCat). The DemoCat was designed by the author and constructed by Harald Stressler. The test approach was elaborated by the author with support of Christoph Schmidl and Manuel Schwabl. The combustion experiments, data evaluation and statistical analysis were mainly conducted by the author with assistance of Harald Stressler (experiments) and Rita Sturmlechner (data evaluation).

Paper [5] presents a comparative evaluation of two test protocols for firewood roomheaters – the official type test standard and a newly developed test protocol focusing on a real-life evaluation of the appliances performance. The test approach was designed by the author with assistance of Christoph Schmidl and Manuel Schwabl. The combustion experiments were conducted by Harald Stressler. The data evaluation and statistical analysis were mainly conducted by the author with assistance of Rita Sturmlechner.

Paper [6] presents the experimental evaluation of two types of oxidizing honeycomb catalysts integrated in five different serial-production firewood stoves under real-life operating conditions. The test approach was designed by the author, Christoph Schmidl and Manuel Schwabl. The combustion experiments were carried out by the author with the support of Marius Wöhler and Harald Stressler. Data evaluation and statistical analysis was done by the author with assistance of Rita Sturmlechner.

The interpretation of results of paper [1] – [6] was done by the author with support of Christoph Schmidl, Walter Haslinger and Professor Christoph Hochenauer.

All papers were written by the author under the supervision of Professor Christoph Hochenauer.

In total the author's own contribution to the whole work is roughly estimated more than 75%.

1.1 Background information[1]

The oldest and even the most common way of woody biomass utilization is the provision of thermal heat for room heating or cooking purposes by using small-scale and batch-wise operated firewood stoves [7] [8]. Worldwide more than 2.7 billion people rely on firewood for heating and cooking [9]. In developing countries cooking is the predominant purpose for the use of firewood [10]. In Europe, most widespread residential wood combustion heaters are batch-wise operated direct room heating devices such as open fireplaces, closed fireplaces, insets, roomheaters and cookers operated with firewood. The stock of this kind of direct room heaters is estimated to be more than 65 million appliances [11], in Austria it is around 1.5 million appliances [12].

For those manually operated wood stoves there are several European standards which are specifically defined for each type of technology:

- EN 13240: Roomheaters fired by solid fuel – Requirements and test methods [13]
- EN 13229: Inset appliances including open fires fired by solid fuels – Requirements and test methods [14]
- EN 12815: Residential cookers fired by solid fuel – Requirements and test methods [15]
- EN 15250: Slow heat release appliances fired by solid fuel – Requirements and test methods [16]
- EN 15544: One off Kachelgrundöfen/Putzgrundöfen (tiled/mortared stoves) – Dimensioning [17]

Although these types of appliances are fairly relevant for reaching European CO_2 emission targets [18]-[20], they were identified to cause high amounts of gaseous as well as particulate matter emissions [21]-[23] which can seriously affect public health [24]-[28]. Especially for fractions of harmful fine particle emissions with aerodynamic diameter smaller than 10 µm (PM_{10}) residential wood combustion has been identified to be a major source of local air pollution, especially in the winter half year in Europe [29]-[32].

Emissions from firewood combustion in room heating devices have a high health impact, because of the emitted respirable dust. Additionally, PM emissions include carcinogenic compounds, e.g. polyaromatic compounds (PAC), like benzo(a)pyrene. An exposure to those emissions can lead to irreversible health diseases till premature death [23]-[26].

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[1] Segments of this section have already been published in [1]-[6].

In Austria, residential wood combustion has been identified to be responsible for around 25% of PM_{10} emissions in 2013 [33]. In Europe, the residential sector contributes with 57% of total $PM_{2.5}$ pollution and is therefore the most relevant polluter of $PM_{2.5}$ emissions [9]. Hence, PM emissions have become an important topic in the European Union since several studies present a regular violation of the European thresholds for PM_{10} in ambient air, which means a maximum annual average value of 40 mg/m^3 or at maximum 35 times of exceedance of daily average value of 50 mg/m^3 per year [34]-[36]. Consequently, public authorities are forced to implement effective measures for emission reduction.

One possibility is the tightening of the emission limit values (ELV) which have to be met during the official-type-testing according to the respective EN standard. At the beginning of the year 2022, new firewood operated room heating appliances have to comply with specific ELV which were elaborated during the Ecodesign and Energy labeling process of the European Union. Accordingly, the maximum emissions measured during EN standard type testing for new stove technologies are 1500 mg/m^3 for CO, 120 mg/m^3 for OGC and 40 mg/m^3 for PM emissions (all ELV at STP conditions: 273.15 K/ 101,325 Pa, measured in the dry flue gas and referred to 13 vol.-% O_2) [37]. These ELV will set equal requirements in Europe and it might be quite challenging for stove manufacturers to comply with these ELV. In this thesis emission concentrations are generally specified in mg/m^3, at STP conditions (273.15 K/ 101,325 Pa), measured in the dry flue gas and referred to 13 vol.-% O_2.

However, to best possibly guide the respective policy measures, there is a need to understand the reasons for the currently high emission level of batch-wise operated firewood room heating appliances in real-life operation. Consequently, the implementation of effective primary and secondary measures for emission reduction is required. Generally, the reasons for increased gaseous and particulate real-life emissions compared to expectations according to current standard type test results can be categorized in four different groups:

- User behavior reasons
- Reasons referring to operating conditions
- Technological reasons
- Type testing reasons

User behavior reasons

All parameters of the operation which can be directly affected by the user are defined by the term user behavior including following aspects:

- Physical and chemical fuel properties
- Ignition technique
- Fuel amount per batch
- Adjustment of air valve settings for combustion air supply
- Number of batches performed during one heating operation cycle

The above listed aspects, comprising the user behavior, can seriously affect the emission level during the combustion process. For example, incorrect fuel dimensions and fuel properties [38] [39], an overloading of fuel [40] or incorrect adjusted air valve settings [41]-[43] result in increased emission levels. Additionally, different types of firewood species can result in different emissions and PM emission compositions [44] [45].

Furthermore, the ignition technique, as an important aspect of user behavior, was found to have a high effect on emission release of gaseous and particulate matter emissions [46]-[48]. In principal, two different ignition techniques can be distinguished – the top-down ignition technique and the bottom-up ignition technique.

Top-down ignition means that firewood pieces are directly placed on the grate. The kindling material is placed above the firewood pieces and on the top of the kindling material the specific starting aids are located. Then, the starting aids are lighted and the combustion process is induced by a subsequent downward ignition of kindling material and the firewood pieces on the grate. In contrast, bottom-up ignition means the placement of kindling material and starting aids directly on the grate below a few pieces of firewood. Then, the starting aids on the grate are lighted and the combustion process is induced upward the fuel batch.

As presented in the studies of NUSSBAUMER et al. [46] [47] and HARTMANN et al. [48] PM emissions of the ignition phase were reduced by about 50% – 80% and CO emissions by about 60% by applying the top-down ignition technique instead of the traditional bottom-up ignition technique. The above mentioned studies [46]-[48] hypothesize that the moderate progress of combustion processes of top-down ignition technique would be advantageous since it would lead to slower pyrolysis and gasification processes and hence reduce the risk of incomplete burn out of gaseous intermediates due to low combustion temperatures above the fuel bed. However, there are only a small number of appliances tested and a systematic evaluation of both ignition techniques at different stoves is still missing.

Reasons referring to operating conditions

Operating conditions define parameters referring to the infrastructure of the heating appliance which cannot be directly affected by the user during operation. Relevant operating conditions concerning emissions and efficiency are mainly the flow conditions induced by natural draught. The most relevant characteristics for induced draught conditions during heating operation are the properties of the chimney system, like material and dimensions (diameter and height), weather conditions and flue gas temperature [46] [49]. Generally, a sufficient draught level in the chimney is important to enable stable and safe operation. Under type testing conditions, for firewood roomheaters, a constant draught of 12 ± 2 Pa is required [13]. If the flue gas draught exceeds 12 Pa the volume flow of combustion air supply increases. Consequently, combustion conditions are affected and hence emission formation and thermal efficiency are influenced with lower thermal efficiency at higher draught conditions [49]-[51]. However, so far no clear correlation between draught and gaseous as well as particulate emissions was observed.

Technological reasons

The used technology has a strong impact on the emission level regarding gaseous carbon monoxide (CO), organic gaseous compounds (OGC) and polycyclic aromatic hydrocarbons (PAH) as well as particulate (PM) emissions [22] [27]. Modern types of appliances are featured with implemented primary measures, like a well dimensioned and insulated combustion chamber design and an air staging concept [52] [53]. These primary measures aim at emission prevention by enabling optimal combustion conditions, especially during the intermediate phase of a combustion batch [54]. According to KELZ et al. [27] and BRUNNER et al. [52] old technologies are not featured with the mentioned primary optimization measures. For example, they have no post combustion chamber and no sophisticated air staging concept for combustion air supply. Those technologies emit around two times higher gaseous CO and OGC emissions and about 40% higher PM_1 emissions. The difference regarding PAH emissions was even higher by a factor of 18. Unfortunately, the stock of firewood room heating appliances in Europe is dominated by old and not state-of-the-art combustion systems [22] [27]. However, even modern and primary optimized stoves have combustion phases characterized by increased emissions, for example during the start-up and burn-out phase of a combustion batch or during the ignition batch [55] [56]. Furthermore, modern types of firewood stoves are prone to a worse emission and efficiency performance when they are improperly operated, e.g. deviations to the suggested operation in the manual [49]. Therefore, the use of secondary emission abatement technologies, like filter precipitators, electrostatic precipitators (ESP) as well as catalysts is a possibility for reduction of emissions.

Catalytic converters, in most cases used as retrofitted application [45] [57] [58], are feasible for an emission reduction. An advantage of catalysts in relation to other mentioned secondary devices, like filters or ESPs, is the reductive effect on both, gaseous and particulate matter emissions [59] [60]. In addition, they operate also under non-optimal phases, e.g. during ignition (if the temperature is sufficiently high), during start-up and burn-out phases, or during critical operating conditions due to user behavior reasons [61]. In general, catalysts do not need electrical power supply. However, a certain temperature level is necessary to enable the catalytic process [61]-[63]. In some studies an external heating of the catalytic system was applied for that reason [58] [64]. Many oxidizing catalysts work effectively when they are operated at temperatures of 300 °C – 450 °C, especially for emission conversion of OGC emissions [62] [63] [65]. Without an external heating device this temperature level is usually not reachable with retrofit applications or only for short time duration in firewood stoves. Furthermore, the duration until the light-off temperature of the retrofitted catalyst is reached lasts around 20 min [59] [61]. An alternative option would be to integrate the catalytic system, for example an oxidizing honeycomb catalyst, in the combustion appliance where flue gas temperatures are sufficiently high and light-off temperatures of the catalyst are reached faster. Honeycomb catalysts are commercially available and they are frequently used in the United States and Canada in firewood stoves [45] [66]. In both countries, standard type test protocols provide even special limits and test procedures for stoves equipped with catalytic systems [67] [68]. Typically, they are used with a bypass in order to avoid safety risks or malfunction due to blockage by deposited material.

However, since up to date there is no legal requirement in most European countries for using such secondary abatement technologies they are only rarely used. Additionally, most of the mentioned secondary emission abatement technologies are still under development and are not yet commercially available [69].

Type testing reasons

In general, the testing of new products should guarantee a minimum of product quality concerning operation performance and safety aspects. Testing conditions and procedures shall be well-defined and transparent to offer equal opportunities for manufacturers. The EN standards for firewood room heating appliances cover definitions and requirements regarding used materials, product declarations, performance testing, and safety testing. During the performance test CO emissions and thermal efficiency are assessed. In addition, the EN testing procedure is used to assess further emissions, like OGC and PM emissions, to comply with national legal requirements.

Standard test methods for firewood stoves evaluate the appliance performance typically only at nominal load. The ignition of the first fuel batch and the heating up of the stove are not considered, except for the testing procedure for slow heat release appliances (EN 15250 [16]). Generally, ignition, different loads, load changes, and the cooling-down phase are not included in the EN test protocols. Consequently, testing according to EN test protocols in a quasi-stationary operation mode and a thermal equilibrium usually leads to best possible emission and efficiency results and should be highly repeatable.

However, operating conditions referring to typical user behavior and transient conditions, like ignition, heating up, and cooling down which occur in each heating operation in real-life, are not evaluated. This leads to official-type-test (ott) results of low emissions and a high thermal efficiency. However, these results are never reached during real-life operation [49] [70]. This effect is well-known for many different product classes. Recently, quite big differences between laboratory and field performance were found for the car industry ("diesel gate"). However, in the case of stove manufacturers the differences do not originate from illegal testing software, as has been reported for car manufacturers, but simply result from different conditions between laboratory testing and field operation.

Consequently, the differentiation of product qualities concerning emissions and thermal efficiency which refer mainly to transient operating conditions and user-related aspects is poor and customers have insufficient information for their selling decision. Furthermore, legal authorities cannot achieve the desired effect of reduced emissions and increased efficiency in the field by tightening ELV. The same criticism was also mentioned for biomass boilers in previous publications [71] [72]. Therefore, there is an interest to develop and implement new test methods that are capable of evaluating the performance of appliances closer to real-life operation to push technological innovation and technological development further and to enable better differentiation of good and poor products for the end-customer. Accordingly, the need for advanced standard type testing procedures that better reflect the real-life behavior and are less endangered by manipulation and rooms for interpretation has reached the awareness of the European public administration [73].

1.2 Objectives and research questions

In this thesis aspects of all reasons mentioned above are covered. Hence, the overall objective was to identify technological and non-technological measures which are suitable to improve the real-life operation performance regarding low emissions and high efficiency of firewood room heating appliances. In detail, the objectives can be structured in four different topics:

1) **Assessment of typical user behavior of manually operated room heating appliances and deduction of effective and customer friendly technological and non-technological optimization approaches for a better and more environmental friendly real-life performance**

   This work was executed by a survey among attendees of a tradeshow. All of the respondents were users of biomass room heating appliances. The main questions to answer were, "*Do the users operate their stoves according to the manufacturers manual?*"; "*What fuel do they typically use?*"; "*How often do they change air valve settings during heating operation?*"; "*How much fuel is typically used for different technologies?*"

   The outcomes of the survey were analyzed taking further available literature data into account. The typical user behavior was defined and based on that evaluation, critical aspects of heating operation towards emissions and thermal efficiency were identified.

   The work dealing with that topic is presented in chapter 2.

2) **Experimental evaluation of the effect of important aspects of user behavior (i.e. ignition technique) and operating conditions (i.e. flue gas draught conditions) regarding emissions and thermal efficiency**

   So far, no studies about systematic evaluation of flue gas draught conditions and ignition techniques at different devices were available. Therefore, comprehensive combustion tests at different serial-production firewood roomheaters were conducted aiming to answer questions, like "*Is the top-down ignition technique in general better compared to the bottom-up ignition technique as it is generally recommended by leaflets for correct heating?*"; "*What effect refers to the type of kindling material used for the ignition batch?*"; "*Are higher draught conditions generally correlated with higher gaseous and particulate emissions?*"; If not, *"What are the effects of higher draught conditions?"*

   Those questions are answered by the outcomes of the experiments presented in chapter 3.

3) **Analysis and evaluation of the applicability of two types of oxidizing honeycomb catalysts, i.e. catalytic conversion characteristics, safety aspects under critical operating conditions and long term durability, integrated in firewood roomheaters for emission reduction**

Integrated catalytic converters are frequently used in firewood room heating appliances in the United States and Canada [45] [66]. So far, European manufacturers focused on the primary optimization approach aiming at emission prevention by optimizing combustion conditions with constructive measures (e.g. air staging, well dimensioned and insulated combustion chambers, etc.). However, the effect of primary optimization is limited due to the high share of transient conditions and the wide influence of the user behavior in case of off-specification heating operation. Therefore, nowadays also European manufactures think about integrating catalytic converters into their appliances in order to reduce emissions and to comply with more stringent ELV. However, there were several open questions, like "*What is the effect of the integrated catalytic system on gaseous (CO, OGC) and PM emissions?*"; "*Are there correlations between space velocity and catalytic conversion rates for all types of emissions (CO, OGC, PM)?*"; "*How much differs catalytic conversion rates during characteristic phases of firewood combustion?*"; "*What are important development aspects for designing catalyst integrated solutions?*"; "*Which emission level can be achieved by integrating catalytic systems?*"; "*How much and what kind of maintenance is necessary to enable safe operating and optimal conversion conditions?*"; "*Are there synergetic effects of primary optimization and catalyst integration?*"

All those aspects and questions were analyzed by experimental evaluations which are presented in chapter 4, 5 and 6. Those chapters thematically synthesize on each other, but can be also read independently from each other.

4) **Analysis and evaluation of a real-life oriented test concept ("*beReal*") for firewood roomheaters compared to the existing EN standard regarding the differences of the procedure and repeatability**

As already mentioned in the introduction, testing of products shall guarantee minimum requirements, sufficient safety and reliability of the products. For firewood room heating appliances, official-type-tests (ott) according to the respective EN standards have to be passed before market introduction of new products. But, since their evaluation is far away from the heating by the end-customers, real-life emissions are higher and efficiency is lower compared to the ott results. Consequently, it is not surprising that nowadays biomass combustion systems are criticized as polluting and harmful technologies. To avoid similar discussions as currently ongoing in the car industry ("diesel gate") and to raise the reliability of biomass combustion systems in terms of low emissions and high efficiency new test concepts with high real-life relevance are requested [73].

Exemplarily for firewood roomheaters, an evaluation of such a real-life testing concept ("*beReal*") compared to the standard type test protocol (EN 13240) was conducted in this thesis. The main research questions were: "*What are the main differences between the procedures and data analysis concepts of both testing protocols?*"; "*Is it possible to achieve official-type-test results with a serial-production appliance?*"; "*What is the repeatability of the real-life test protocol compared to the standard type test protocol?*"; "*Is the repeatability of the real-life test concept significantly worse due to included transient conditions?*"; "*Is the real-life test protocol capable to reflect all possible situations in real-life operation?*"

All those questions are answered in chapter 7 of this thesis.

1.3 Impact of this work on scientific community

The outcomes of this thesis have been relevant for the scientific community in different R&D work. For example, the questions of the survey presented in chapter 2 were also applied for a European survey about user behavior [74]. The outcomes of both surveys were then used as a basis to define the procedure of the newly developed real-life testing procedure for firewood roomheaters, called *"beReal"* (chapter 7).

Also the results of chapter 3 about the effect of draught conditions and the ignition technique on emissions and thermal efficiency were considered for the development of the *"beReal"* test procedure. For example, the flue gas draught was not changed compared to the official-type-test protocol since the effect on emissions was found as appliance specific. Furthermore, no general recommendation for the top-down ignition technique within the *"beReal"* test protocol is given since it was clarified that for some appliances the bottom-up ignition technique might be more advantageous. Therefore, the ignition technique has to be determined appliance specific by the manufacturer within the Quick-User-Guide (QUG) of the *"beReal"* test protocol.

The outcomes of the combustion experiments with the integrated honeycomb catalysts (chapter 4, 5, and 6) clarified general aspects of its applicability and provide a basis for further R&D work (see "Outlook" in chapter 9). Using the DemoCat test facility it was possible for the first time, to analyze the catalytic effect of the catalyst during characteristic phases of firewood combustion, meaning start-up, intermediate and burn-out phase [75]. This clearly reveals the potential of synergetic effects of primary and secondary optimization by catalyst integration. Furthermore, for the first time, the correlation of space velocity and catalytic conversion rate in real flue gas from firewood combustion was assessed. This was possible since the DemoCat test facility enabled the simultaneous measurement in catalytically treated and untreated flue gas. Additionally, first data about the effect of off-specification and long term operation towards the applicability and the need of maintenance of integrated solutions were collected. The results confirmed many findings about catalytic conversion rates of previous publications, where similar or equal catalytic systems were used in retrofit applications. But the results provide also explanations on the question "*Why integrated systems are more efficient compared to retrofit applications*". The primary effect of the carrier material of the catalyst was analyzed and quantified for the first time within this thesis.

The comparison of test methods provided potential reasons and explanations why official-type-testing leads to test results which are far away from real-life situations. Further R&D work can use the descriptions of procedures and measurements of the new *"beReal"* test concept (chapter 7).

1.4 Impact of this work on industry

For manufacturers this work is valuable, since they learned many aspects about "*How their products are used in real-life operation*". Thus, they got information about typical mistakes and off-specification heating operation of end-customers in real-life operation which results in increased emissions and lower thermal efficiency. Based on that, they should focus their further technological development on real-life operating conditions and enhance measures to achieve the appliance specific best-practice heating operation in real-life operation.

The results about catalyst integrated solutions provide manufacturers information about potential emission values for CO, OGC and PM emissions which can be achieved in real-life heating operation. They got to know about the best way to develop catalyst integrated solutions – 1. Integrate a dummy, 2. Doing the primary optimization and defining the operational heating aspects (e.g. adjustment of air valve settings) and 3. Change the dummy against the catalyst. However, they also learned about the risk of blocking and consequently the need for cleaning the catalyst regularly.

The comparison of official-type-test protocol and the *"beReal"* test protocol illustrates that ott results do not represent real-life performance criteria. However, they can use the descriptions of procedures and measurements to implement the *"beReal"* test protocol in their own development work.

2 USER SURVEY – ASSESSMENT OF TYPICAL USER BEHAVIOR[2]

This chapter presents briefly the stock and market situation of residential biomass room heating appliances and the typical design of state-of-the-art appliances. Additionally, the current (Austria, Germany) and future (Europe) legal framework conditions regarding emissions and thermal efficiency are presented. Following the "state of the art", the methodology and the results of a user survey are presented. The user survey was conducted to evaluate the common user behavior for most relevant types of batch-wise operated room heating appliances in Austria. The user survey set the basis for the technological and non-technological measures evaluated and discussed in this thesis.

2.1 State of the art

2.1.1 Stock and market

In Austria, the total number of biomass room heating appliances was quantified by BENKE and LEUTGÖB [12] at around 1.5 million appliances.

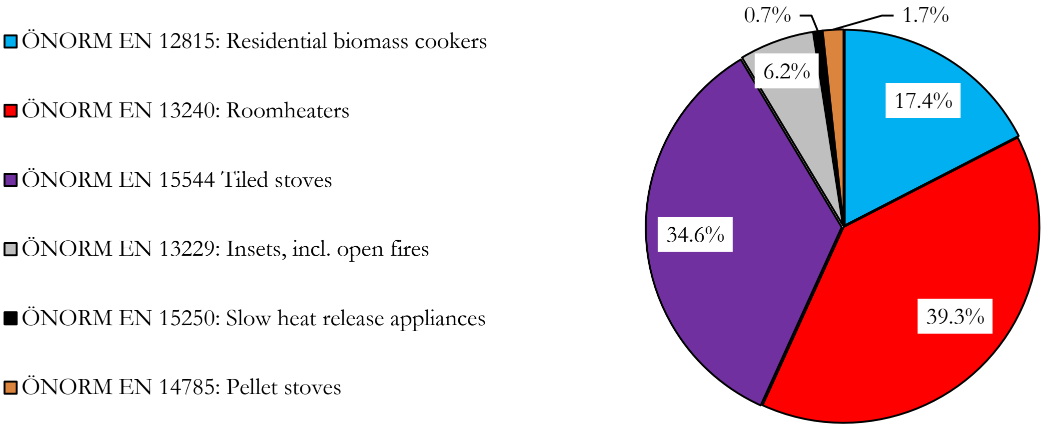

Figure 1: Share of residential room heating appliances in Austria according to figures and specifications presented in [12].

As illustrated in **Figure 1**, firewood roomheaters (~580,000 = 39.3%) and tiled stoves (~510,000 = 34.6%) are the most popular and widespread types of room heating appliances in Austria.

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[2] Segments of this section have already been published in [1].

Slow heat release appliances (~11,000 = 0.7%) and pellet stoves (~25,000 = 1.7%) have the lowest share of the stock of biomass room heating devices in Austria. The main difference of slow heat release appliances compared to tiled stoves is that tiled stoves are designed according to the chimney dimensions and are established directly on-site at the end-users, whereas slow heat release appliances represent prefabricated products which can be bought as ready-to-use products.

In Germany, the total stock of biomass room heating appliances is estimated in the range of 10.2 million [76] to 11.0 million appliances [77]. Detailed proportions referring to the different types of biomass room heating appliances are provided in [70] and illustrated in **Figure 2**.

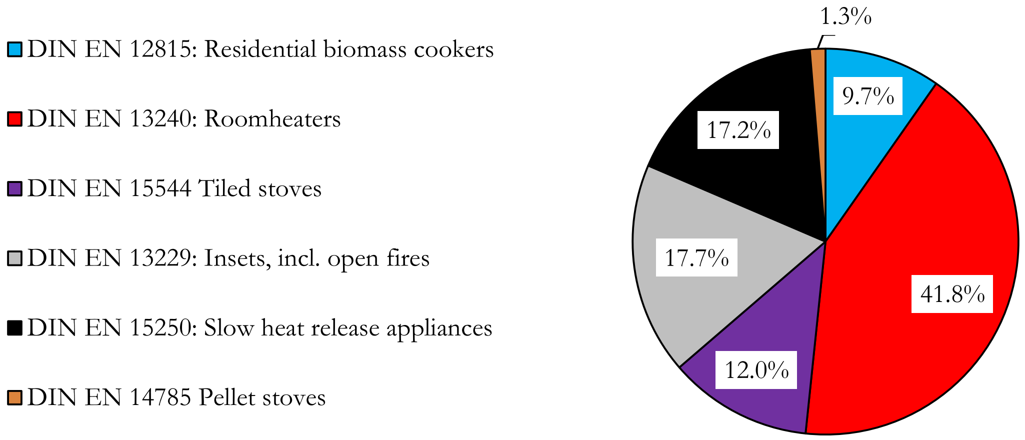


Figure 2: Share of residential room heating appliances in Germany according to figures and specifications presented in [77] and [70]

Based on the total number of [77] the share of firewood roomheaters is comparable to Austria with 41.8% (~4.60 million). Also the share of pellet stoves is comparable (~0.143 million =1.3%). The share of tiled stoves and residential biomass cookers is clearly lower compared to Austria at around 12% (~1.3 million) and 9.7% (~1.07 million). In contrast, insets (~1.95 million = 17.7%) and slow heat release appliances (~1.89 million = 17.2%) are more widespread in Germany compared to Austria.

In Europe, the total stock of biomass room heating appliances is estimated at about 65 million appliances. About 45 million appliances are supposed to be used as secondary heating source whereas the remaining appliances (~20 million) are regarded to be used as primary heating source [11].

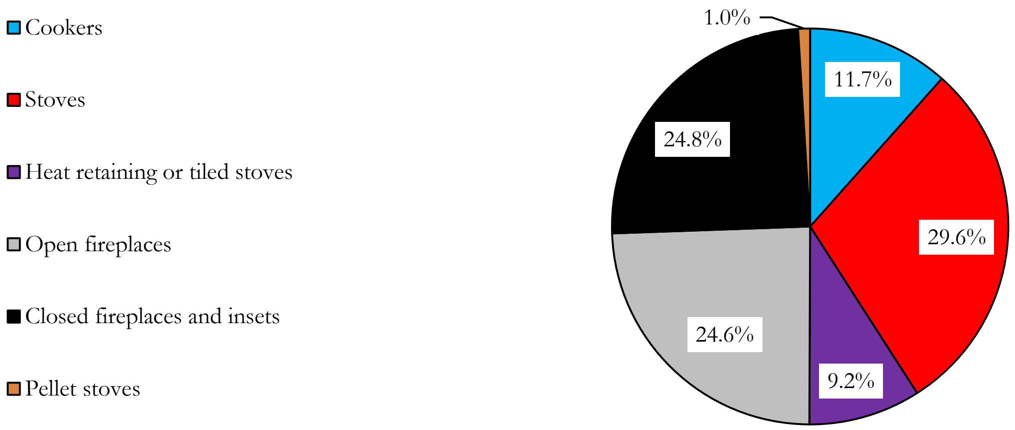


Figure 3: Share of residential room heating appliances in Europe according to figures and specifications presented in [11]

Compared to Austria and Germany the share of appliances looks differently in Europe (**Figure 3**). Most dominant types of appliance are open and closed fireplaces (including insets). In total they are quantified at around 32 million appliances (~50%) with a dominant distribution in France and Italy [11]. Stoves classified according to EN 13240 ("Stoves") represent 29.6% which means around 19.3 million devices. The share of heat retaining or tiled stoves is only 9% (= 6 million). However, it is mentioned that, for example in Finland and Sweden, the share of those technologies is very high (Finland: 100%, Sweden: 50%). On a European level the share of pellet stoves is lower compared to Austria and Germany at 1% (~637,000).

Table 1: Stock of residential room heating appliances in Europe, Austria and Germany in relation to the number of private households

	Stock of residential room heating appliances [Mio.] *	**Number of private households [Mio.]****	**Private households with room heating appliance**
Europe	65	221.3	29.4%
Austria	1.5	3.89	38.9%
Germany	11.0	41.3	26.6%

*…according to [11] (Europe), [12] (Austria), [77] (Germany)

**…according to https://de.statista.com (number of private households based on data for 2017)

As illustrated in **Table 1** around 29.4% of European private households have an installed residential room heating appliance. This share is slightly lower for Germany with a theoretical number of 11.0 million households equipped with a residential room heating device. In Austria, the share of private households is clearly higher compared to Europe and Germany at 38.9%. This illustrates the high popularity of those appliances for residential room heating in Austria. However, it has to be mentioned that the share of households with installed residential room heating appliances are most probably lower since there are households with more than one appliance installed. This was also confirmed by the user survey. During the survey 108 persons were interviewed, but totally 114 appliances were covered by the answers (chapter 2.3.1, **Table 4**). According to [11] the share of private households in Europe with installed residential room heating devices was estimated at 20%.

In Austria, the annual sales of biomass room heating appliances are around 28,100 appliances [78] (based on 2017) which represents around 1.9% of the stock. According to [78], firewood roomheaters represent the highest share of annual sold appliances with an absolute number of around 13,000 devices (= 46%), followed by tiled stoves (~8,100 devices), insets (~4,900 devices) and pellet stoves (~2,100) representing 29%, 17% and 7% of sold appliances.

In Germany, the annual sales are between 400,000 and 450,000 units [76] based on data between 2007 and 2014). This represents 3.6% to 4.5% of the stock and is clearly higher compared to Austria. The highest share can be attributed to firewood roomheaters with around 300,000 to 350,000 devices. This represents around 75% of sold appliances. The annual sales of inset appliances are around 50,000 units and around 25,000 units for each of both, solid fuel cookers and pellet stoves.

For Europe, the annual sales according to [11] are estimated at around 0.85 million for open fireplaces and 0.849 million for closed fireplaces and insets. For stoves the annual sales are quantified at around 1.037 million appliances, for pellet stoves at around 0.270 million and for cookers at around 0.464 million. Therefore, for all residential room heating appliances the total number of annual sales is around 3.47 million appliances which represent around 5.3% of the total stock. This is even higher compared to Germany. The product prices for biomass room heating appliances range from around 300 € (e.g. cheap firewood roomheater) to more than 10.000 € (e.g. tiled stove). However, the average product price for such heating devices was assumed between 2,000 € and 3000 € on a European level [11]. Therefore, under the assumption of an average product price of 2,500 € the total annual market volume is around 8.7 billion €.

2.1.2 Typical design of a state-of-the-art firewood stove

Firewood stoves, classified according to standard EN 13240 [13], are the most popular and widespread type of firewood room heating appliances in Austria and Germany. On a European level this type of solid fuel heating appliance represent around one third of the total stock and is the most frequently sold type of appliance (see previous section). Furthermore, the experimental work presented in this thesis refers to firewood roomheaters. Therefore, the design and most relevant technical characteristics of a state-of-the-art firewood roomheater are presented in **Figure 4**.

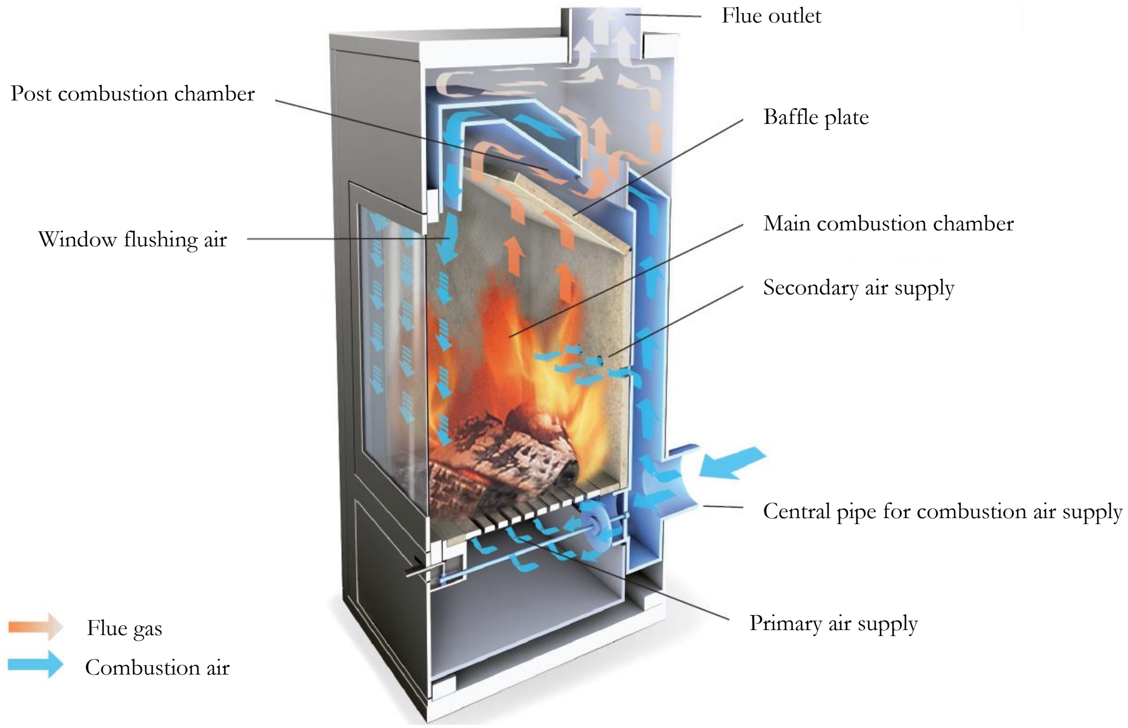


Figure 4: Technical design characteristics of a state-of-the-art firewood stove (EN 13240) [79] (translated in English)

State-of-the-art firewood roomheaters consist of a main combustion chamber where the firewood is manually fueled and a post combustion chamber where the heat is transferred to the surrounding envelope and /or to a specific designed heat storage material. The heat is released by radiation and convection into the room. Sometimes modern firewood roomheaters are also equipped with a fan in order to transfer a proportion of the thermal heat to another room. The combustion chamber and the post combustion chamber are separated by a baffle plate which enhances the turbulences of flue gases when they enter the post combustion chamber.

The total combustion air supply of modern firewood stoves is conveyed by a central pipe. This is important, especially for modern buildings where the combustion air is not supplied from the installation room but from outside air. In most cases the combustion air supply of such technologies is provided by a specifically designed chimney system (chapter 3.1.2, **Figure 11**). Modern state-of-the-art firewood stoves are featured with an air staging concept [52] [53]. Therefore, the total amount of combustion air supply is divided in primary, secondary and tertiary air (window flushing air). The primary air is predominately used for processes of pyrolysis and gasification whereas the secondary air is used for oxidation of gaseous intermediate products. The window flushing air, often defined as tertiary air, is usually contributing to the secondary air supply. However, the main purpose of window flushing air is to keep the window free from particulate agglomerations (e.g. tar and soot).

The technical design of other types of modern firewood room heating appliances, e.g. residential biomass cookers, tiled stoves and fireplaces, is in general very similar. Therefore, the principals of the design – combustion chamber, post combustion chamber and air staging concept – are also implemented in those products. The main differences regarding the technical design are the amount of heat storage capacity and the thermal heat management.

2.1.3 Legal framework conditions

The European standards define minimum requirements regarding emissions and thermal efficiency. For instance, firewood roomheaters (EN 13240) have to achieve CO emissions < 1 vol.-% (based on 13 vol.-% O_2) and a minimum thermal efficiency of 50% [13]. However, in many European countries the requirements regarding emissions are much more stringent and covering additional components, like, PM, OGC and NO_x emissions. Therefore, manufacturers have to comply with the national emission limit values (ELV) when they want to introduce new products into the market. As already mentioned in the introduction (chapter 1.1) the new Ecodesign requirements will set a common basis of ELV in whole Europe from the beginning of 2022 [37].

Table 2 provides and overview of the current national ELV of Austria and Germany in comparison with future Ecodesign requirements coming into force in 2022 [37]. Additional ELV of other European countries (e.g. Switzerland, Sweden) can be found in [11] and [76].

For Austria the emission requirements refer to the energy input based on the net calorific value of fired fuel (mg/MJ). Based on the fuel composition and the net calorific value of the fired test fuel the data can be transformed to mg/m³ (STP, dry, 13 vol.-% O_2). For a rough estimation those figures can be multiplied by a factor of 1.538 to achieve the ELV in mg/m³ (STP, dry, 13 vol.-% O_2) [80].

In general appliances have to comply with the respective requirements during official-type-testing at nominal load according to the specific EN standards [13]-[17]. Official-type-testing is carried out by notified testing bodies using the final prototype of the newly developed appliance which is provided by the manufacturer. Only for Austria all listed types of appliances of **Table 2** have to be evaluated obligatorily also during part load operation (e.g. 50% of nominal load for firewood roomheaters). However, during part load the appliances only have to comply with the ELV of CO and OGC emissions.

Table 2: Overview of current national (Austria, Germany) and future (Europe) emission limit values (ELV) for residential biomass room heating systems which have to be achieved during official-type-testing (ott)

Regulation	**Type of Technology**	**CO**	**OGC**	**NOx**	**PM**	**Efficiency**
Art 15a B-VG (Austria) [81]	Roomheaters	1,100 mg/MJ	50 mg/MJ	150 mg/MJ	35 mg/MJ	80%
	Insets	1,100 mg/MJ	50 mg/MJ	150 mg/MJ	35 mg/MJ	80%
	Slow heat release appliances and tiled stoves	1,100 mg/MJ	50 mg/MJ	150 mg/MJ	35 mg/MJ	80%
	Cookers	1,100 mg/MJ	50 mg/MJ	150 mg/MJ	35 mg/MJ	72%
	Pellet stoves	500 mg/MJ	30 mg/MJ	100 mg/MJ	25 mg/MJ	80%
1 BImSchV (Germany) [82]	Roomheaters	1250 mg/m³	-	-	40 mg/m³	73%
	Insets	1250 mg/m³	-	-	40 mg/m³	75%
	Slow heat release appliances	1250 mg/m³	-	-	40 mg/m³	75%
	Cookers	1500 mg/m³	-	-	40 mg/m³	70%
	Pellet stoves (without water jacket)	250 mg/m³	-	-	30 mg/m³	85%
Ecodesign requirements (Europe, coming into force in 2022) [37]	Solid fuel local space heater (firewood, closed fronted)	1500 mg/m³	120 mg/m³	200 mg/m³	40 mg/m³	65%*
	Solid fuel local space heater (firewood, open fronted)	2000 mg/m³	120 mg/m³	200 mg/m³	50 mg/m³	65%*
	Solid fuel local space heater (pellets, closed fronted)	300 mg/m³	60 mg/m³	200 mg/m³	20 mg/m³	79%*

All emission concentrations (mg/m³) refer to STP conditions (273.15 K/101,325 Pa), dry flue gas and 13 vol.-% O_2

* Seasonal space heating energy efficiency (specifically calculated based on ott results)

Official-type-testing is carried out under well-defined operating conditions (e.g. using specifically designed wood pieces) without respecting transient conditions, like start and stop phases. Therefore, operating conditions which refer to typical user behavior (e.g. ignition, different loads, and typically designed firewood) are not respected during official-type-testing. Concluding, the ELV of **Table 2** represent best possible values which are not representative for real-life operation performance at the end-users [49] [70]. The official-type-testing results are not reachable with serial-production appliances when they are tested according to the respective EN standard (chapter 7) [133]. In detail, this topic is addressed and discussed in chapter 7.

2.2 Approach

As already mentioned in the introduction (chapter 1), the need for advanced testing procedures for official-type-testing that better reflect real-life user behavior has reached the awareness of the European public administration [73]. The user survey, which is described in the following, evaluates "*How firewood room heating appliances are typically operated in real-life by end-users*". Therefore, users of firewood roomheaters (EN 13240), residential biomass cookers (EN 12815) and tiled stoves (EN 15544) were asked about their common mode of heating operation. Together with results of the user survey of SCHIEDER et al. [83] similarities and differences of user behavior of different technology classes were analyzed. Based on that, the typical user behavior for all three technology classes was defined. Thereby, a clear picture about typical user behavior in Austria could be drawn. Shortly, after publishing this study, the study of WÖHLER et al. [74] was published providing results of an online user survey regarding user behavior of firewood room heating appliances in Europe. The Austrian survey (chapter 2) was the basis for defining the questions of this European survey [74].

All those information together represent a useful basis for development and implementation of technological and non-technological primary measures for improving user behavior regarding minimized emissions and optimized efficiency in real-life operation. Furthermore, the findings were used to develop and define the procedure of the real-life oriented test protocol *"beReal"* (see chapter 7).

2.3 Material and methods

The user survey based on a questionnaire and was conducted at the tradeshow "Buildings and Energy" in Lower Austria in September 2012. The survey was performed by face-to-face interviews with attendees of the tradeshow who are users of firewood room heating appliances. In total 108 randomly selected persons met this requirement and agreed to take part in the survey. No personal data, like name, age, education level or gender, were recorded. Further, no questions regarding specific living conditions were asked. In advance of the interviews all respondent users were informed that no personal data will be published and that all data analysis will be done anonymously. This was done in order to fulfill data privacy protection requirements.

The questionnaire consisted of 15 multiple-choice questions. All questions as well as the specific answer options were defined by representatives of certified testing bodies, associations for firewood room heating appliances, air quality authority and research experts in the field of small-scale biomass combustion technology.

2.3.1 Types of room heating appliances covered by the survey

The questionnaire defined three groups of different types of firewood room heating appliances.

- Firewood roomheaters (EN 13240:2007 [13])
- Tiled stoves (ÖNORM B 8303:1999 [84] or EN 15544:2009 [17])
- Residential biomass cookers (EN 12815:2007 [15])

According to BENKE and LUTGÖB [12], these types of appliances have a market share of nearly 92% of biomass room heating appliance in Austria (**Figure 1**). These types of firewood room heating appliances are commonly used for batch-wise operation. Additionally, for all three types of appliances the operation takes place under natural draught conditions. The main difference between the three technologies is that firewood roomheaters as well as residential biomass cookers are supplied as industrial end-user products or as pre-fabricated construction sets, whereas tiled stoves are constructed directly on site at the end-customer using pre-fabricated tiles or fireclay stones.

2.3.2 Structure and content of the questionnaire

The questionnaire was structured in four parts – part 1 for basic information, part 2 about used fuels, part 3 about real-life operating conditions and part 4 about the mode of heating operation, the so called “user behavior”.

Part 1 – Basis information

In the first part of the questionnaire the general information is conveyed. The purpose of the first question was the classification of respondent's user behavior to a certain group of firewood room heating appliances. Further basis information about the general operation according to manual as well as the availability of any external training arrangements before initial operation was queried.

Part 2 – Fuels

Two questions about the commonly used fuels were asked in order to assess relevant fuel properties in real-life operation.

Part 3 – Operating conditions

Two questions about natural draught conditions and the adjustment of the chimney system to the respective type of appliance were asked. Since the user is not able to determine draught values quantitatively, only qualitative options were given in order to get an impression about the draught conditions in real-life operation.

Part 4 – Mode of operation (user behavior)

The main part of the questionnaire aimed at the identification of the user behavior in real-life operation. Therefore, eight questions were asked within this part. Three questions aimed at the investigation of the ignition procedure. In particular the mode and form of fuel placement of the ignition batch, the use of different kinds of starting aids as well as the common position for lighting the ignition batch were queried supported by explanatory photographs (**Table 3**).

Table 3: Overview of selectable options for answering the question regarding the mode of placement of the ignition batch in the combustion chamber

Cross joint, on top shavings	Firewood, at the bottom shavings	Cross joint, without shavings	Campfire
			

Further, four questions were asked to enable an estimation of the average duration of one heating operation cycle as well as to identify the common user behavior after starting the heating operation of the appliances. Therefore, the questions tended to identify the instant of time for recharging a new fuel batch, the number of firewood pieces used per fuel batch, the air valve settings during heating operation and the absolute number of performed batches of one heating operation cycle. The final question was about the air valve settings after finishing the heating operation cycle. This question was important to enable a conclusion regarding potential thermal heat losses during cooling down phase.

2.3.3 Data analysis and data evaluation

Data analysis was performed for each specific type of technology. For all multiple-choice questions the frequency of answers for each parameter was calculated and analyzed separately for each type of technology. For defining the typical user behavior the required frequency of answers about the respective operation parameter was defined at least 60%. For enhancing the informative value the results of the survey were compared and validated with available literature data of the study of SCHIEDER et al. [83] about user behavior in Austria. In the frame of this study 652 users of biomass room heating appliances were asked about technical data of their appliances, the number of heating operation days per year as well as about the amount of used fuel. 139 users of biomass room heating appliances were also asked in detail about their user behavior during heating operation. In the study of SCHIEDER et al. [83]the survey was performed as direct interviews personally at several different events or via telephone. However, the survey described in this chapter covered several more additional operational aspects that are essential for real-life emission as well as efficiency performance compared to the study of SCHIEDER et al. [83]. Therefore, in this survey more detailed questions about the mode of storage of the firewood, draught conditions, and adjustment of air supply settings during and after heating operation as well as more specifications about each single fuel batch were asked.

2.4 Results and discussion

2.4.1 Survey

During the survey 108 persons were interviewed. The sum of firewood room heating appliances comprised by the survey was 114 heating devices, which were almost equally distributed to the defined classes of technology (**Table 4**).

Table 4: Results about absolute numbers as well as the percentage share of technologies covered by the survey

Number	Question	Type of Technology	Absolute value = n	Percentage value
1.1	What kind of technology do you have?	Firewood roomheater	43	38%
		Tiled stove	34	30%
		Residential biomass cooker	37	32%
		Total values	**114**	**100%**

n = number of answers

According to the Austrian market analysis of BENKE and LEUTGÖB [12], the percentage of firewood roomheaters is around 39% (= 580,000) of the stock of small scale biomass room heating appliances in 2009 (= 1,473,700). The share of tiled stoves (also including fireplace inserts) is around 41% (= 601,000). However, the percentage share of residential biomass cookers is only around 17% (= 257,700) (**Figure 1**). Hence, it is obvious that the segmentation of appliances of the survey is not representative for the Austrian market share of tiled stoves and residential biomass cookers. In this survey the share of tiled stoves was underrepresented, whereas the share of residential biomass cookers was disproportionately high. Therefore, the results of each type of technology were analysed separately in order to respect each type of technology equally.

Part 1 – Basis information

Table 5 represents the results of answers about relevant basis information regarding the respective type of appliance.

Table 5: Results of the survey about basis information classified to each type of technology

	Number	**Question**	**Options for answer**	**Firewood roomheaters**		**Tiled stoves**		**Residential biomass cookers**	
				n [-]	[%]	n [-]	[%]	n [-]	[%]
Basis Information	1.2	Do you operate your stove according to manual?	Yes	12	28%	12	35%	10	27%
			No	16	37%	13	38%	12	32%
			To some extend	9	21%	5	15%	5	14%
			I don´t know	6	14%	4	12%	10	27%
	1.3	Were there any external training arrangements before initial operation? Results given in **Figure 5**							

n = number of answers

The results of basis information show, that only about one third of the interviewed persons operate their heating appliances knowingly according to the manufacturers' manual. The same amount of about one third of respondent users declared that they knowingly do not operate the appliance in accordance to manufacturers manual. Further, no significant difference was found between the different types of technologies regarding this aspect. Hence, these results indicate a tendency that users of firewood room heating appliances either spend insufficient attention to, or simply ignore the operation mode recommended by manufacturer's manual. For external training arrangements before initial operation significant differences between the different types of technology were found (**Figure 5**).

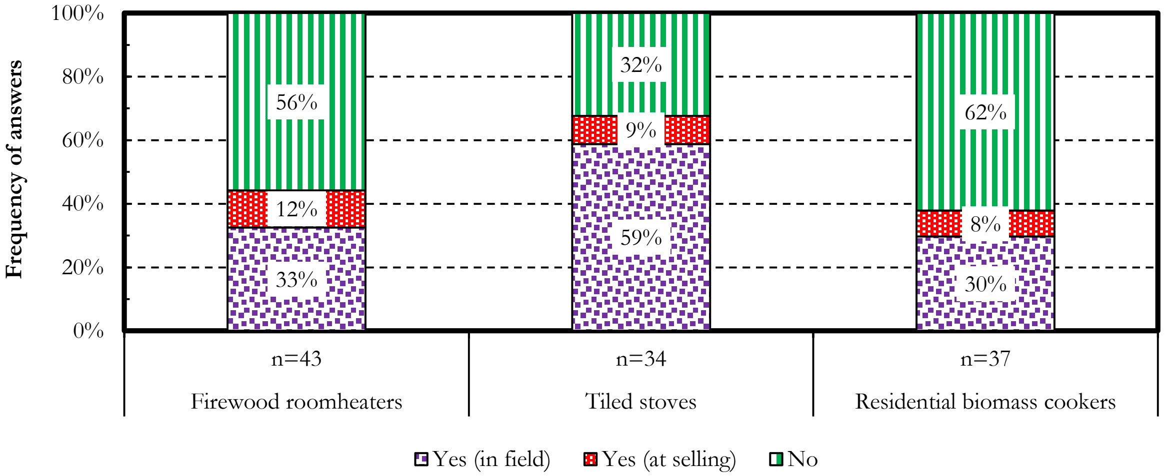


Figure 5: Results of question 1.3 about the availability of any external training arrangements before initial operation

About one third of users of firewood roomheater and residential biomass cookers got an external training in the field before initial operation. But in contrast, nearly 60%, of tiled stove users got an external training in field before initial operation. However, it is interesting that there are only less training arrangements regarding correct operation of the appliances at selling (only 8% – 12%), especially for firewood roomheaters and residential biomass cookers. For tiled stoves this outcome is explainable since tiled stoves are commonly sold, designed and set up by professional stove fitters directly at the home of the user. Therefore, potential trainings are arranged directly at the user before or during the official transfer of the appliance to the user. These results correspond also to the findings of SCHIEDER et al. [83]. According to their findings, only 8% of respondent users of biomass room heating appliances got a professional training at selling. Finally, the results of the basic information section of the survey illustrate that there might be a lack of information, especially for users of firewood roomheaters and residential biomass cookers, how to operate the respective appliance correctly.

It is claimed that there is a considerable potential for improving the emission and efficiency performance of real-life heating operation by effective measures that guarantees that users of firewood room heating appliances get all necessary information for correct operation before initial operation. Beside external training arrangements in the field as well as at selling a further possibility could be an obligatory Quick-User-Guide comprising the most important operation characteristics on only one page. For example, this page could be fixed to the window of the combustion chamber door and therefore the user is forced to take a look on this information at least once before initial operation.

Part 2 – Fuels

Table 6 summarizes the results of questions about used fuels in real-life operation.

The results about the kind of used fuel are very similar for all three types of technology. The mainly used kind of fuel is firewood of hardwood with a length of 0.25 m or 0.33 m. The utilization of softwood firewood or wooden briquettes is of comparably lower importance. However, the results regarding commonly used length of firewood confirm the trend that in tiled stoves in principle longer pieces of firewood are used. These results can be explained by the usually higher amount of fuel per batch which is linked with a higher combustion chamber volume of tiled stoves compared to firewood roomheaters and residential biomass cookers.

Table 6: Results of the survey about questions related to fuel characteristics

	Number	Question	Options for answer	Firewood roomheaters		Tiled stoves		Residential firewood cookers	
				n [-]	[%]	n [-]	[%]	n [-]	[%]
Fuels	2.1	What kind of fuel do you use usually? Results given in **Figure 6**							
	2.2	What is the length of the firewood?	0.25 m	22	53%	5	15%	20	54%
			0.33 m	17	40%	24	73%	15	41%
			0.50 m	3	7%	4	12%	2	5%
			> 0.50 m	0	0%	0	0%	0	0%
	2.3	What is the storage mode of the firewood until it is used?	Outside uncovered	2	4%	3	8%	1	3%
			Outside covered	18	40%	11	31%	15	38%
			Indoor	13	29%	8	22%	13	33%
			Others (e.g. garage...)	12	27%	14	39%	11	28%
	2.4	What is the average storage duration of firewood (from felling till thermal utilization)?	<6 months	7	17%	3	9%	2	5%
			6-12 months	13	32%	8	24%	7	19%
			12-18 months	13	32%	15	44%	17	46%
			>18 months	8	20%	8	24%	11	30%

n = number of answers

Figure 6 presents the frequencies of answers of question 2.1 about the kind of usually used fuel in the respective type of firewood room heating appliance.

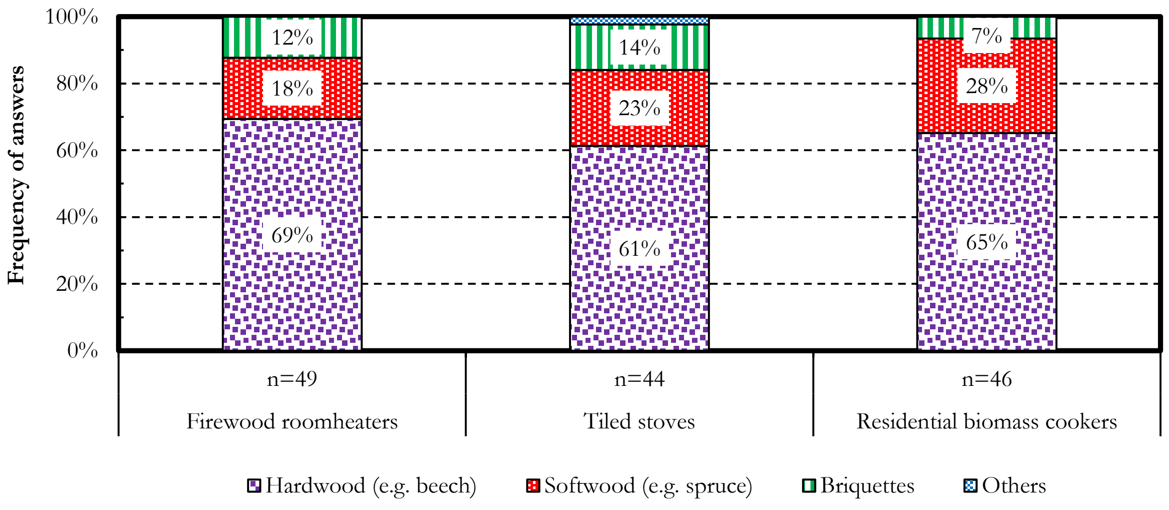


Figure 6: Results of question 2.1 about the kind of usually used fuel

Regarding the mode and duration of firewood storage the results indicate that the common use of firewood storage is either covered outside or at any place indoor for at least 6 months. The amount of respondent users that declared that their firewood is stored less than 6 months was below 10% for users of tiled stoves and residential cookers and below 20% for users of roomheaters. However, it might be possible that these users usually buy the firewood as air-dried firewood that is usable without any longer storage duration. Corresponding to the results of HARTMANN et al. [48], this leads to the assumption that the moisture content of the used firewood in real-life operation is usually between 12% and 20%. They found out that covered and split firewood dry to that range within 4 – 6 months of outdoor storage. This is also confirmed by findings of previous field measurements of moisture content in real-life done by SPITZER et al. [85]. According to their measurements of 29 fuel samples, the average moisture content of firewood in real-life operation was 12% in Austria. The range of measured moisture contents was between 8% and 21%.

In this survey no specific question about the utilization of litter in firewood room heating appliances was asked. The use of other fuels than firewood or briquettes was only declared by one tiled stove user. However, the findings of SCHIEDER et al. [83] indicate a considerable role of litter that is used in firewood room heating appliances. Corresponding to their findings based upon 139 surveys, in 31.9% of all firewood room heating appliances litter is used as fuel to some extent. The results of their study about the share of used litter for combustion showed that untreated waste wood (29%), used paper (23.2%), cardboard boxes (15.2%), packaging material and leftovers (4.3%) are most frequently used types of litter in firewood room heating appliances. But anyway, it has to be considered that the estimated number of unreported cases is not known and also difficult to estimate. According to the opinion of experts and manufacturers, the share of litter that is used as inappropriate fuel in firewood room heating appliances is estimated to be up to 30% in Austria [86]. This assumption is also in line with the findings of the study of SCHIEDER et al. [83].

Part 3 – Operating conditions

Table 7 represents the results of answers about a qualitative estimation of operating conditions, respectively draught conditions, dedicated for each type of appliance.

Draught conditions are essential operation conditions in respect of efficiency performance and consequently also indirectly for emission performance. Poor draught conditions could lead to operational problems and poor combustion conditions. Extremely high draught conditions, i.e. more than 30 Pa, result in a decrease of efficiency and in intensive combustion conditions. This could also increase the emission level as well as the thermal heat output [49] [51] (see also chapter 3).

Table 7: Results of the survey about real-life operating conditions allocated to each type of technology

	Number	Question	Options for answer	Firewood roomheaters		Tiled stoves		Residential biomass cookers	
				n [-]	[%]	n [-]	[%]	n [-]	[%]
Operating conditions	3.1	How are the operating conditions of your heating system regarding draught level?	Poor draught conditions	4	9%	2	6%	6	16%
			Optimal draught conditions	38	88%	30	91%	30	81%
			Exorbitant draught conditions	1	2%	1	3%	1	3%
	3.2	Is the draught level of the chimney system adjusted to the stove?	Yes	9	21%	14	42%	13	36%
			No	12	28%	4	12%	3	8%
			I don´t know	22	51%	15	45%	20	56%

n = number of answers

Draught conditions are in most cases estimated as "optimal" by the interviewed users. Poor draught conditions are denoted by more users than too high draught conditions. However, it has to be mentioned that the estimations of the responded users are only based on their feelings about draught conditions and not on quantified data or calculations. This is also confirmed by the answers of question 3.2 which shows a lack of information about the adjustment of the respective appliance to the chimney system. The results indicate that possibly residential biomass cookers could tend to operating problems due to poor draught conditions.

However, based on these qualitative results it generally can be assumed that draught conditions are mostly sufficient regarding usual operation as well as safety aspects in real-life operation. However, it is not possible to conclude to the common absolute draught level in the field as well as to the resulting emission and efficiency impact by the findings of the survey. The question regarding the adjustment of draught conditions to the respective chimney system shows that nearly 50% of users don't know that. But it seems that especially firewood roomheaters are at least adjusted to the chimney system, whereas nearly 40% of users of tiled stoves and residential biomass cookers declared that their appliances were adjusted to the respective chimney systems in terms of draught conditions. Generally, it seems that there is less awareness of the user for the importance of the optimal adjustment of chimney system and heating appliance to enable adequate draught conditions for low emission and high efficiency performance.

Part 4 – Mode of operation (user behavior)

Table 8 presents the results of answers about the mode of operation.

Table 8: Results about the mode of operation (user behavior) allocated to each type of technology

	Number	**Question**	**Options for answer**	**Firewood roomheaters**		**Tiled stoves**		**Residential biomass cookers**	
				n [-]	[%]	n [-]	[%]	n [-]	[%]
Mode of operation	4.1	How often is a new fuel batch recharged?	Only ignition batch	3	7%	8	24%	1	3%
			One time of recharging	11	26%	6	18%	5	14%
			Two times of recharging	9	21%	8	24%	8	22%
			Three times of recharging	6	14%	3	9%	9	24%
			> Three time recharging	14	33%	9	26%	14	38%
	4.2	How is the ignition batch placed in the combustion chamber?	Cross joint, on top shavings	5	12%	4	12%	0	0%
			Firewood, at the bottom shavings	21	51%	14	42%	29	78%
			Cross joint, without shavings	5	12%	7	21%	5	14%
			Campfire	10	24%	8	24%	3	8%
	4.3	What kind of starting aids do you use for ignition?	Small firewood pieces, specific starting aids	8	19%	8	22%	1	3%
			Small firewood pieces, paper	22	51%	18	50%	33	87%
			Only specific starting aids	7	16%	2	6%	0	0%
			Only paper	4	9%	5	14%	3	8%
			Others (i.e. straw...)	2	5%	3	8%	1	3%
	4.4	Where do you light the ignition batch? Results given in **Figure 7**							
	4.5	Do you rule the combustion air of each batch by adjusting the air valve settings?	Yes, once per batch	22	51%	11	32%	15	41%
			Yes, several changes per batch	7	16%	7	21%	9	24%
			No, steady settings	14	33%	14	41%	13	35%
			Others (i.e. at need, …)	0	0%	2	6%	0	0%
	4.6	When do you recharge a new fuel batch?	Only little flames visible	19	45%	13	46%	20	54%
			After a certain time interval	4	10%	4	14%	3	8%
			No flames visible	17	40%	11	39%	13	35%
			When signalized by the stove	2	5%	0	0%	1	3%
	4.7	How many firewood pieces do you recharge per batch usually? Results given in **Figure 8**							
	4.8	What are the air valve settings AFTER finishing the heating operation?	Opened	5	12%	3	10%	2	5%
			Closed	21	49%	19	63%	16	43%
			Not changed	17	40%	8	27%	19	51%

n = number of answers

Question 4.1 aimed at the identification of the average number of rechargings that is done within one heating operation cycle (from "cold to cold stage"). Results show no clear tendency towards a specific number of recharging a new fuel batch. However, the single use of the ignition batch as a whole heating operation cycle is only relevant for tiled stoves (24%). Residential biomass cookers tend to be operated most frequently with more than four batches (52%). For firewood roomheaters the distribution is wider, but it seems that the use of at least three batches for one heating operation cycle is a common operation mode for firewood room heating appliances. According to the findings of SCHIEDER et al. [83], the duration of a heating operation cycle is in the range of 3 – 6 h for these types of firewood room heating appliances. This corresponds to the results about number of rechargings of the survey when it is assumed that a batch in real-life operation lasts for around 0.75 – 1.5 h.

The results about the ignition procedure (Question 4.2 – 4.4) indicate no general difference between users of all three types of firewood room heating appliances. The placement of the ignition batch is commonly done by placing firewood pieces above small shavings at the bottom of the combustion chamber. This preferred mode of placement of the ignition batch is followed by the traditional campfire placement, especially for users of firewood roomheaters and tiled stoves. The most commonly used starting aids are the combination of small firewood pieces and paper. Especially the ignition of first fuel batch of residential biomass cookers is performed predominantly in this way. Less than one quarter of respondent users of firewood roomheaters and tiled stoves light the ignition batch with small firewood pieces and specific starting aids. Further, a significant trend regarding the lighting position of the ignition batch was found (**Figure 7**).

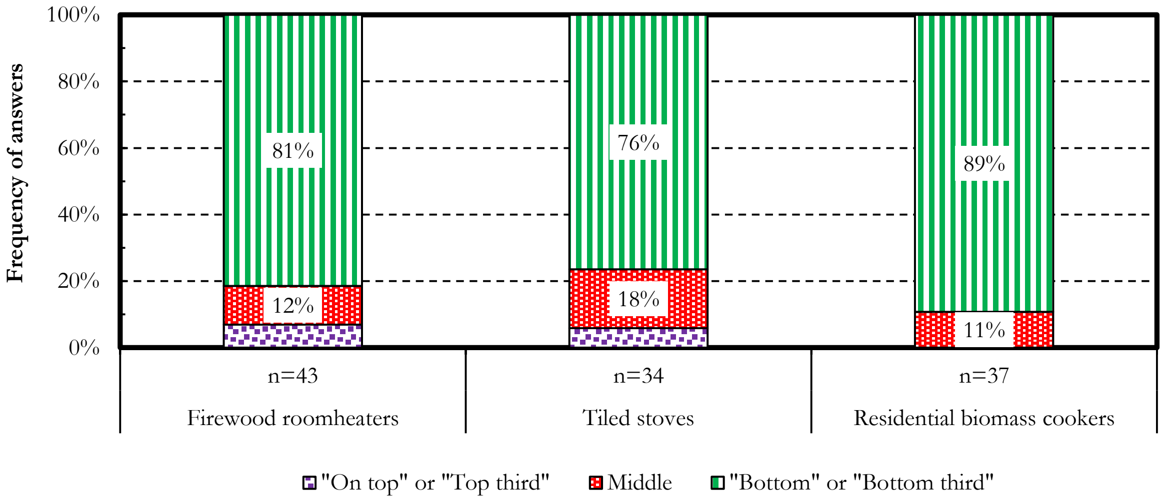


Figure 7: Results of question 4.4 about the position where the first fuel batch is usually lighted

As illustrated in **Figure 7** most of the users of all three classes of technology light the fire at the bottom or at the bottom third of the ignition batch (76% – 89%). This was also found out when comparing only the answers of respondents, who declared to operate their appliances according to manufacturer's manual. Previous research has shown that this kind of lighting procedure leads to high emissions in real-life operation. Corresponding to HARTMANN et al. [48] and NUSSBAUMER et al. [47] the procedure of lighting the ignition batch of a firewood room heating appliance has a strong impact on the emission level. Within both studies it was found out that higher emissions are caused by the traditional procedure of lighting the ignition batch from the bottom (bottom-up method). In contrast, using the method of lighting the ignition batch from the top (top-down method) resulted in a particulate emission reduction of 50% – 80%. Furthermore, CO emissions were significantly reduced by around 70% by applying the top-down ignition method. Consequently, the results of the survey emphasize the assumption that there is a significant emissions reduction potential in real-life operation of firewood room heating appliances by improving the lighting procedure. Only 4% of all respondents declared to apply the top-down ignition mode by lighting the ignition batch at the top or top third. This is even less compared to the findings of SCHIEDER et al. [83] that showed that 10.2% of Austrian users declared to apply the top-down ignition method. Consequently, the results about the ignition procedure indicate the need to improve the user behavior referring to the ignition procedure in real-life operation by effective measures. Therefore, a systematic evaluation of the top-down and bottom-up ignition technique was conducted at two different firewood roomheaters in order to increase the number of tests and to evaluate if the top-down ignition is generally advantageous compared to the bottom-up ignition technique (see chapter 2).

The mode of regulating the air valve settings for combustion air supply during one fuel batch were identified by question 4.5. The results indicate mainly two common modes of air valve adjustment for operation of a firewood room heating appliance. One part of users negates changing air valve settings for combustion air supply. This is the most frequent operation mode for users of tiled stoves (41%). But there is another part of users who changes air valve settings at least once a batch. This mode of air valve adjustment is predominantly seen for users of firewood roomheaters and residential cookers (51%, 41%). However, according to the data of the survey there is no possibility to assess at which stage of combustion process the air valve adjustments are done. Therefore, it is not possible to evaluate if the air valve adjustments for the combustion process for low emission and high efficiency performance. According to several studies, insufficient combustion air supply leads to an increase of high gaseous as well as particulate emissions [21] [41]. It is claimed that stoves equipped with implemented automatically controlled air supply devices could significantly contribute to support correct air valve settings during heating operation. In addition, this would also increase the user comfort.

The instant of time for recharging a new fuel batch, investigated by Question 4.6, indicates a clear tendency for all classes of technology. The most frequent answer is when “Only little flames visible” (45% – 54%) followed by the point “No flames visible” (35% – 40%). Only less than 15% of respondents recharge a new fuel batch “After a specific time interval”. SCHIEDER et al. [83] asked a similar question in the scope of their survey. However, they predetermined different time intervals as answer options. They found out that 18.9% of users recharge a new fuel batch after 0.5 h and 25.4% after 50 min. According to HARTMANN et al. [48], a new fuel batch should be recharged shortly before the flames extinguish in order to guarantee sufficient temperatures for a fast ignition of the new fuel batch and subsequently for lower emissions. Consequently, it has to be mentioned that according to the findings of the survey this is commonly done in real-life operation. Therefore, this aspect of user behavior should not be responsible for increased emissions in real-life operation.

Since the fuel amount of a batch significantly influences the emission level [38] [40] [87] the question about the number of firewood pieces used for recharging a new fuel batch (Question 4.7) was asked (**Figure 8**).

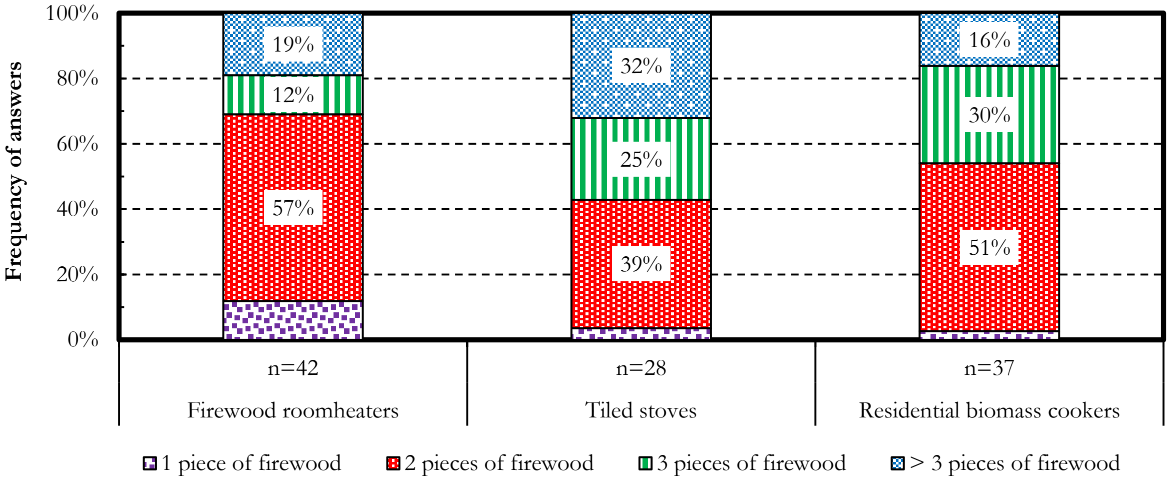


Figure 8: Results of question 4.7 about the number of firewood pieces commonly used for recharging a new fuel batch

Figure 8 illustrates that respondent users of all three classes of technology predominantly recharge a new fuel batch using two pieces of firewood. According to these results a general trend for a real-life operation in overload causing high emissions can be excluded. Users of tiled stoves tend to use generally more pieces of firewood for recharging compared to users of firewood roomheaters and residential biomass cookers. This can be explained by the specific technological aspects and the commonly higher nominal heat output of tiled stoves. However, there seems to be a tendency of overloading for nearly 20% of firewood roomheaters in real-life operation by using more than three pieces of firewood for one batch with an average mass of one firewood piece of around 0.75 – 1.0 kg.

The final question (Question 4.8) of the survey was linked to air valve adjustment when finishing the heating operation. Closing air valve settings after finishing heating operation is essential for achieving less thermal losses during the cooling down phase. Additionally, this enables a maximum possible efficiency factor in real-life operation. The most frequent air valve adjustment after finishing the heating operation of firewood roomheaters and tiled stoves are closed settings. In contrast, for residential biomass cookers the most frequent way is to do no specific air valve adjustments when finishing the heating operation cycle. This is also declared by 40% of users of firewood roomheaters as well as by 27% of users of tiled stoves. Around 5% – 12% of all users declared to complete the heating operation cycle with open air valves. Whereas the air valve settings of tiled stoves are often temperature controlled and automatically closed the air valve settings of firewood roomheaters and residential cookers are most frequently only manually controlled. Consequently, it can be assumed that the air valve settings of more than 50% of all firewood roomheaters and residential biomass cookers remain open, at least partly, after finishing the heating operation cycle. This leads to permanent thermal heat losses, especially during cooling down phase but also during times without any operation. Finally, this results in a decrease of the efficiency factor of the respective firewood room heating appliance. Therefore, it is essential to develop and implement effective measures that guarantee the closing of air valve settings after finishing the heating operation.

2.4.2 Assessment of typical user behavior

The results of this survey in addition to the survey of SCHIEDER et al. [83] show generally a very similar user behavior for all three types of appliances. Therefore, below the typical user behavior during real-life operation is described for users of firewood roomheaters, tiled stoves and residential biomass cookers in Austria.

For operation of firewood room heating appliances predominantly hardwood with a length of 0.25 m or 0.33 m is used. Until thermal utilization the firewood is stored either covered outside or indoor for duration of mainly 6 – 18 months. The placement of the ignition batch is either done with firewood pieces placed on shavings at the bottom of the combustion chamber or by using the traditional campfire placement. Small firewood pieces together with paper or specific starting aids are commonly used as starting aids. The ignition batch is lighted applying the bottom-up ignition method. Recharging a new fuel batch is done at least once per heating operation cycle using two or three pieces of firewood. This is done when only little or no flames are visible. The air valve settings during a heating operation cycle are either adjusted to steady settings over the whole batch or adjusted once per batch. After the last batch air valve settings are either closed or are not changed compared to the settings during the heating operation.

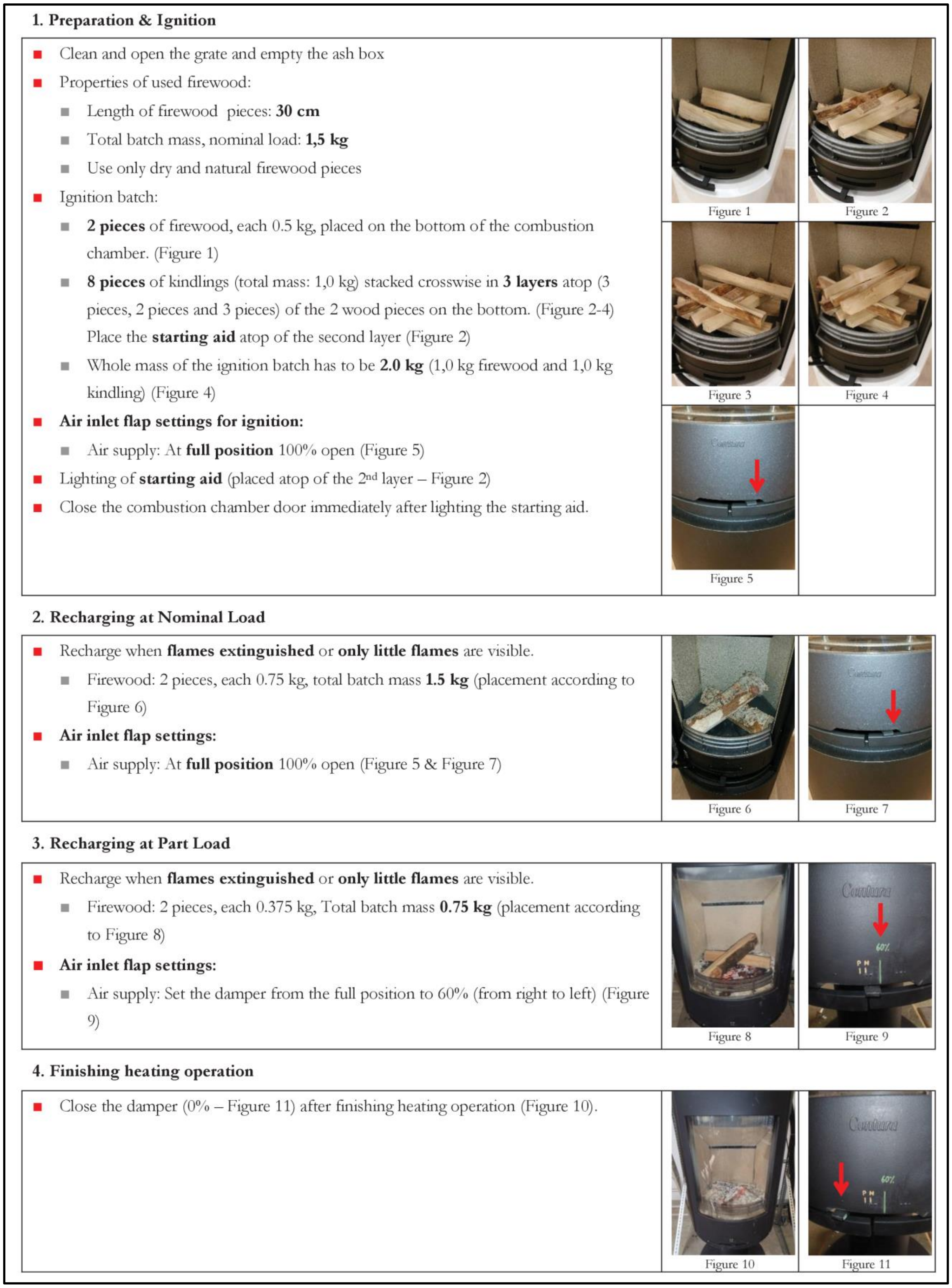
The main differences between the typical modes of operation of the three types of firewood room heating appliances are the number of performed batches of one heating operation cycle. For firewood roomheaters and residential biomass cookers the number of fuel batches per heating cycle is by trend typically higher compared to tiled stoves.

2.4.3 Potential customer friendly technological and non-technological optimization approaches

As shown in the previous section, the typical operation mode is very similar for all considered types of firewood room heating appliances. Overall, the results of the survey reveal an appropriate operation mode. Hardwood of suitable dimensions is used in general. According to the storage mode as well as the storage duration no excessive moisture content is expected. Furthermore, a clear trend of permanent overloading is not indicated. However, the results indicate a clear optimization potential regarding emission minimization and efficiency increase in real-life. Most obvious is the optimization potential regarding thermal efficiency performance by ensuring correct air valve settings for combustion air supply. Further, the use of specific starting aids instead of paper and the application of top-down ignition method instead of the commonly used bottom-up ignition method are expected to decrease emissions significantly.

As potential non-technological optimization measures external training arrangements and Quick-User-Guides are suggested. For example, obligatory Quick-User-Guides can be used to clearly specify correct and appliance specific heating operation. Hence, they should contain the most relevant specifications for environmental friendly and most efficient operation on one page. These implement the right choice of fuel and fuel properties, the fuel amount per batch for nominal and part load operation, procedure of ignition technique and correct air valve settings during and after heating operation.

The idea of the Quick-User-Guide for defining the optimal appliance specific best-practice heating operation for the end-user was later taken up in the European R&D project "*beReal*" [88] (more details described in chapter 7). An exemplary Quick-User-Guide is illustrated in **Figure 9**.



1. Preparation & Ignition

- Clean and open the grate and empty the ash box
- Properties of used firewood:
    - Length of firewood pieces: **30 cm**
    - Total batch mass, nominal load: **1,5 kg**
    - Use only dry and natural firewood pieces
- Ignition batch:
    - **2 pieces** of firewood, each 0.5 kg, placed on the bottom of the combustion chamber. (Figure 1)
    - **8 pieces** of kindlings (total mass: 1,0 kg) stacked crosswise in **3 layers** atop (3 pieces, 2 pieces and 3 pieces) of the 2 wood pieces on the bottom. (Figure 2-4) Place the **starting aid** atop of the second layer (Figure 2)
    - Whole mass of the ignition batch has to be **2.0 kg** (1,0 kg firewood and 1,0 kg kindling) (Figure 4)
- **Air inlet flap settings for ignition:**
    - Air supply: At **full position** 100% open (Figure 5)
- Lighting of **starting aid** (placed atop of the 2nd layer – Figure 2)
- Close the combustion chamber door immediately after lighting the starting aid.

Figure 1 Figure 2 Figure 3 Figure 4 Figure 5

2. Recharging at Nominal Load

- Recharge when **flames extinguished** or **only little flames** are visible.
    - Firewood: 2 pieces, each 0.75 kg, total batch mass **1.5 kg** (placement according to Figure 6)
- **Air inlet flap settings:**
    - Air supply: At **full position** 100% open (Figure 5 & Figure 7)

Figure 6 Figure 7

3. Recharging at Part Load

- Recharge when **flames extinguished** or **only little flames** are visible.
    - Firewood: 2 pieces, each 0.375 kg, Total batch mass **0.75 kg** (placement according to Figure 8)
- **Air inlet flap settings:**
    - Air supply: Set the damper from the full position to 60% (from right to left) (Figure 9)

Figure 8 Figure 9

4. Finishing heating operation

- Close the damper (0% – Figure 11) after finishing heating operation (Figure 10).

Figure 10 Figure 11

Figure 9: Example of a Quick-User-Guide elaborated in the R&D project "*beReal*" as a non-technological measure to enable the optimal appliance specific best-practice user behavior [88]

Additionally, non-technological optimization measures should be also supported and combined by the development and implementation of technological measures. For instance, the development and implementation of automatic control systems for correct adapted combustion air supply. Especially for firewood roomheaters and residential biomass cookers, such systems can reduce gaseous and particulate emissions and also increase thermal efficiency during heating operation (for details see chapter 3.4.3). Furthermore, they would increase the user comfort during heating operation. Additionally, the systems are useable for an automatically close of air valves after finishing heating operation for avoidance of thermal heat losses during cooling down and stand-by conditions. Consequently, this will enhance the efficiency factor and will also decrease emissions since the heat demand of the room will be lower and therefore less operation times are required. Possibly, other kinds of secondary abatement technologies, like catalysts or filters, should be combined with the mentioned non-technological and technological measures in order to minimize emissions also in transient conditions or during maloperation. Especially catalysts are expected as a suitable secondary abatement measure since they are supposed to reduce gaseous and particulate emissions, have a low pressure drop as well as work without electrical current.

2.5 Summary – Survey about user behavior in Austria

For defining suitable measures to improve the real-life operation performance of firewood room heating appliances a user survey was conducted. Thereby, the typical user behavior was evaluated and potential differences of the operation mode of the respected technologies were identified.

In total 108 users were interviewed and the results were analyzed in comparison with further data of more than 600 Austrian wood stove users [83]. The typical user behavior was defined for users of firewood roomheaters, tiled stoves and residential biomass cookers. These three types of appliances represent almost 95% of biomass room heating appliances that are used in Austria. The evaluation of the user data showed a clear picture of typical operation mode of firewood room heating appliances in Austria regarding used fuels, mode of ignition and air valve settings during and after heating operation.

Generally the results confirmed that in principal the operation mode is very similar for all considered types of firewood room heating appliances. Overall, the results showed a convenient typical operation mode. Hardwood of suitable dimensions is used in general (61% – 67%). For fire wood roomheaters and biomass cookers typically two pieces of firewood are used whereas for tiled stoves three and more pieces are used for one batch. However, since tiled stoves are often only fueled with one or two batches no overload was indicated. According to the storage mode as well as the storage duration no excessive moisture content of the firewood is expected. Although the results pointed out that only about one third operate their appliances knowingly according to the manual, a clear trend of permanent off-specification operation, e.g. by overloading, was not observed.

However, the results indicate a clear optimization potential regarding minimized emissions and increased efficiency in real-life. Most obvious is the optimization potential regarding thermal efficiency performance by ensuring correct air valve settings for combustion air supply. Especially the closing of the air valves after finishing heating operation would guarantee less thermal heat losses and therefore a higher overall efficiency. Further, the use of specific starting aids instead of paper and the application of top-down ignition method instead of the commonly used bottom-up ignition method are expected to decrease emissions significantly. Moreover, the user survey confirmed the need to develop user friendly education measures, especially for users of firewood roomheaters and residential biomass cookers. For example external training arrangements and Quick-User-Guides are suggested. Furthermore, the non-technological optimization measures should be supported by the development and implementation of technological measures, e.g. automatically air control systems for correct adapted combustion air supply or secondary abatement systems, e.g. catalysts or filters.

3 IMPORTANT OPERATIONAL ASPECTS – IGNITION TECHNIQUE AND DRAUGHT CONDITIONS[3]

3.1 State of the art

3.1.1 Ignition technique

In principal, two different ignition techniques can be distinguished – the top-down ignition technique and the bottom-up ignition technique. The principal design of fuel and kindling material of both ignition techniques is illustrated in **Figure 10**.

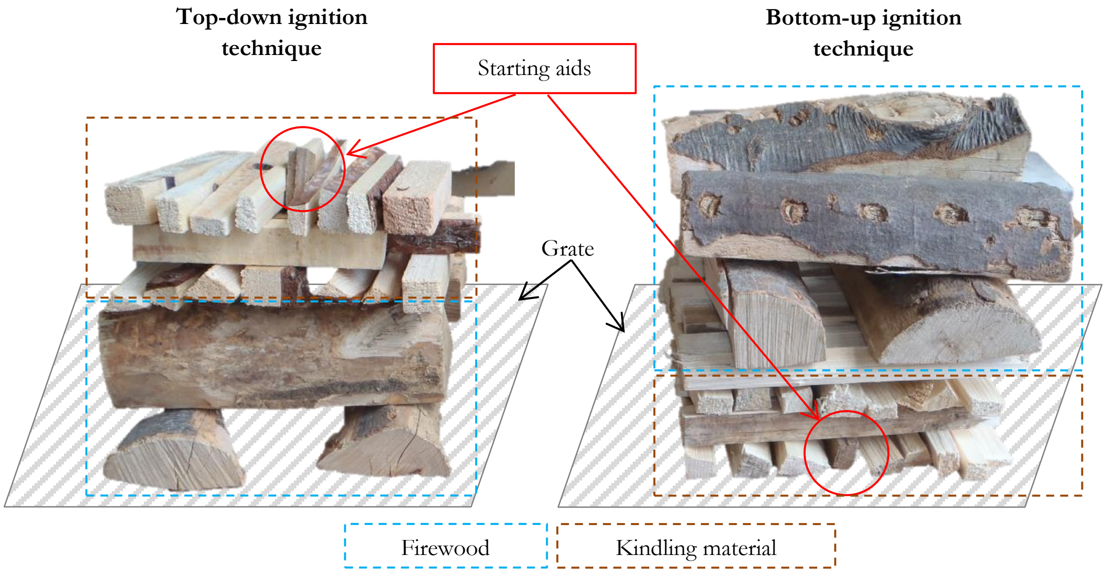


Figure 10: Design of fuel and kindling material of the top-down ignition technique (left) compared to predominantly used bottom-up ignition technique (right)

As already described in the introduction (chapter 1.1), top-down ignition means that firewood pieces are directly placed on the grate. The kindling material is placed above the firewood pieces and on the top of the kindling material the specific starting aids are located. Then, the starting aids are lighted and the combustion process is induced by a subsequent downward ignition of the kindling material and

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[3] Segments of this section have already been published in [2].

firewood pieces on the grate. In contrast, bottom-up ignition means the placement of kindling material and starting aids directly on the grate below a few pieces of firewood. Then, the starting aids on the grate are lighted and the combustion process is induced upward the fuel batch (**Figure 10**). Instead of specific starting aids, paper is used frequently. According to the Austrian user survey (chapter 2) the use of paper is preferred compared to the use of specific starting aids (**Table 8**, Question 4.3). In contrast, specific starting aids are predominantly used in Europe [74].

According to the results of the user survey described in the previous chapter only around 10% of the Austrian users of firewood roomheaters commonly apply the top-down ignition technique. Also the European survey showed that the bottom-up ignition technique is the predominant ignition procedure [74]. However, most of the available leaflets concerning "correct heating operation" propose the top-down ignition technique as advantageous compared to the bottom-up ignition technique due to lower emissions. But, since there are only limited experimental data available, the question is still, if the top-down ignition technique is in general better compared to the bottom-up ignition technique. Furthermore, the assumption that the ignition technique may be a significant contribution to reduce real-life emissions of firewood stoves is not yet underpinned by a satisfying number of systematic studies, even more as recent works showed that top-down ignition technique can even result in higher gaseous CO and OGC emissions compared to bottom-up ignition technique [90].

3.1.2 Draught conditions

Draught conditions represent one of the most relevant operating conditions referring to the infrastructure of the heating appliance (chapter 1.1). Flue gas draught conditions determine the flow conditions and the overall mass flow of the combustion air supply at specific air valve settings. Therefore, draught conditions significantly influence the combustion conditions and consequently are important for emissions and thermal efficiency performance of the appliance. The relevant characteristics for induced natural draught conditions during heating operation are the properties of the chimney system, like material and dimensions (diameter and height), weather conditions and flue gas temperature [46] [49].

In principal, natural draught conditions are induced by natural convection processes which base on the density difference of the flue gases. The density difference results from the different temperatures at the inlet and outlet of the chimney system. Modern state-of-the-art chimney systems enable the supply of combustion air and the evacuation of flue gases (**Figure 11**). This enables the operation of the stove independently from the air of the installation room which is often required for airtight modern residential buildings.

The principal characteristics of the natural draught conditions within the chimney and the typical components of a state-of-the-art chimney which is suitable for roomsealed heating systems are illustrated in **Figure 11**.

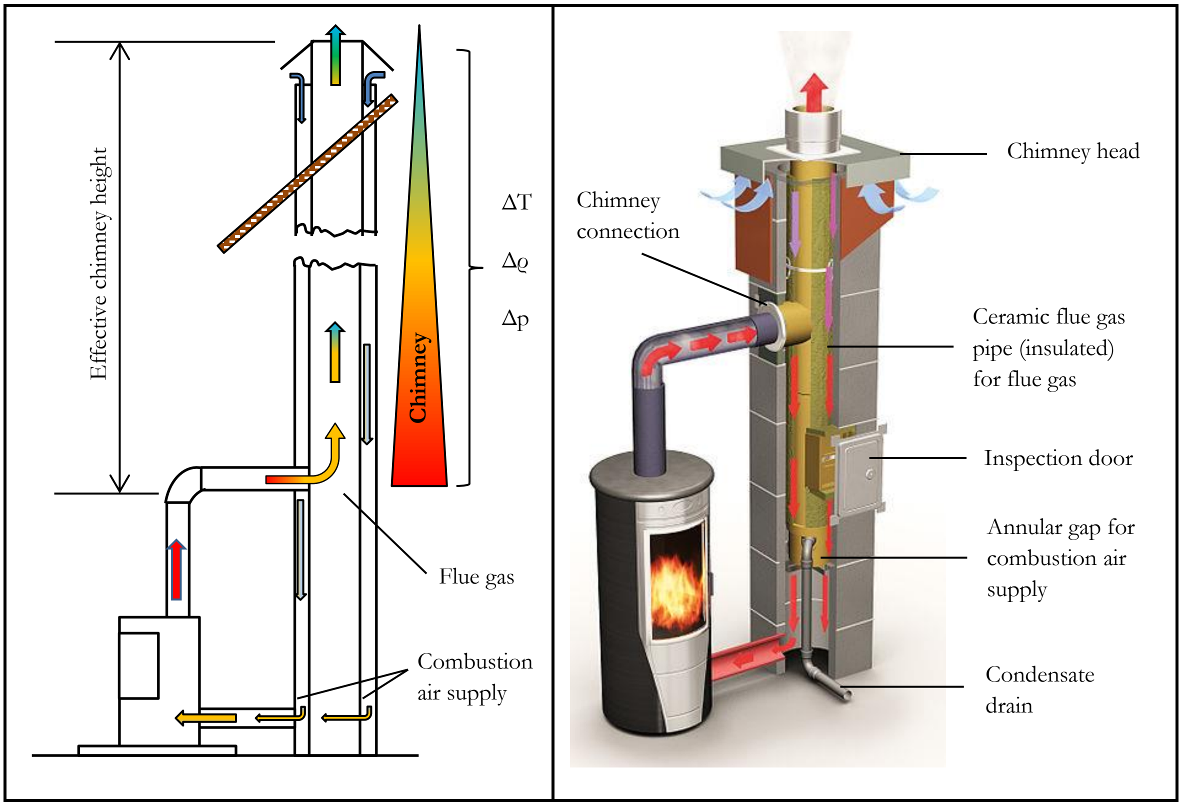

Figure 11: Caharcteristic pressure and temperature conditions of the flue gas within the chimney (left) and the typical components of a sate-of-the-art chimney suitable for roomselaed firewood heating appliances (right) [89] (translated in English)

Generally, a sufficient draught level in the chimney is important to enable stable and safe operation. Under official-type-testing conditions a constant flue gas draught is applied for firewood roomheaters, [13]. Therefore, the flue gas draught is controlled at a certain point to constant settings of 12 ± 2 Pa during the whole test by using a ventilator. If the flue gas draught exceeds 12 Pa the volume flow of combustion air supply increases (at same air valve settings). Consequently, combustion conditions are affected and hence emission formation and thermal efficiency are influenced with lower thermal efficiency at higher draught conditions [49]-[51]. However, so far no clear correlation between flue gas draught and gaseous as well as particulate emissions was observed.

As identified by the Austrian user survey (chapter 2) the majority of respondents evaluated the draught conditions of their appliances as “optimal draught conditions” (**Table 7**). In addition, the findings of the European user survey showed that 66% of the related chimneys are between 5 m and 10 m high [74]. Measurements under lab conditions [3] (chapter 6, **Figure 61**) and during long term field tests performed with respective chimney heights revealed that these chimney heights typically correspond with a flue gas draught between 20 Pa and 30 Pa (**Figure 12**, according to [49]).

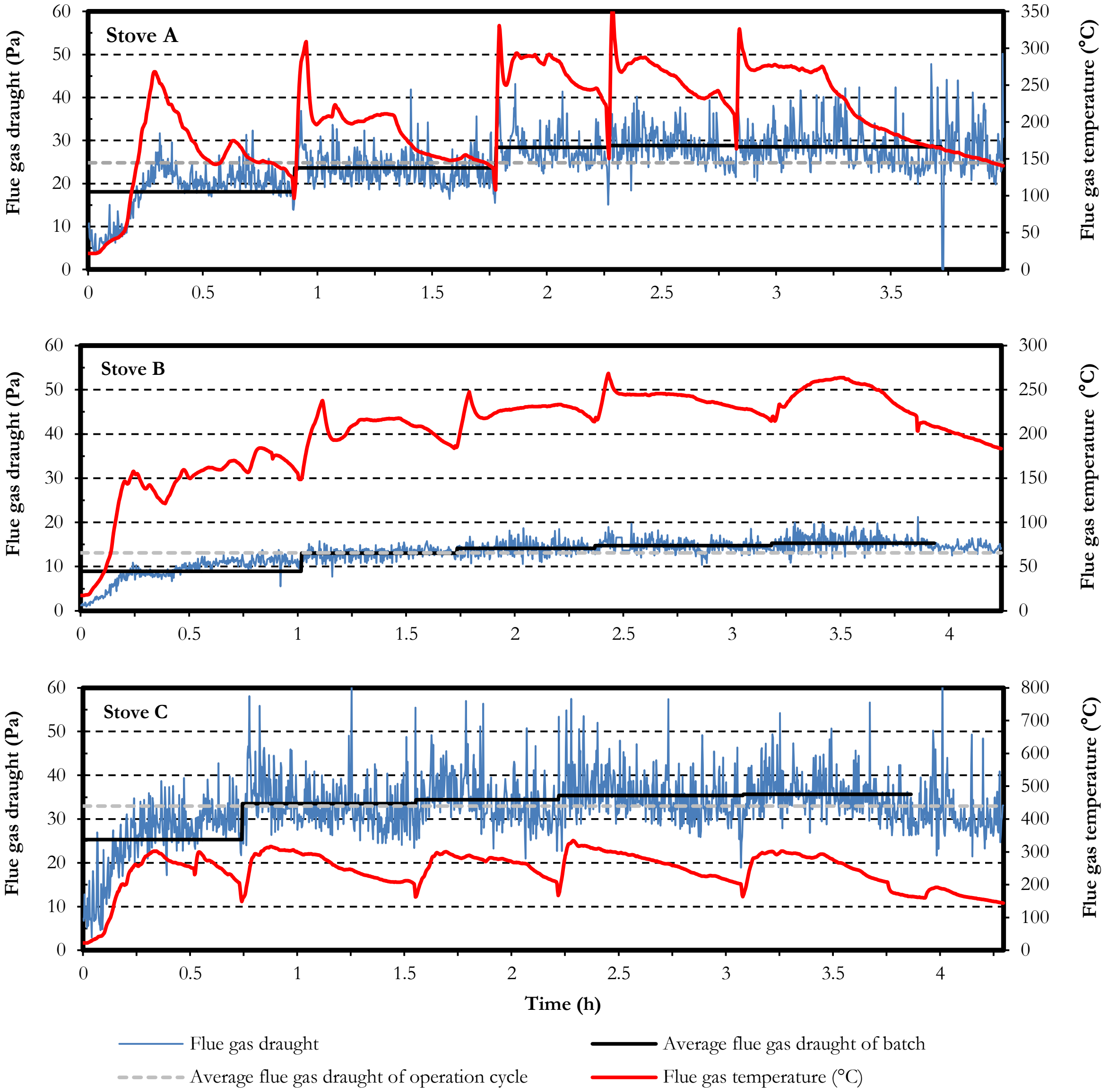

Figure 12: Flue gas draught measurements at three field test sites with different effective chimney heights (Stove A: 6.0 m, top; Stove B: 3.4 m, middle; Stove C: 7.2 m, bottom) during a five batch heating cycle according to measurements of [49]

As illustrated in **Figure 12**, the average flue gas temperatures were similar for all three stoves in the range of 200 °C to 300 °C. The combustion air supply of stove A and B were adapted manually and remained on constant settings during the single batches. Stove C was a stove equipped with an automatic combustion air control system (open-loop control) which adapts the air valve settings during the batch based on temperature measurements of a temperature sensor located in the combustion chamber. The highest effective chimney height was measured for stove C (7.2 m) and resulted also in the highest average flue gas draught conditions of 32 Pa. The lowest average flue gas draught of 13 Pa was observed for stove B which had also the lowest effective chimney height (3.4 m). The average flue gas draught of stove A was measured at 24 Pa. Stove A was connected to a chimney with an effective chimney height of 6.0 m. Consequently, the measurement results clearly confirmed the principal correlation of the effective chimney height and average flue gas draught conditions. The progress of chimney draught conditions was similar for all three stoves. During the first 30 minutes of heating operation the flue gas draught measurements increased clearly. But after the first batch, the average flue gas draught conditions increased only slightly between the batches 2 – 5. For example, the average flue gas draught conditions of batch 2 to 5 varied between 13 Pa and 14 Pa for stove B or between 25 Pa and 29 Pa for stove A (**Figure 12**). The fluctuations of draught conditions which were measured with a measuring interval of 10 s resulted most probably due to varying combustion conditions, measurement precision and weather conditions. For stove C the fluctuations which might derive from the air valve adaptions of the automatic combustion air supply control system are highest (**Figure 12**).

In REICHERT et al. 2016 [88] flue gas draught conditions from nine different firewood roomheaters are presented. The data were evaluated based on long term field measurements of a measuring period of three to four months in the winter season. As illustrated in **Figure 13**, chimney draught conditions were in a range between 3 Pa and 28 Pa[4]. The average real-life flue gas draught calculated on the basis of those measurements was specified at 18 Pa.

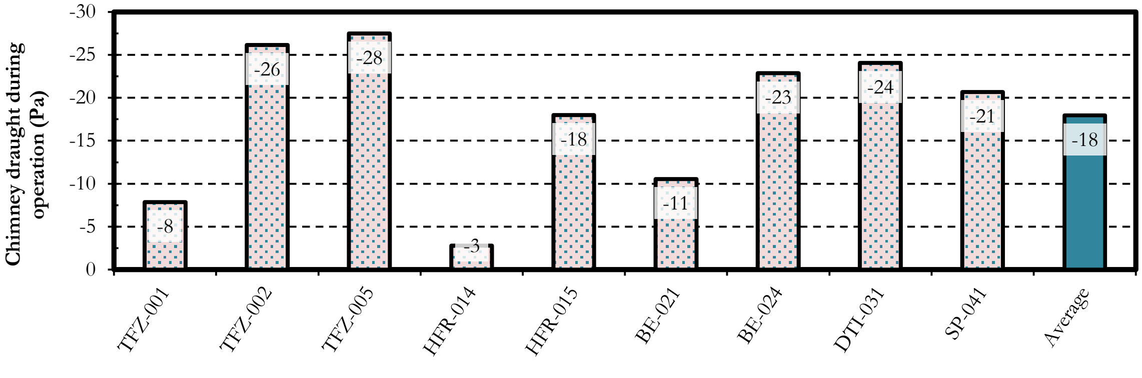

Figure 13: Avearge flue gas draught conditions of nine selected firewood stoves (classified according to EN 13240) during real-life heating operation at the end-user evaluted for a time period of three to four months [88]

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[4] The draught conditions of this thesis are specified by positive numbers although they refer to a static under pressure. This is due to the definitions of the respective EN standards (e.g. EN 13240 [13]) where flue gas draught condition are always specified by positive numbers.

The results of [49] and [88] confirmed that the draught level of 12 Pa applied during standard type testing procedure [13] is exceeded in most cases under real-life operating conditions at the end-users. However, so far, there are no systematic studies that evaluate the effect of the flue gas draught on the combustion performance of different appliances.

3.2 Approach

In the following, comprehensive experimental combustion tests are presented. They were carried out at different firewood roomheaters for a systematical evaluation of the effect of different ignition techniques and flue gas draught conditions regarding gaseous and particulate emissions as well as thermal efficiency. For repeatability reasons the measurements were performed under constantly controlled draught conditions. Furthermore, this seems appropriate since it was shown by field test results that draught conditions in real-life increase to a stable level within a short time (**Figure 12**).

The findings shall clarify the results of previous studies. Additionally, they shall identify potential measures to improve the real-life behavior of firewood roomheaters and support the respective technological development. Moreover, the results may be useful contributions to legislative and normative processes and procedures.

3.3 Materials and methods

3.3.1 Performed test series and general testing procedure

For evaluating the effect of the ignition technique and draught conditions two different test series were conducted:

I. Ignition technique test series – Performance of comparative ignition combustion tests regarding differences of gaseous (CO, OGC) and particulate matter (PM) emissions and thermal efficiency (η): The test runs were conducted under controlled constant draught conditions of 12 ± 2 Pa at two different firewood roomheaters (**Table 10**) applying the top-down and bottom-up ignition technique. Softwood (spruce – "*picea abies*") and hardwood (beech – "*fagus sylvatica*") were used as kindling material for each ignition technique. For each variation three ignition combustion tests were performed.

II. Draught conditions test series – Investigation and assessment of the effect of draught conditions on CO, OGC and PM emissions as well as on thermal efficiency (η): Combustion test cycles were conducted at 12 ± 2 Pa, 24 ± 2 Pa and 48 ± 2 Pa. For each draught setting, combustion test cycles were performed with three different firewood roomheaters (**Table 10**).

All combustion tests were conducted under constant draught conditions. For test series I only the ignition batch was performed. For test series II several combustion test cycles consisting of five consecutive batches were carried out starting from cold conditions (**Figure 14**).

The ignition procedure was done consistently over all tests of a test series either top-down or bottom-up. As kindling material spruce or beech kindling together with specific starting aids were used. The second fuel batch was recharged when flames of batch 1 were extinguished. The subsequent fuel batches (batch 3-5) were placed directly on the firebed when the CO_2 concentration (v/v) of the flue gas declined to 25% of the maximum peak of CO_2 during the respective batch. This corresponded well with the recharging criteria of a maximum variation of firebed mass ±0.05 kg according to the standard EN 13240 [13]. The adjustment of air valve settings for combustion air supply was done manually. During the ignition (Test series I) and preheating batch (Test series II) the air valve settings for primary and secondary air were fully open for all test runs. After the preheating batch the air valve settings were adapted according to the specifications of the user manual on constant settings (Test series II).

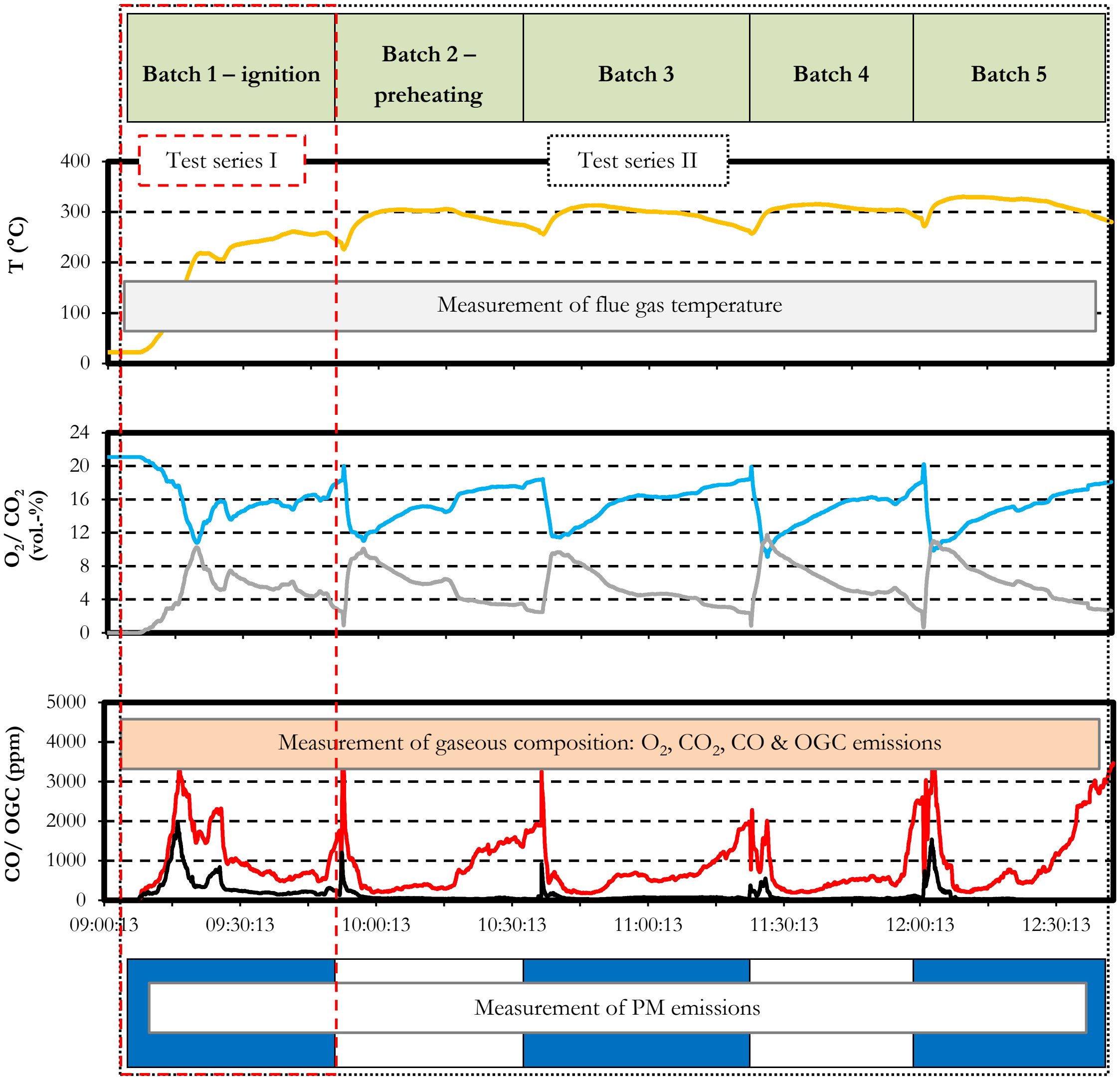


Figure 14: Testing procedure of combustion test cycles of test series I (only batch 1) & II (five consecutive batches)

Flue gas temperature, flue gas draught and gaseous emissions were measured continuously over the whole test duration. The flue gas temperature for determination of thermal heat losses for indirect efficiency calculation was measured with a thermocouple centrally placed in the flue gas pipe (**Figure 15**). The gravimetric PM measurement was done discontinuously over the complete batch duration (Test series I: ignition batch/Test series II: batch 1, 3, 5). The PM sampling started just before opening the combustion chamber door for recharging and was terminated right before the recharging criteria for the subsequent batch was reached. The applied test procedure reflects better real-life operation than EN standard type test procedure as described and discussed REICHERT et al. [91].

3.3.2 Used fuels

For all tests the appliances were operated with beech firewood according to ÖNORM EN 14961-5:2011 [92]. The used fuel was provided by the local firewood producer HOFEGGER REINHARD

(A-3250 Wieselburg) and was stored covered outside until thermal utilization. **Table 9** summarizes relevant physical and chemical data of the used firewood and kindling material.

Table 9: Relevant physical and chemical data of used firewood (beech = "*Fagus sylvatica*"; spruce = "*picea abies*")

	Ø Length (m)	**Moisture content* (W) (kg/kg)**	**Net calorific value (H_u) (MJ/kg d.b.)**	**Ash content (a) (g/kg d.b.)**	**Carbon (C) (kg/kg d.b.)**	**Hydrogen (H) (kg/kg d.b.)**	**Nitrogen (N) (kg/kg d.b.)**	**Sulfur (S) (mg/kg d.b.)**
Standard for analysis	-	EN 14774-1:2009 [93]	EN 14918: 2010 [94]	EN 14775:2009 [95]	EN 15104:2011 [96]			EN 15289:2011 [97]
Beech ("*Fagus sylvatica*") firewood	0.25	0.11 – 0.14	17.7	8.6	0.473	0.062	0.0015	0.1
Beech ("*Fagus sylvatica*") kindling	0.25	0.11 – 0.14	17.7	8.6	0.473	0.062	0.0015	0.1
Spruce ("*Picea abies*") kindling	0.25	0.09	18.3	4.5	0.487	0.631	< 0.001	< 0.1

d.b. = dry based/ * as received

3.3.3 Overview of used appliances, scheme of test setup and detailed testing procedure

Four different firewood roomheaters according to EN 13240 were used [13] (**Table 10**). The ignition tests (Test series I) were performed using only roomheater A and B.

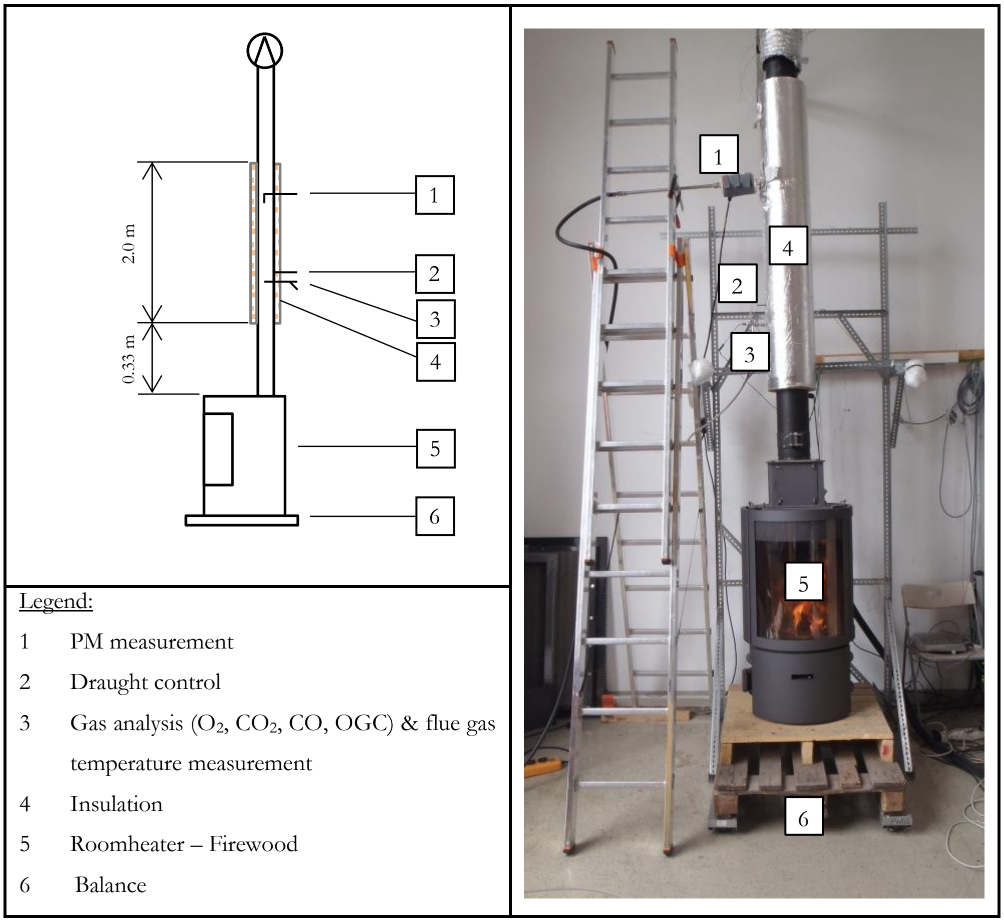
Table 10: Overview of relevant data of used firewood roomheaters

Parameter	**Roomheater A**	**Roomheater B**	**Roomheater C**	**Roomheater D**
Nominal heat output	10 kW	8 kW	6 kW	8 kW
Mass	215 kg	140 kg	76 kg	256 kg
Flue outlet	0.15 m	0.15 m	0.15 m	0.15 m
Year of production	2014	2010	2010	2013
Primary and secondary air valves independently adjustable?	no – one stageless valve adjustable from 0% to 100%	no – one stageless valve adjustable from 0% to 100%	yes – two stageless valves adjustable from 0% to 100%	yes – two stageless valves adjustable from 0% to 100%
Primary air supply	from the front	through the grate	through the grate	through the grate
Secondary air supply	from the back & window flushing air			
Volume and dimensions of combustion chamber	0.0433 m³ (0.35 m diameter*, 0.45 m height)	0.0530 m³ (0.35 m diameter*, 0.55 m height)	0.0230 m³ (0.3 m width, 0.32 m depth, 0.31 m height)	0.0593 m³ (0.38 m width, 0.3 m depth, 0.52 m height)

Parameter	Roomheater A	Roomheater B	Roomheater C	Roomheater D
Material of combustion chamber	grey cast iron and fire clay	grey cast iron and fire clay	grey cast iron and vermiculite	grey cast iron and fire clay
Test series I	✓	✓	-	-
Test series II	✓	-	✓	✓

* round combustion chamber

The ignition tests referring to roomheater B and the draught tests referring to roomheater D were performed at the TFZ Straubing. The combustion tests on the effect of different draught conditions (Test series II) were carried out with roomheater A, C and D representing typical state-of-the-art firewood roomheaters. Furthermore, roomheater A and B were used for the ignition tests since they had different designs of the primary air supply (**Table 10**). According to the guidance for correct heating of ESSER et al. 2014 [100] the bottom-up ignition technique should be advantageous when the primary air is supplied via the grate. The top-down ignition technique should be advantageous if this is not the case. **Figure 15** shows the scheme of the test setup applied for both test series.



Legend:
1 PM measurement
2 Draught control
3 Gas analysis (O_2, CO_2, CO, OGC) & flue gas temperature measurement
4 Insulation
5 Roomheater – Firewood
6 Balance

Figure 15: Scheme of the test setup (left) applied for test series I and test series II and exemplary picture of roomheater A

The flue gas pipe diameter of the measurement section was 0.15 m. The insulation of the measurement section started 0.33 m downstream the flue outlet. Gas analysis was carried out 0.6 m downstream the flue outlet. The measuring point of pressure drop for draught control was located 0.1 m after the gas analysis. The measuring point for PM sampling was located 1.5 m after the flue gas outlet.

3.3.4 Detailed testing procedure: test series I – ignition technique

Roomheater A and B were used for the ignition tests. Length of the used firewood pieces was 0.25 ± 0.03 m and the mass of each firewood piece was 0.6 ± 0.06 kg. For both ignition techniques four firewood pieces were used. The total mass of kindling material was 0.5 ± 0.02 kg. The total mass of an ignition batch was 2.9 ± 0.06 kg. Firewood pieces as well as kindling material were placed crosswise for both ignition techniques (**Table 11**).

Table 11: Conducted variations of test series I – ignition technique (exemplary pictures at roomheater A for both ignition techniques at lighting and 5 minutes later)

Top-down ignition technique		Bottom-up ignition technique	
			
Variation 1	**Variation 2**	**Variation 3**	**Variation 4**
Beech firewood & spruce kindling (Spruce + Beech)	Beech firewood & beech kindling (Beech + Beech)	Beech firewood & spruce kindling (Spruce + Beech)	Beech firewood & beech kindling (Beech + Beech)

The kindling material was lighted using commercial starting aids (wax-covered wood wool). For the top-down ignition technique the starting aid was placed on the top of kindling material, whereas for the bottom-up ignition technique the starting aid was placed directly on the grate next to the kindling material. For each variation, three single test runs were performed.

3.3.5 Detailed testing procedure: test series II – draught conditions

Roomheaters A, C and D were used for this test series. For each draught level one test cycle according to **Figure 14** was applied. The operation procedure was equal for each test run of the respective appliance. For all roomheaters the bottom-up ignition technique was applied using a defined amount of spruce kindling material, two specific starting aids and several pieces of beech firewood including bark. For all roomheaters primary and secondary air supply was fully open during the ignition and preheating batch. Subsequently, air valve settings were adapted manually and then remained unchanged till the end of the test cycle. The ambient air conditions were similar for all conducted tests. The ambient temperature at the test room was 22 ± 3 °C and the relative humidity ranged between 40% and 60%. Therefore, the total amount of air supply at a certain damper setting was sufficiently comparable for the respective stove and combustion tests. Furthermore, flue gas draught conditions that control the absolute flue gas mass flow were fixed at constant settings for respective test runs. More detailed information about the specific operating procedure is presented in **Table 12**.

Table 12: Fuel specifications and air valve settings of each roomheater applied during test series II

Parameter	Roomheater A	Roomheater C	Roomheater D
Ignition batch	4 pieces (each 0.6 kg, 0.25 m) + 0.5 kg kindling	4 pieces (each 0.4 kg, 0.25 m) + 0.5 kg kindling	3 pieces (each 0.66 kg, 0.25 m) + 0.6 kg kindling
Recharging	3 pieces (each 0.8 kg, 0.25 m)	2 pieces (each 0.8 kg, 0.25 m)	2 pieces (each 1.0 kg, 0.25 m)
Air valve settings (after ignition & preheating)	80% (primary and secondary air controlled)	Primary air: 0% Secondary air: 100%	Primary air: 15% Secondary air: 100%

3.3.6 Measurements, data evaluation and statistical analysis

The following measurements were done (see also **Figure 15**):

Continuous measurements:

- Gas analysis of CO_{low} (0 – 5000 ppm), CO_{high} (0 – 10 vol.-%), CO_2 (0 – 20 vol.-%) – NDIR, O_2 (0 – 25 vol.-%) – paramagnetic (NGA 2000 – MLT4 gas analyzer), OGC (0 – 100 ppm /10000 ppm) – FID (M&A Thermo-FID PT63LT)
- Temperature measurement with thermocouples, Type K – class 1 (-50 – 1000 °C, ± 1.5 °C of measured value) – thermoelectric effect (Seebeck)
- Static pressure difference for draught control (0 – 100 Pa, ± 1.5% of measured value) – piezoelectric effect (Thermokon DPT 2500-R8)
- Mass loss of fuel combustion: Palette balance, Mettler Toledo PTA 455-600 (0 – 600 kg, ± 0.05 kg of measured value)

Discontinuous measurements:

- Gravimetric measurement of PM emissions (out-stack) with stuffed quartz filter cartridges and downstream plane filters using a constant sampling rate of 10 l/min, STP and a diameter of sampling nozzle of 12 mm. Rinsing of sample probe was done with acetone. Pre- and post-conditioning of filters and rinsing tanks as well as heating of filter holder device were conducted at 160 °C. Cooling down of filters and rinsing tanks in a desiccator lasted for at least 8 h.

- Mass measurement of stuffed PM filter cartridges and plane filters before and after measurement was performed with a precision balance for laboratory (Sartorius ME 235P, 0 – 60 g, ±0.01 mg)
- For measurements of fuel batch mass a precision balance was used (Sartorius AW-8201, 0-8201 g, ± 0.1 g)

The analysis of CO emissions was done according to the standard EN 13240 [13], the analysis of OGC emissions was done according to CEN/TS 15883 [98]. Thereby, the average emission concentration of each combustion test cycle was calculated including all measurements of the combustion test (mg/m^3, STP, dry, based on 13 vol.-% O_2). For PM emissions the time-weighted average emission concentration of the combustion test cycle was calculated using all conducted PM measurements.

The lambda value (λ) was calculated based on flue gas composition using the residual oxygen measurement values ($O_{2,mean}$) (**Eq. 1**).

$$\lambda = 21/(21 - O_{2,mean}) \quad \textbf{(Eq. 1)}$$

$O_{2,mean}$ Average O_2 concentration of the respective test batch in dry flue gas; in % of volume

Thermal efficiency was determined indirectly according to ÖNORM EN 13240:2001 + AC: 2003 standard [13] (**Eq. 2**). Therefore, the thermal ($q_{a,EN}$) and chemical ($q_{b,EN}$) flue gas losses were determined (**Eq. 3** – **Eq. 4**). For the losses due to combustible constituents in the residues ($q_{r,EN}$) a constant value of 0.5% was used as proposed by the EN test protocol.

$$\eta = 100 - (q_{a,EN} + q_{b,EN} + q_{r,EN}) \quad \textbf{(Eq. 2)}$$

η	Thermal efficiency; in %
$q_{a,EN}$	Proportion of thermal flue gas losses; in %
$q_{b,EN}$	Proportion of chemical flue gas losses; in %
$q_{r,EN}$	Proportion of losses through combustible constituents in the residues; in % For firewood: 0.5% absolutely is predefined by the standard EN 13240

The specific thermal ($Q_{a,\,EN}$) and chemical ($Q_{b,\,EN}$) heat losses are calculated on the basis of average batch values of the flue gas and room temperature measurements as well as on fuel composition. The specific heat of the dry flue gases (C_{pmd}) and water vapor (C_{pmH2O}) are calculated in relation to the flue gas temperature, ambient temperature and the CO_2 concentration of the dry flue gas. The factors in the equations are specified in the standard and base on empirical data and on combustion calculations. In **Eq. 3.a** and **Eq. 4a** for example, the factor 0.536 represents the volume of CO_2 (in m^3, STP) generated from the stoichiometric combustion of 1 kg carbon (C) with oxygen, the factor 1.244 represents the specific volume of water vapor from the oxidation of 1 kg hydrogen with oxygen under stoichiometric conditions (**Eq. 3a**). In **Eq. 4a** the factor 12644 represents the volumetric heating value of carbon monoxide (in kJ/m^3, STP).

$$q_{a,\,EN} = \frac{Q_{a,\,EN}}{H_{u,\,f}} \times 100 \tag{Eq. 3}$$

$$Q_{a,\,EN} = \left(T_{flue\ gas} - T_{ambient\ air}\right)\left(\frac{C_{pmd}\ (C - C_r)}{0.536\,\left(CO_{mean} + CO_{2,\,mean}\right)} + C_{pmH2O}\ \times 1.244\ \frac{(9H + W)}{100}\right) \tag{3a}$$

$$\begin{aligned} C_{pmd} = 3.6 \times (0.361 + 0.008\ &\frac{T_{flue\ gas}}{1000} + 0.034 \times \left(\frac{T_{flue\ gas}}{1000}\right)^2 \\ &+ \left(0.085 + 0.19 \times \frac{T_{flue\ gas}}{1000} - 0.14\ \times \left(\frac{T_{flue\ gas}}{1000}\right)^2\right)\frac{CO_{2,\,mean}}{100} \\ &+ \left(0.3\ \times \frac{T_{flue\ gas}}{1000} - 0.2 \times \left(\frac{T_{flue\ gas}}{1000}\right)^2\right)\frac{CO_{2,\,mean}}{100} \end{aligned} \tag{3b}$$

$$C_{pmH2O} = 3.6 \times (0.414 + 0.038\ \times \left(\frac{T_{flue\ gas}}{1000}\right) + 0.034\ \times \left(\frac{T_{flue\ gas}}{1000}\right)^2\right) \tag{3c}$$

$$C_r = 1.4925 \times\ H_{u,\,f}\ \times 10^{-5} \tag{3d}$$

$q_{a,\ EN}$	Proportion of losses through specific heat in the flue gas (Q_a), referred to the net calorific value of the fired test fuel ($H_{u,\ f}$); in %
$Q_{a,\,EN}$	Specific thermal heat losses in the flue gas; in kJ/kg
$T_{flue\ gas}$	Average flue gas temperature of the test batch; in °C
$T_{ambient\ air}$	Average temperature of ambient air in the test room (during the test batch); in °C
C	Carbon content of used firewood (as fired); in % of mass
C_r	Carbon content of the residue, calculated based on the net calorific value ($H_{u,\ f}$) acc. to Eq. 3d
CO_{mean}	Average CO concentration of the respective test batch in the dry flue gas; in % of volume
$CO_{2,\,mean}$	Average CO_2 concentration of the respective test batch in dry flue gas; in % of volume
H	Content of hydrogen of used firewood (as fired basis); in % of mass
W	Moisture content of used firewood; in % of mass
C_{pmd}	Specific heat of dry flue gas in standard conditions, depending on temperature and flue gas composition; in $kJ/(Km^3)$

C_{pmH2O}	Specific heat of water vapor in the flue gas in standard conditions, depending on temperature; in kJ/(Km³)

$$q_{b,EN} = \frac{Q_{b,EN}}{H_{u,f}} \times 100 \qquad \text{(Eq. 4)}$$

$$Q_b = 12644 \times CO \times \left(\frac{C - C_r}{0.536\,(CO_{mean} + CO_{2,mean})\,100}\right) \qquad \text{(4a)}$$

$q_{b,EN}$	Proportion of losses through latent heat in the flue gas (Q_b), referred to the net calorific value of the fired test fuel ($H_{u,\ f}$); in %
$Q_{b,EN}$	Specific chemical heat losses in the flue gas; in kJ/kg

For statistical analysis two sided Students' t-tests were used. The following interpretations are used for the p values resulting by Students' t-tests:

- $p \leq 0.05$ significant difference or correlation
- 0.32 $p > 0.05$ no significance, but a clear trend of difference or correlation
- $p > 0.32$ no significance and no trend of difference or correlation

In chapter 3.4 exact p-values are given in brackets for each statistical test.

3.4 Results and discussion

3.4.1 Test series I – ignition technique

Effect kindling material

The use of small firewood pieces as kindling material for the ignition procedure is typical for all types of batch wise fired room heating appliances (chapter 2) [74]. Commercially sold kindling material is usually made of softwood, e.g. spruce. But if the kindling material is prepared by the users themselves, the kindling material is most probably small pieces of the same firewood than used for recharging. In that case it is predominantly hardwood, e.g. beech (chapter 2) [74]. Therefore, the potential effect of hardwood or softwood as kindling material on the combustion performance of the ignition batch was investigated. The statistical analysis by Students' t-test of different test runs of each ignition technique showed no significance regarding differences of CO, OGC and PM emissions as well as thermal efficiency due to the use of different kindling material (**Figure 16 – Figure 18**).

Further, the curves of CO, OGC, CO_2, O_2 and flue gas temperature confirmed that the ignition process is very similar when using spruce or beech as kindling material (**Figure 19 – Figure 22**). Nevertheless some trends were visible regarding the impact of kindling material on the ignition performance.

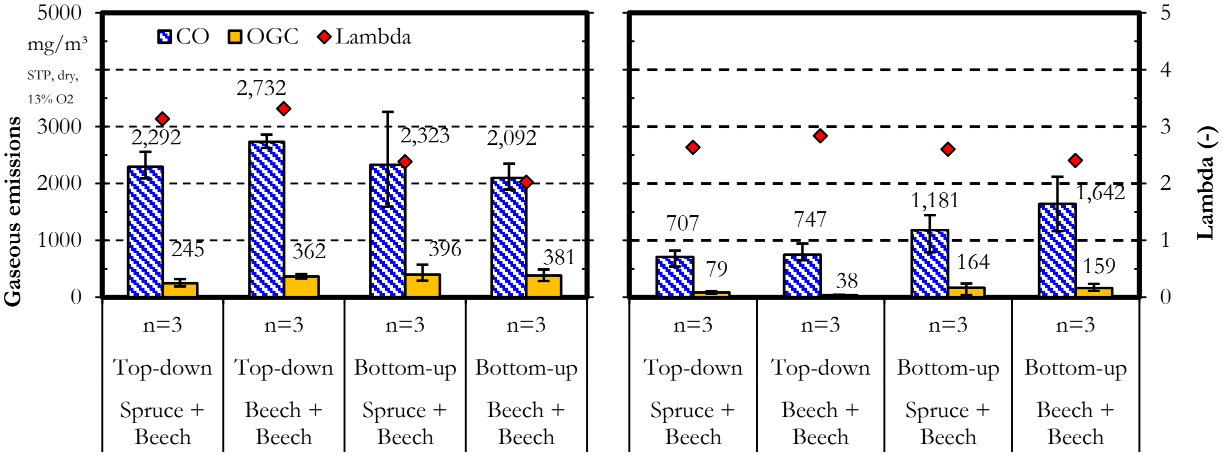


Figure 16: Average results of ignition test runs of roomheater A (left) and B (right) regarding CO, OGC and lambda. Error bars represent the maximum and minimum values determined.

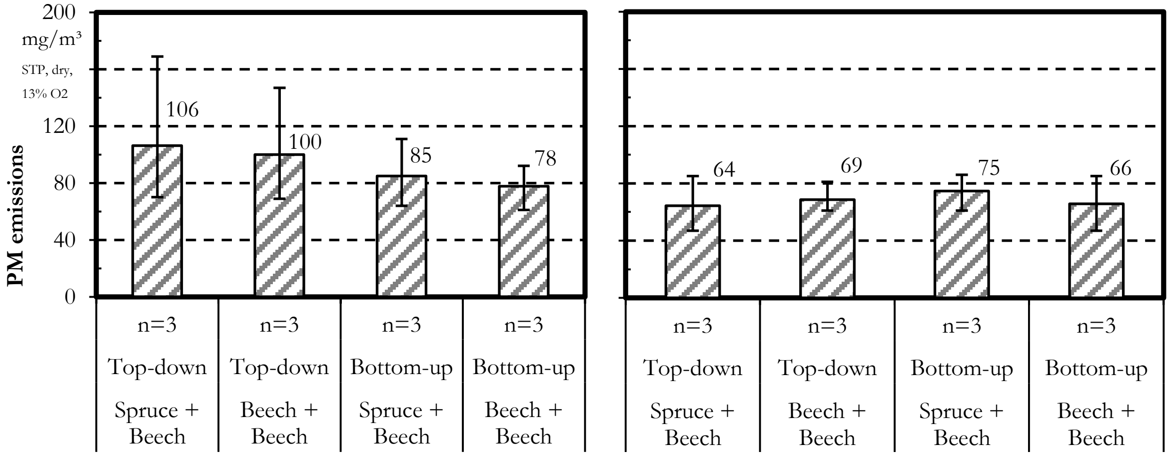


Figure 17: Average results of ignition tests of roomheater A (left) and B (right) regarding PM emissions. Error bars represent the maximum and minimum values determined.

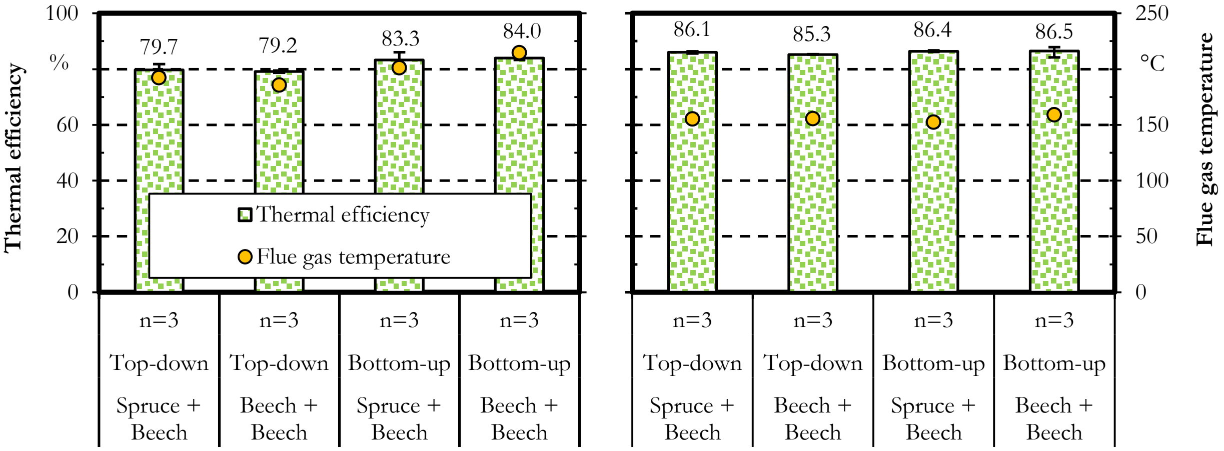


Figure 18: Average results of ignition tests of roomheater A (left) and B (right) regarding thermal efficiency and lambda. Error bars represent the maximum and minimum values determined.

Thermal efficiency of the ignition batch was only marginally different using spruce or beech kindling material. Also differences for PM emissions were low, at about 10%. The highest impact of kindling material was found for CO and OGC emissions. The highest deviations regarding OGC results for top-down ignition technique revealed the trend that top-down ignition might be more sensitive towards the kind of kindling material. However, no clear correlation was evident since for roomheater A spruce kindling led to lower OGC emissions ($p = 0.077$). In contrast, beech kindling led to lower OGC emissions for roomheater B ($p = 0.082$). For CO emissions a trend of an impact for top-down (roomheater A, $p = 0.066$) and bottom-up ignition technique (roomheater B, $p = 0.254$) was evident. In both cases CO emissions were higher when using beech kindling material. The higher influence of kindling material regarding CO and OGC emissions might result from a lower number of kindling pieces for beech due to the differences in gross density between spruce (softwood) and beech (hardwood). Another influencing factor might be the higher burning rate of softwood compared to hardwood. Further, the chemical composition of spruce, for example a higher content of natural resin, can influence the gaseous emissions as discussed by McDONALD et al. [99] and PETTERSSON et al. [75].

Effect ignition technique

Since the impact of kindling material was only marginal, all test runs applying top-down ignition were compared with the bottom-up ignition test runs. This comparison confirmed the influence of the ignition technique on the combustion quality for each roomheater. The bottom-up ignition led to an improved emission performance for roomheater A. In comparison with top-down ignition, ($p = 0.283$) and 21% lower PM emissions ($p = 0.269$). In contrast, for OGC emissions top-down ignition resulted in an emission reduction of about 21% ($p = 0.181$) (**Figure 16**). Regarding thermal efficiency the bottom-up ignition technique was on average 4.5% higher, indicating a significantly higher thermal efficiency ($p = 0.004$) (**Figure 18**). For roomheater B the top-down ignition technique led to significantly better CO ($p = 0.012$) and OGC ($p = 0.024$) emissions indicated by around 50% lower CO and 65% lower OGC emissions (**Figure 16**). PM emissions showed no significant difference between both ignition techniques ($p = 0.875$) (**Figure 17**). Regarding thermal efficiency the absolute difference was below 1%, but by trend the bottom-up ignition technique reached better results ($p = 0.197$) (**Figure 18**).

For roomheater A batch duration of bottom-up ignition technique was significantly shorter (> 15 min, $p < 0.001$), for roomheater B the same trend was observed, but differences were lower (~3 min, $p = 0.231$) (**Figure 19 – Figure 22**).

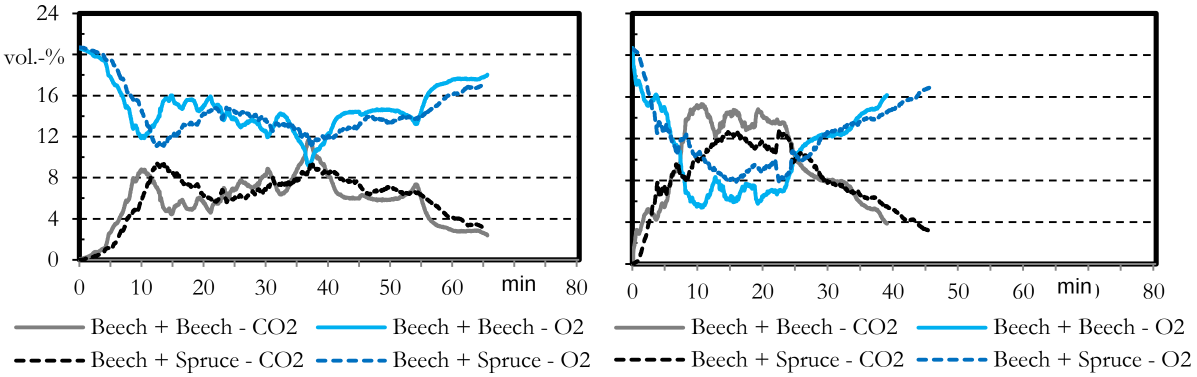


Figure 19: O_2 and CO_2 measurements of ignition tests for top-down (left) and bottom-up ignition technique (right) of roomheater A using different kindling material (Test run 1)

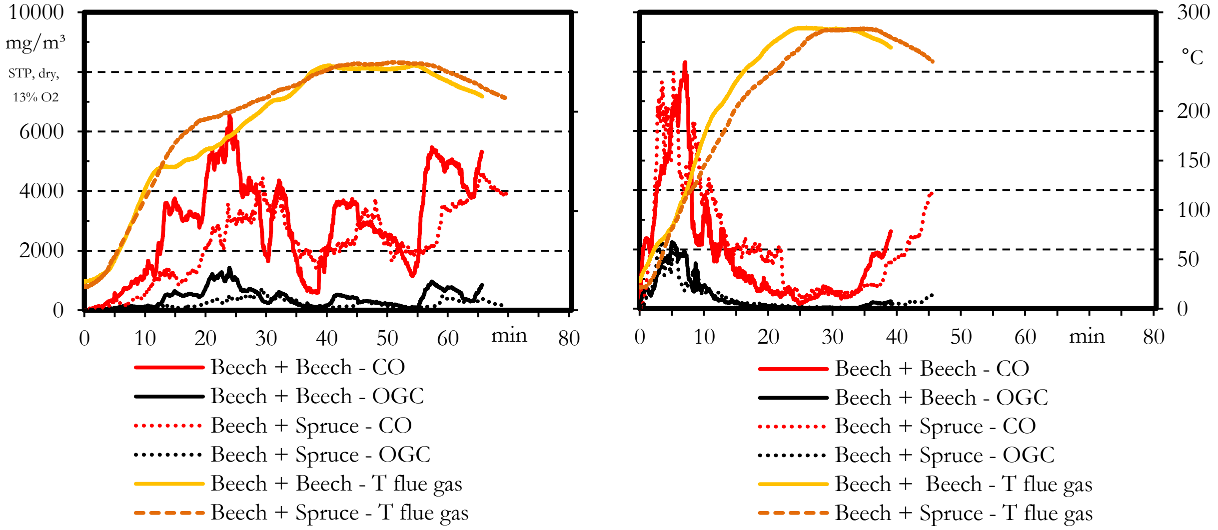


Figure 20: CO, OGC and flue gas temperature measurements of ignition tests for top-down (left) and bottom-up ignition technique (right) of roomheater A using different kindling material (Test run 1)

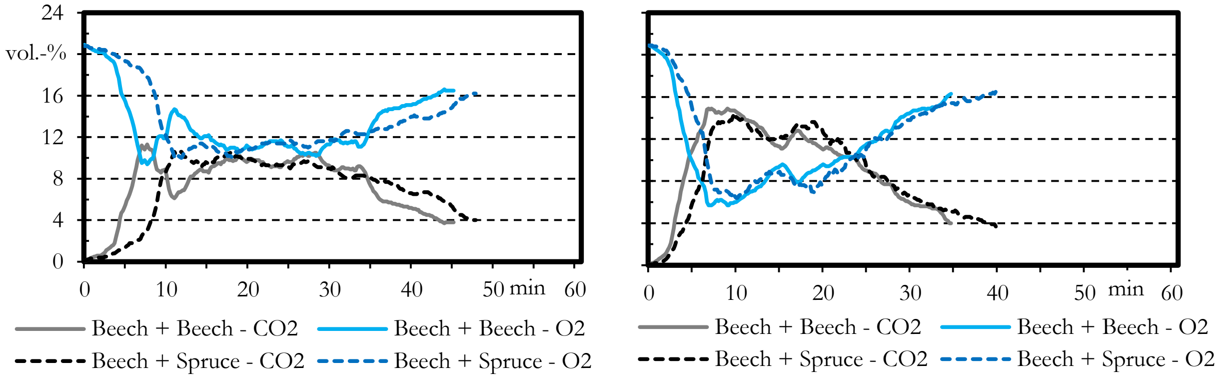


Figure 21: O_2 and CO_2 measurements of ignition tests for top-down (left) and bottom-up ignition technique (right) of roomheater B using different kindling material (Test run 2)

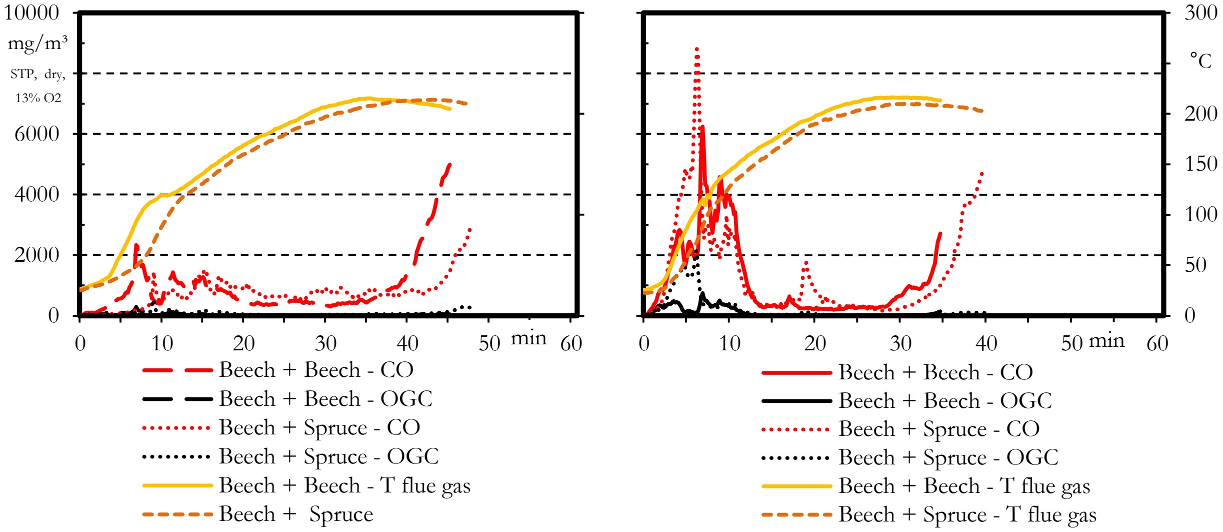


Figure 22: CO, OGC and flue gas temperature measurements of ignition tests for top-down (left) and bottom-up ignition technique (right) of roomheater B using different kindling material (Test run 2)

Comparing the curves of continuously measured gaseous components and flue gas temperatures several characteristics for each ignition technique were found. In general, the increase of the flue gas temperature was faster for the bottom-up ignition technique (**Figure 20**; **Figure 22**). Further, the concentration of the residual flue gas oxygen declined faster. Additionally, lower absolute values compared to top-down ignition technique occurred (**Figure 19**; **Figure 21**). CO and OGC emissions showed a characteristic peak in the start-up phase during the first 10 – 15 min (**Figure 20**; **Figure 22**). The constant course of CO_2 and O_2 concentration of the top-down ignition test runs indicates more stable conditions, especially during the intermediate phase. However, this was not in any case correlated with lower CO, OGC (**Figure 20**; **Figure 21**) or PM emissions (**Figure 17**). For roomheater A the lower batch duration was linked with higher oxygen levels (**Figure 19**) and led to lower flue gas temperatures (**Figure 20**). Consequently, this resulted in higher gaseous emissions as well as in a lower thermal efficiency (**Figure 20**; **Figure 18**, left). Interestingly, the top-down ignition technique for roomheater B resulted in lower CO and OGC emissions during the start-up phase, whereas PM emissions, measured over the whole batch duration, were almost at the same level (**Figure 22**; **Figure 17**).

The 50% CO emission reduction by application of top-down ignition technique for roomheater B is in agreement with the results of HARTMANN et al. [48]. However, the reductive effect of top-down ignition technique on PM emissions, as described by NUSSBAUMER et al. [46] [47] could not be confirmed with those measurements. In general, differences of PM emissions between both ignition techniques were low and statistically not significant.

Overall, test results revealed that top-down ignition might not always be the best solution for each firewood room heating appliance. Furthermore, no correlation was found between the mode of primary air supply and the effect of the ignition technique. A recently published guidance for correct firewood heating [100] is recommending to use the bottom-up ignition technique when primary combustion air is supplied via the grate at the bottom of the combustion chamber (roomheater B). In contrast, the top-down ignition technique should be applied for types of appliances where primary air supply does not enter the combustion chamber via the grate (roomheater A). The results of this work clearly contradict these recommendations. This is also confirmed by the results of VICENTE et al. [90]. In their study comparative ignition tests were carried out using hardwood (beech – "*fagus sylvatica*") and softwood (pine – "*pinus pinaster*") in a wood stove with primary air supply via the grate. Their results showed significantly lower CO and by trend also lower OGC emissions for the bottom-up ignition technique. Regarding PM emissions, the ignition tests conducted with pine firewood led to around four times higher emissions for the bottom-up ignition technique compared to the top-down ignition technique. For beech firewood PM emissions measured during bottom-up ignition test runs were higher by around 15%. Concluding it is evident, that several factors are influencing the emission behavior of the ignition batch. In contrast to the bottom-up ignition technique, the top-down ignition technique led to a slower and more gradual combustion process resulting in longer batch duration and lower thermal efficiency.

3.4.2 Test series II – draught conditions

For all three roomheaters no influence of flue gas draught conditions on PM emissions was found (**Figure 24**, left). Both diagrams of **Figure 23** illustrate that, by trend, increased draught conditions influenced the combustion performance for all three tested roomheaters regarding CO and OGC emissions.

Increased draught conditions of 48 Pa influenced the combustion performance regarding CO emissions for all three roomheaters (A: p = 0.094; C: p = 0.109; D: p = 0.089). The draught conditions influenced the OGC emissions of roomheaters A and C by trend (A: p = 0.172; C: p = 0.130). For roomheater A and C emissions of CO and OGC decreased, whereas for roomheater D an increase of CO and OGC emissions was observed at higher draught conditions. The thermal efficiency was influenced similarly for all combustion tests (**Figure 24**, right). All three roomheaters showed a significant decrease of thermal efficiency at a draught level of 48 Pa compared to a draught level of 12 Pa (A: p = 0.004; C: p = 0.019; D: p = 0.006). Also the increase of draught conditions up to 24 Pa led by trend (D: p = 0.102) or even significantly (A: p = 0.019; C: p = 0.002) to a decreased thermal efficiency performance.

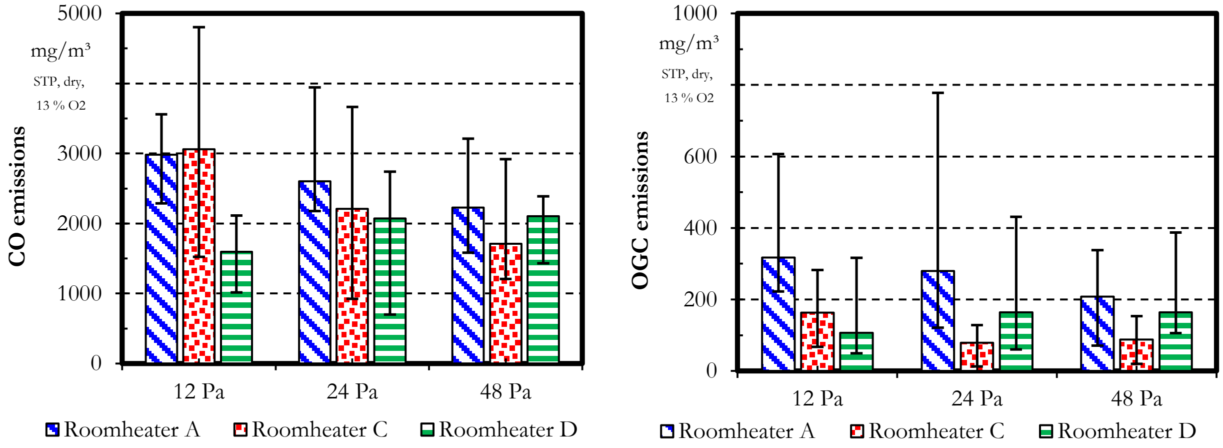


Figure 23: Average results of test cycles at different draught levels regarding CO emissions (left) and OGC emissions (right). The error bars represent the maximum and minimum values of batch results of the test run.

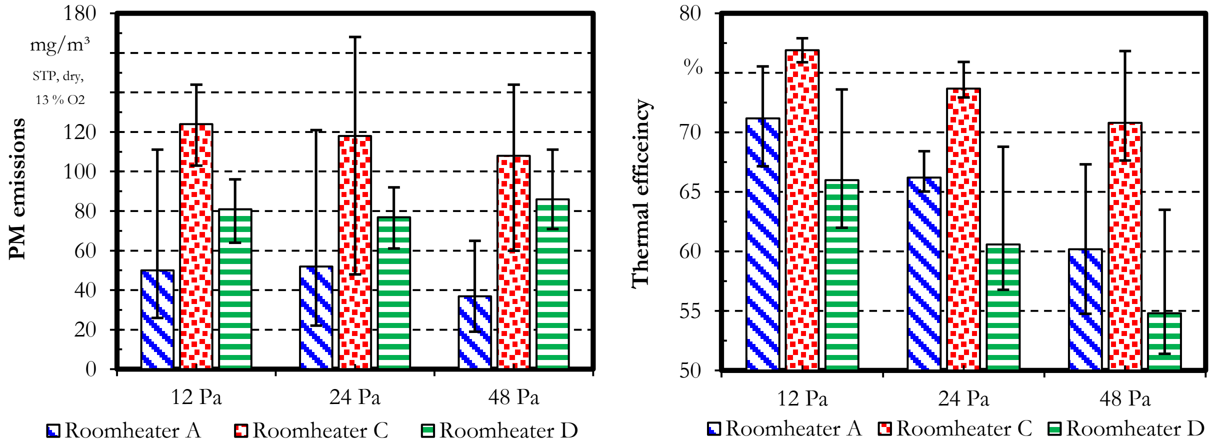


Figure 24: Average results of test cycles at different draught levels regarding PM emissions (left) and thermal efficiency (right). The error bars represent the maximum and minimum values of batch results of the test run.

Due to the calculation method, burn rates are linked with average batch duration. Accordingly, shorter batch durations were accompanied by higher burn rates (**Table 13**). Generally, the decrease of thermal efficiency at higher draught conditions was also evident for single batches. Furthermore, burn rates and flue gas temperatures were increased for batch results at higher draught conditions (see **Table A2**). These findings contradict the results of PRAPAS et al. [101]. According to their findings the burn rate was not influenced by the draught conditions. However, in their study the draught level was varied only in the range of 1 – 7 Pa which is clearly lower compared to the draught conditions of this study. Moreover, draught conditions in real-life are higher and ranged around 15 Pa to 30 Pa [49].

The effect of increased draught conditions on lambda values was similar for roomheater A and D. Both roomheaters showed a significant increase of lambda values between the test cycles of 12 Pa and 48 Pa (A: p = 0.010; D: p = 0.003). In addition, roomheater A and C showed a significant increase of flue gas temperature (A: p = 0.016; D: p = 0.020). The effect of increased lambda values at higher draught conditions was also found by PRAPAS et al. [101]. However, in their study flue gas temperature was decreased by trend at higher draught conditions. In this work the flue gas temperature was either increased (roomheater A and C) or remained on a similar level (roomheater D). This can explain the different findings regarding the effect of draught conditions on burn rates. The higher draught conditions induced a higher amount of combustion air supply as it was indicated by the increased lambda values and also confirmed by PRAPAS et al. [101]. This higher amount of combustion air supply can either accelerate the combustion process (i.e. increase of pyrolysis and gasification rates) or decelerate the combustion process by cooling effects as it was shown by PRAPAS et al. [101].Which effect prevails depends highly on the distribution of the additional combustion air in the combustion chamber. The additional air supply can be divided in three main parts: Primary air, secondary air (e.g. by secondary air inlets and window flushing air) and undefined leakage air (e.g. leaky sealing of combustion chamber door). For roomheater D CO and OGC emissions as well as lambda values were increased whereas flue gas temperature remained on an equal level. Burn rates were decreased at batch 1 at higher draught conditions (see **Table A2**) and varied only little during batch 2 to 5. The average burn rates remained on a similar level (**Table 13**). This indicates that the higher amount of combustion air supply led to a cooling effect which negatively influences CO and OGC emissions.

Table 13: Duration of test cycles (batch 1 – 5) and average results of flue gas temperature, lambda and burn rate

Roomheater	Draught (Pa)	$T_{flue\ gas}$ (°C)	Lambda (-)	Duration (min)	Burn rate (kg/h)
Roomheater A	12 ± 2	271	2.9	348	2.0
	24 ± 2	285	3.4	315	2.2
	48 ± 2	316	3.7	249	2.7
Roomheater C	12± 2	240	5.2	182	2.5
	24 ± 2	273	3.9	152	2.9
	48 ± 2	311	3.9	121	3.7
Roomheater D	12 ± 2	280	3.7	214	2.9
	24 ± 2	278	4.4	208	2.9
	48 ± 2	282	5.1	194	3.1

The effect of increased draught conditions on combustion quality on batch-wise operated firewood stoves was also studied by LENZ et al. [51]. In that study an inset appliance and a firewood roomheater were tested under different draught conditions between 12 Pa and 34 Pa. For the inset appliance a slow increase of CO, OGC and PM emissions was found. Regarding the firewood roomheater also no effect of increased draught conditions on PM emissions was found. Furthermore, CO and OGC emissions did not correlate with the draught condition variations. However, thermal efficiency correlated clearly with varied draught conditions. For both firewood room heating appliances a significant decrease of thermal efficiency was found at increased draught conditions. In addition, field measurements at three different roomheaters under natural draught conditions confirmed the major impact of draught level on thermal efficiency [49].

Based on these findings it was confirmed that the main effect of increased draught conditions on real-life operation performance of firewood roomheaters is a reduced thermal efficiency performance. This leads to an increase of firewood or fuel demand. Consequently, there is an increasing PM emission freight, since more fuel has to be combusted for covering a certain thermal heat demand.

3.4.3 Suitable measures to adapt the stove to different draught conditions

As previously discussed, a higher amount of combustion air supply induced by higher draught conditions can either accelerate the combustion process (i.e. increase of pyrolysis and gasification rates) or decelerate the combustion process by cooling effects. The respective effect can be seen by analyzing the burn rate in correlation to the flue gas temperature and lambda value. As shown in the previous section this has a direct impact on the thermal efficiency (**Table 13**, **Figure 24**, left). The effect of a higher amount of combustion air supply induced by higher draught conditions on gaseous emission concentrations depends on the appliance specifics. Therefore, gaseous emissions (CO/ OGC) could either increase or decrease (**Figure 23**). For PM emission concentrations no impact was observed (**Figure 24**, left). But, as mentioned above, a reduced thermal efficiency results in higher fuel consumption. Consequently, there is an impact on total emissions even if the emission concentrations are on a similar level.

In general, the thermal efficiency depends mainly on the flue gas temperature and the lambda value which directly influence the thermal flue gas losses when calculated according to the indirect determination approach of the EN standard (**Eq. 2 – Eq. 4**). The chemical flue gas losses and the losses due to the combustibles in the residues are clearly lower (**Figure 25**).

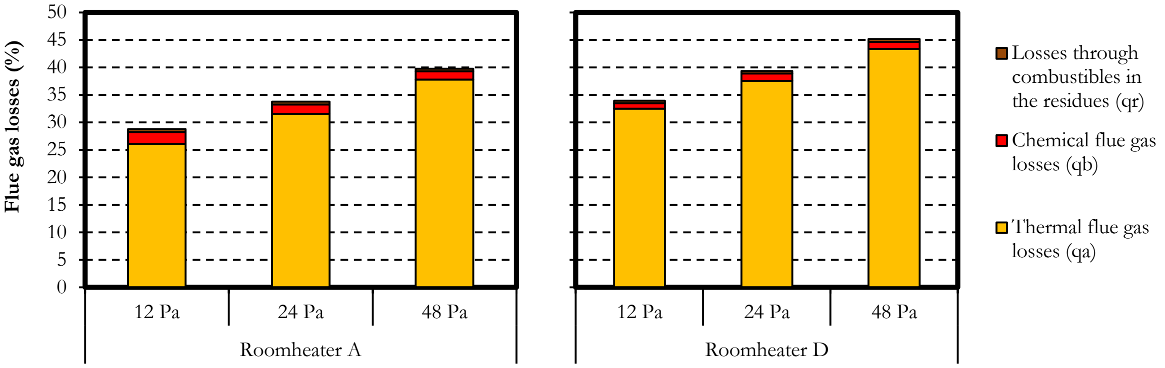


Figure 25: Proportions of flue gas losses for the for the different test cycles (12 Pa – 24 Pa – 48 Pa) calculated according to the standard EN 13240

If the flue gas temperature increase and the lambda value remain quite stable (or vary comparatively slightly) the thermal flue gas losses increase and therefore the thermal efficiency decreases. For example this is illustrated for roomheater A within to the draught tests (**Figure 24**, right, **Figure 25**, left, **Table 13**). If the flue gas temperature is quite stable, but the lambda value increases clearly, the thermal flue gas losses increase as well and consequently thermal efficiency decreases. This can be exemplarily seen for roomheater D within to the draught test series (**Figure 24**, right, **Figure 25**, left, **Table 13**).

Based on the correlations of flue gas temperature, lambda value and thermal efficiency it is obvious that the absolute amount of combustion air supply has to be adapted appropriately for the appliance and the respective load settings. In general, there are two different possibilities to achieve this:

- Adapt air valve settings for combustion air supply either manually or by suitable automatic control systems
- Adapt flue gas draught conditions using external draught control devices

1) Adapt air valve settings for combustion air supply

At higher draught conditions (e.g. > 30 Pa) the air valve settings have to be more closed, e.g. compared to draught conditions of 12 Pa as applied during EN testing. However, since the operating procedure of the manual is usually defined according to the draught conditions required by the EN standard (12 ±2 Pa) there is the risk, that the air valve settings under typical real-life draught conditions (**Figure 12**, **Figure 13**) are inappropriate. However, there are several appliances or retrofit devices on the market that enable an automatic adaption of the combustion air supply by regulating the air valve settings. These control systems are either designed as open-loop or closed-loop control systems.

For firewood stoves, open-loop control systems typically adapt the air valve settings according to specific criteria in a predetermined manner. For example, the air valve settings are adapted (continuously between closed or opened) according to the temperature gradient measured by a temperature sensor (e.g. located in the combustion chamber or at the flue outlet). Such control systems are either implemented in the serial-production stoves, e.g. FOX II RIKATRONIC3[5] of the Austrian stove manufacturer Rika, or are available as retrofit applications which are installed at the central pipe for combustion air supply (**Table 14**/ **Figure 26**). Retrofit applications have to be calibrated for each appliance specifically by pretests [102]. However, the actual combustion conditions give no permanent feedback towards the open-loop control systems.

In contrast, closed-loop control systems adapt the air valve settings according to an O_2 and temperature sensor which are placed in the flue gas path (e.g. flue outlet of the post combustion chamber). The regulation system adapts the air valves specifically in order to achieve a stable residual O_2 level and flue gas temperatures during heating operation. Consequently, the actual combustion conditions induce a permanent feedback towards the closed-loop control system. **Figure 26** presents an example of such an "intelligent heating system (IHS)" integrated and implemented in a firewood roomheater as well as a retrofit control system for combustion air supply (TATAREK RT-08 OS).

Figure 26: Retrofit open-loop control system for combustion air supply (TATAREK RT-08 OS) (left) [103] and closed-loop combustion air supply control system integrated in a firewood stove (right) [104]

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[5] http://www.rika.at/de/fox-II-rikatronic3/ (accessed 26/11/2018)

2) External draught control devices

As second possibility, the amount of combustion air supply can be adapted by external draught control devices. One type of external draught control devices adapts the draught conditions by a controlled dilution of the flue gas. When draught conditions are too high a damper of the draught control device opens and dilution air form outside of the heating system enters the chimney. The principle of such an external draught control device is illustrated in **Figure 27**.

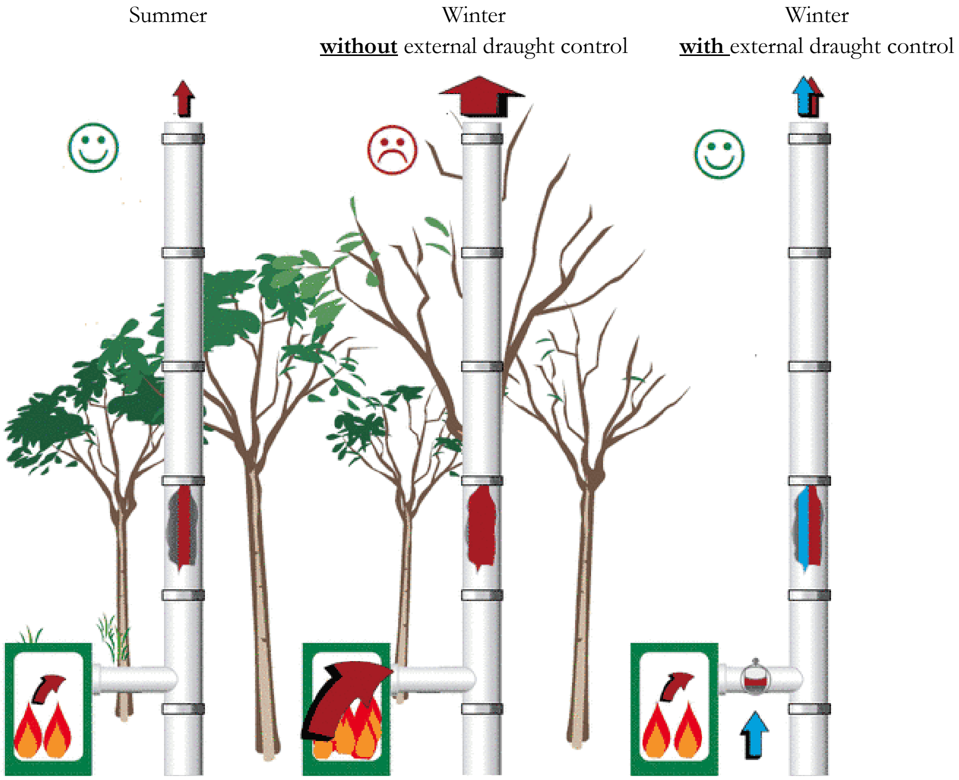

Figure 27: Operating conditions of a stove at summer and winter seasons referring to draught conditions and functionality of external control system [105] (adapted & translated in English)

During warm temperature conditions of around 15 °C, e.g. at spring or autumn, the chimney system induces optimal draught conditions. The stove can be operated at optimal load settings and the thermal heat losses are comparatively low (**Figure 27**, left). However, in the winter season at cold temperature conditions and windy weather conditions the chimney could induce exorbitant draught conditions which results in higher burn rates and increased thermal flue gas losses (**Figure 27**, middle). Using an external draught control device the flue gas is diluted by ambient air and the flue gas draught could be reduced. Therefore, thermal heat losses should be reduced and a high thermal efficiency can be achieved (**Figure 27**, right). However, it should be considered, that the external draught control device results in thermal heat losses of the installation room during operation, if it is installed in the installation room, e.g. in the connecting flue gas pipe of the chimney. Furthermore, this could also increase standby heat losses, for example during windy weather conditions [102].

Therefore, external draught control devices which dilute the flue gases should use ambient air from outside the heated room or building as illustrated in **Figure 28**.

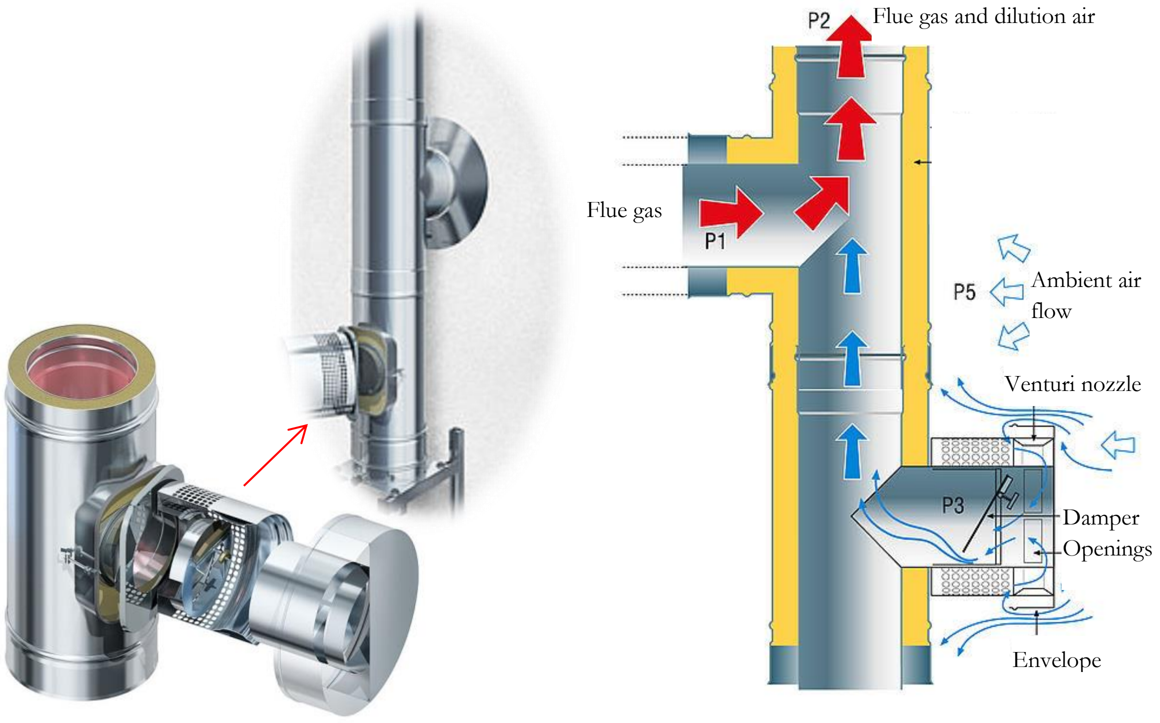

Figure 28: External draught control device, ZUK 130 DW (left), and its functionality (right) of the company Kutzner + Weber (K+W) [106] (translated in English)

There are also external draught control devices which vary only the open cross-sectional diameter of the flue gas pipe. One example represents the FLORIAN device from the company "fumis by ATeech electronics" (**Figure 29**).

The FLORIAN adapts the damper settings either manually according to different stages or automatically according to a self-learning regulation approach (fuzzy-logic regulation) on the basis of the flue gas temperature conditions. Additionally, the FLORIN has also an integrated ventilator in order to increase flue gas draught conditions if it is needed, e.g. during ignition of the first fuel batch [107] [102].

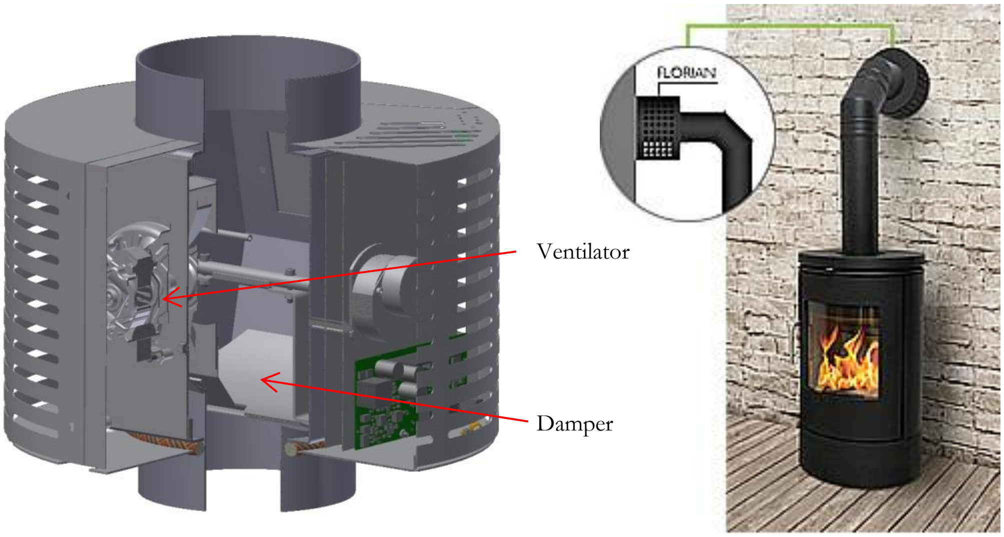

Figure 29: External draught control device, FLORIAN, of the company “fumis by ATech electronics” [107]

In MACK et al. [102] an overview of three retrofit devices for regulation of combustion air supply and two different external draught control devices are presented (**Table 14**).

Table 14: Overview of evaluated control systems for combustion air supply and external draught control devices [102] (translated in English)

Testing conditions	**Controlled draught conditions**			**Natural draught conditions**	
Type	**TATAREK RT-08 OS**	**SCHMID SMR**	**K+W compact**	**FLORIAN (ATech electronics)**	**K+W external draught control device (ZUK 130 DW)**
Functional principle	Thermocouple + electronic damper			Thermocouple + pressure and temperature sensor + electronic damper + ventilator	Mechanical damper
Position of installation	Central pipe of combustion air supply			Connecting flue gas pipe between chimney and stove	Connecting flue gas pipe between chimney and stove or at the bottom of the chimney
Open diameter during standby	6.2 cm^2 (8%)	tight	9 cm^2 (8%)	-	-
Approximate price for end-customers	276 €	1,100 €	1,070 € (without display)	300 €	300 €

The systems were evaluated under real-life operating conditions in the laboratory by using the "*beReal*" test cycle [102] (see chapter 7, **Figure 70**). Therefore, the test cycle consisted of eight consecutive batches and includes ignition, preheating, nominal and part load as well as considers the cooling down phase for the efficiency determination. The control systems for combustion air supply regulation were evaluated under controlled draught conditions (12 ± 2 Pa), whereas the external draught control devices were tested at a specific test facility connected to a natural draught chimney. All tests ("*beReal*" heating cycle) were repeated twice (n=3). The operation mode was carried out according to Quick-User-Guides which were supplied by the manufacturers [102]. An example of a Quick-User-Guide is presented in **Figure 9** (chapter 2.4.3).

Figure 30 to **Figure 33** illustrate the results of the respective combustion tests regarding gaseous and particulate emissions, average batch durations as well as thermal efficiency results according to experimental evaluations of MACK et al. [102].

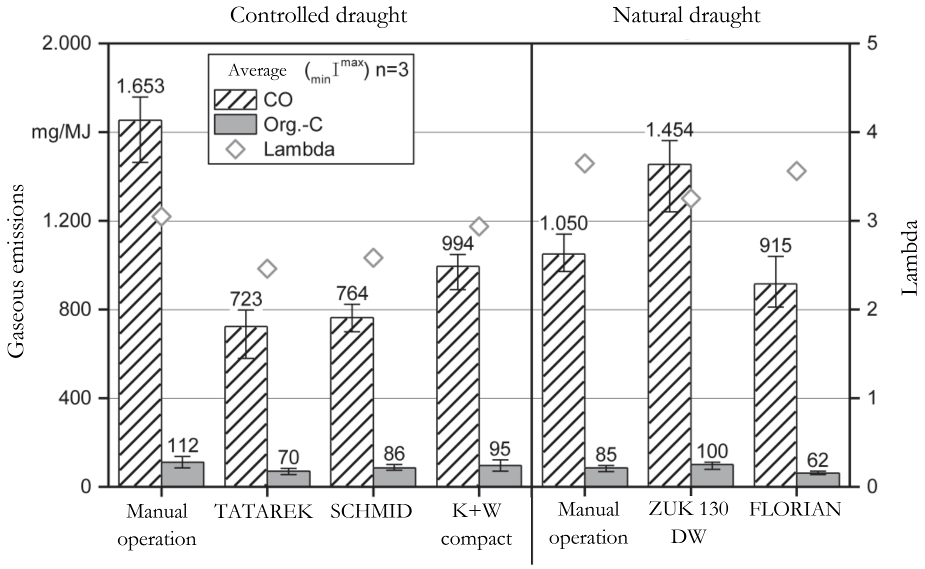

Figure 30: Average gaseous emissions and lambda values of heating operation using different applications of combustion air supply control systems and external draught control devices compared to manual heating operation [102] (translated in English)

The gaseous emissions were lower compared to the reference of manual heating operation for all tested retrofit applications for combustion air supply regulation (**Figure 30**). For the TATAREK RT-08 OS system, CO emissions were reduced by 56% and OGC emissions by 38% which represents the best results of the tested systems. All tested systems are characterized by lower lambda values during heating operation compared to the manual heating operation with constant air settings for ignition, nominal load and part load.

For the combustion tests conducted under natural draught conditions an emission reduction effect was only observed for the FLORIAN system. The reductive effect was around 13% for CO emissions and 27% for OGC emissions. For CO emissions the reductive effect was clearly lower compared to the evaluated combustion air control systems. For the second evaluated external draught control device (ZUK 130 DW) the gaseous emissions were even higher compared to the manual heating operation. According to Mack et al. [102] it is assumed that the FLORIAN which was installed in the connecting flue gas pipe (**Figure 29**) induced further turbulences with a positive effect on emissions. This was not the case for the ZUK 130 DW which was installed at the bottom of the natural draught chimney system (see **Figure 28**).

Interestingly, gaseous emissions of the same stove were lower when operated manually under natural draught conditions compared to the manual heating operation under constantly controlled draught conditions (**Figure 30**). The higher lambda values of the heating cycle conducted under natural draught conditions indicate that the average natural draught conditions were higher than 12 Pa. Therefore, these findings are in line with the results of draught tests presented in chapter 3.4.2.

The recharging of a new fuel batch was indicated by the combustion air control systems and resulted in all cases in a shorter batch duration compared to the manual heating operation (**Figure 31**).

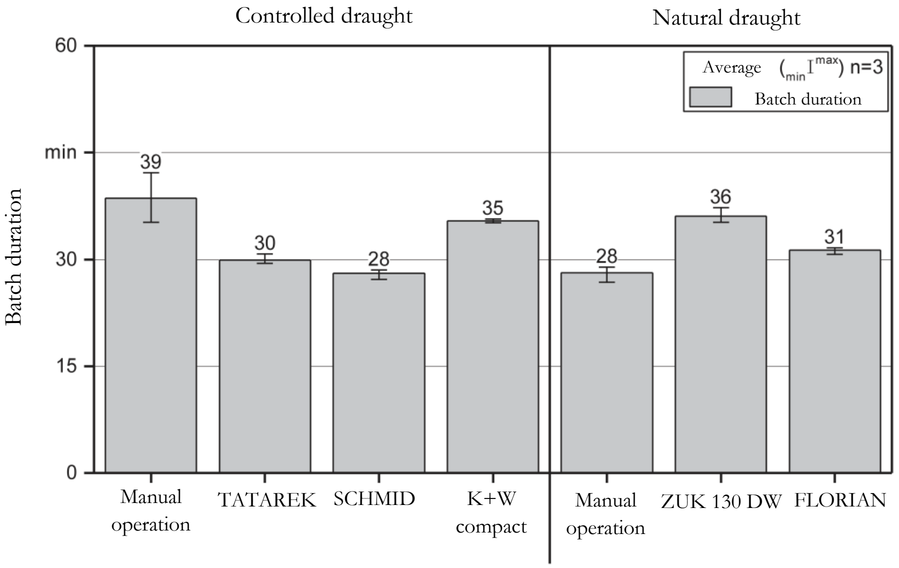

Figure 31: Average batch duration of heating operation using different applications of combustion air supply control systems and external draught control devices compared to manual heating operation [102] (translated in English)

The shorter batch durations resulted in a less share of burn-out phase emissions and higher temperatures at the beginning of the following fuel batch. Additionally, the combustion air supply control systems reduced the combustion air supply, especially during the start-up phase of the batch. Both aspects were identified as the most relevant reasons for the positive effect of the combustion air supply control systems [102]. However, it has to be considered that all systems were calibrated according to measurements of pretest by experts. It is also critically mentioned in the report of MACK et al. [102] that optimal calibration is hardly possible for an end-customer only by visual inspection.

In contrast to the combustion air supply control systems, the external draught control devices led to longer batch durations (**Figure 31**). Consequently, the heating operation using the combustion air control systems led to higher burn rates and thermal heat outputs whereas the external draught control devices led to reduced burn rates and thermal heat outputs compared to the manual heating operation, respectively.

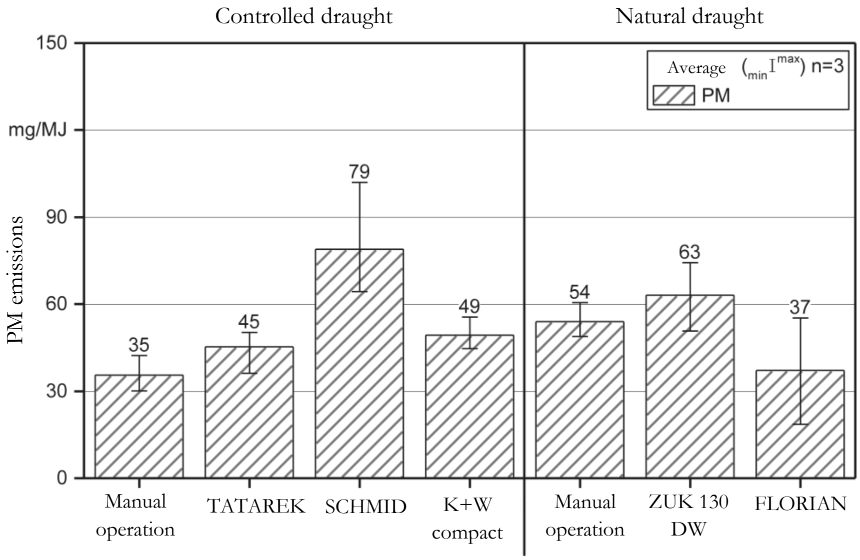

Figure 32: Average PM emissions of heating operation using different applications of combustion air supply control systems and external draught control devices compared to manual heating operation [102] (translated in English)

The particulate emissions increased for the combustion tests with the applied combustion air control systems (**Figure 32**). As mentioned in MACK et al. [102] the reasons for that were most probably the shorter batch durations and therefore the higher effect of particulate emissions of the start-up phase on total PM emissions. This is because most of particulate emissions are released during the start-up and the first part of the intermediate phase of a firewood batch whereas the PM emissions during the burn-out phases are comparatively low [46]. Consequently, the sampled flue gas volume of gravimetric PM measurements are less for shorter batch durations and the impact of the start-up and intermediate phases on total PM emission concentrations are higher. But considering the range of measurements, the PM emissions were on a similar level, at least for the systems the TATAREK RT-08 OS and K+W compact.

The effect of the external draught control devices was contrary. The combustion tests with the FLORIAN system led to lower PM emissions whereas the combustion tests with the ZUK 130 DW led to slightly higher PM emissions (**Figure 32**). However, the range of PM emission results was similar and indicates comparatively low differences. This is in line with the results of draught tests presented in chapter 3.4.2 where no significant effect of changed draught conditions on PM emission concentrations was observed (**Figure 24**, left). However, the results found for the FLORIAN device indicate that this device does not only lowering the flue gas draught, but might also influence the primary combustion conditions as assumed by MACK et al. [102].

The effect of the different evaluated automatic combustion air supply control systems (retrofit application) and the external control devices on thermal efficiency is presented in **Figure 33**. The air valve settings remained on the respective settings at the end of heating operation (after the eighth batch). This is indicated in the specifications of the x-axis of **Figure 33** as "last position" and corresponds to typical real-life use of end-customers [102]. This was also confirmed by the findings of the survey (chapter 2.4, **Table 8** "Question 4.8").

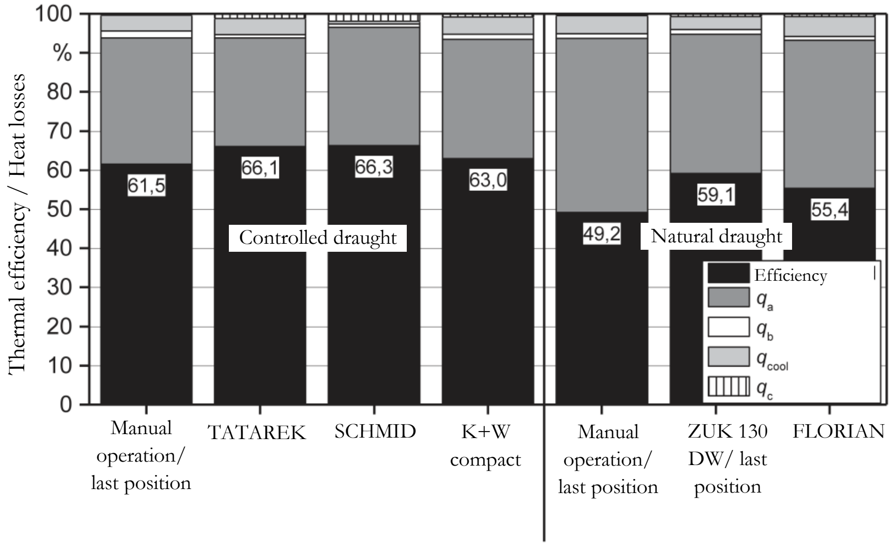

Figure 33: Thermal efficiency and different losses of heating operation using different applications of combustion air supply control systems and external draught control devices compared to manual heating operation [102] (translated in English)

The thermal efficiency of combustion tests increased when both, the combustion air supply control systems and the external draught control devices, were used. For the combustion air control systems the increase of thermal efficiency ranged between 1.5% (K+W compact) and 4.8% (SCHMID SMR) absolutely (**Figure 33**, left).

The increase of thermal efficiency was higher for the external draught control devices and ranged between 6.2% (FLORIAN) and 9.9% (ZUK 130 DW) absolutely (**Figure 33**, right). However, due to the different draught levels the thermal efficiencies under natural draught conditions were lower compared to the tests performed under constantly controlled draught conditions. In all cases the thermal heat losses of the flue gas represented the highest losses. Improvements of thermal efficiency were predominantly gained by lowering thermal heat losses of the flue gas. This is in line compared to results of the draught test series and the descriptions about general correlations of flue gas temperature, lambda value and thermal efficiency which were mentioned at the beginning of this section (**Figure 25**).

In MACK et al. [102] retrofit applications, external draught control devices and retrofitted open-loop combustion air supply control systems (**Figure 30** – **Figure 33**), are evaluated whereas in ILLERUP et al. [104] a stove with an integrated automatic combustion air supply control system (closed-loop) is presented (**Figure 26**). In **Figure 34** the curves of flue gas temperature, O_2, CO and thermal efficiency from that stove are presented. Thereby, the principle of such a closed-loop control concept is illustrated.

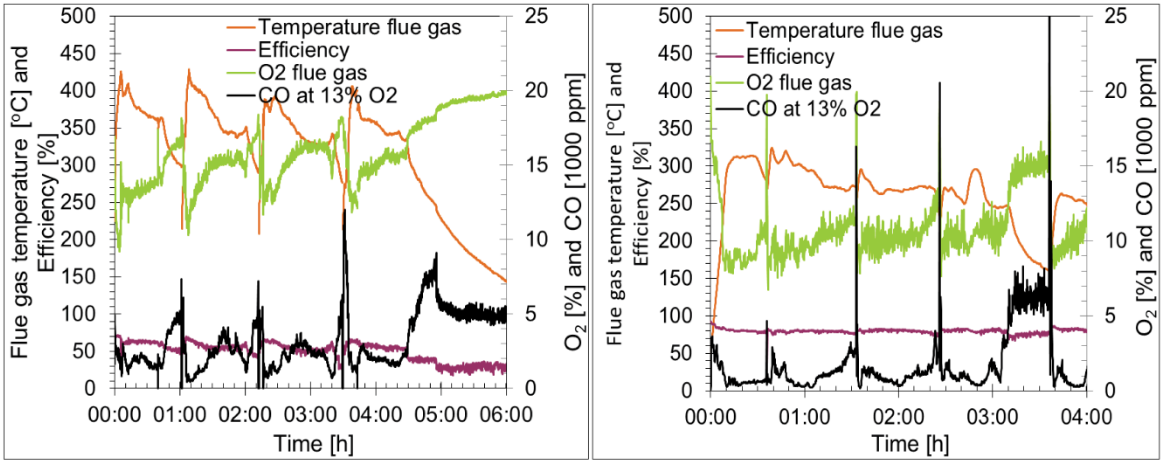

Figure 34: Curves of flue gas temperature, O_2, CO and thermal efficiency during a batch without and with closed-loop control of O_2 and flue gas temperature referring to the heating operation of the same user [104]

The stove was operated by the same user without and with the closed-loop combustion air supply control system. As already described, closed-loop control systems react permanently towards the actual combustion conditions based on the sensor signals of flue gas temperature and O_2. The air valve settings are adapted in order to achieve stable flue gas temperature conditions and a low residual O_2 concentration. As illustrated in **Figure 34** the flue gas temperature was reduced by around 100 °C from 380 °C to around 280 °C and the residual O_2 content by around 5 vol.-% from about 15 vol.-% to 10 vol.-% when using the closed-loop combustion air supply control system. Thereby, the efficiency was increased by more than 10% to around 80% absolutely during the whole heating operation of four batches (**Figure 34**). Furthermore, CO emission concentrations decreased, especially during the intermediate phases of the respective batches.

Concluding, the previously mentioned systems, external draught control devices and combustion air supply control systems, aim to improve the combustion conditions by influencing the amount of combustion air supply. Due to high draught conditions or other user related aspects an inappropriate amount of combustion air supply can be available. The evaluated systems are suitable to increase thermal efficiency and additionally have the potential to decrease emissions. However, in some cases emissions were increased using those some systems compared to the manual heating operation. However, it has to be considered that the manual heating operations which represent the reference cases according to [102] refer to best-practice user behavior which is not always the case in real-life operation at the end-users. Therefore, the mentioned control systems can be recommended as potential technological measures to improve real-life heating operation. Especially closed-loop control systems which adapt air valve settings during the combustion phases to a certain set-point (e.g. O_2 value) reveal a suitable and effective solution to reduce emissions as well as to increase thermal efficiency significantly. Retrofit applications which adapt the amount of combustion air supply with an open-loop control approach have to be calibrated for each appliance specifically. This should be carried out by experts based on measurement data of pretests on-site. Furthermore, it should be considered that all retrofit combustion air control systems represent a significant change of the combustion appliance. Thus, it loses its CE-mark. Consequently, each installation of a retrofit combustion air supply control system needs obligatorily an approval of the chimney sweeper [102].

External draught control devices increase thermal efficiency and can reduce emissions. However, their general benefit towards lower emission concentrations is still not clear and there might be even increased emissions due to the installation of external draught control devices. As recommended by MACK et al. [102] the installation of external draught control devices should be clarified together with the manufacturer of the appliance.

3.5 Summary – Effect of ignition technique and draught conditions on emissions and thermal efficiency

Following the outcomes of the user survey, the effect of the ignition technique as well as draught conditions on emissions and efficiency was clarified by experimental combustion tests. Those tests confirmed that draught conditions as well as the ignition technique (top-down compared to bottom-up) can significantly influence the emission and efficiency performance of firewood roomheaters.

The ignition technique in general affected the level of CO, OGC and PM emissions during the ignition batch of a firewood roomheater. However, the comparison of both ignition techniques showed no general advantage of the top-down ignition technique as it is often claimed by leaflets for correct heating. The high reductive effect of top-down ignition on PM emissions, as reported by literature, was not observed for the tested roomheaters. A potential link between the design of primary air supply via the grate and the better performance of the top-down ignition technique was also not confirmed. The impact of using beech or spruce as kindling material indicated only marginal or rather no influence on the combustion performance of the ignition batch independently of the applied ignition technique. The bottom-up ignition technique was found to be faster compared to the top-down ignition technique and even most advantageous concerning thermal efficiency performance. Consequently, the general application of the top-down ignition technique as best-practice operation cannot be reasoned. The best-practice ignition technique shall be specified for each appliance in the manual or in a Quick-User-Guide, as already proposed in chapter 2.

The comparative combustion tests at different draught conditions of 12 Pa, 24 Pa and 48 Pa showed that different appliances are influenced either positively (30% – 60%) or negatively (13% – 5%) regarding gaseous emissions at higher draught conditions. However, at draught levels of around 15 Pa – 30 Pa, which are common in real-life operation, the impact of draught conditions on gaseous emissions is negligible. Additionally, the results showed no direct effect of draught conditions on PM emissions for all three types of used firewood roomheaters. A significant correlation between increased draught conditions and lower thermal efficiency performance was observed for all tested roomheaters.

Concluding, the main effect of draught conditions refers to the thermal efficiency and the burn rate. The higher the draught conditions, the higher are burn rates and thermal heat losses at the same air valve settings. Since stoves are tested at 12 ± 2 Pa and manufacturers specify the heating operation in accordance to this level the risk of low efficiency performance at exorbitant draught conditions increases significantly, even when the air valve settings are consistent with the manual.

The applicability of retrofitted or integrated combustion air supply control systems and external draught control devices represent suitable technological measures to adapt the stove towards different draught conditions. Those systems were also identified as suitable measures to decrease gaseous as well as particulate emissions and increase thermal efficiency. However, since the stoves react differently towards higher draught conditions those systems, especially external draught control devices, do not reduce emissions in every case, especially compared to best-practice heating operation. Furthermore, the systems need a proper installation and calibration specifically for each appliance. Therefore, the installation of retrofit combustion air supply control systems as well as external draught control devices should be done by experts and the installation should be clarified together with the manufacturer of the respective stove. Finally, the installations need the approval of the chimney sweeper.

4 APPLICABILITY OF OXIDIZING HONEYCOMB CATALYSTS – CATALYTIC EFFICIENCY[6]

In chapter 2 the user survey illustrated that common end-user heating operation has potential for improvement regarding some aspects of heating operation (e.g. air valve settings during operation and at the end of heating operation, optimal ignition technique, etc.). Furthermore, operating conditions referring to the installation conditions of the appliance could significantly influence the combustion conditions. As illustrated in chapter 3 an unsuitable ignition technique or increased draught conditions could increase emissions as well as lowering thermal efficiency. Based on the outcomes of the survey external training arrangements and short guidelines (Quick-User-Guides) specifying correct and appliance specific best-practice heating operation were recommended as suitable non-technological measures to optimize the user behavior and improve the emission and thermal efficiency performance of appliances during real-life heating operation. The level of draught conditions depends to the most extent on the chimney design and weather conditions and is only less influenced by the operating habits of end-users. As discussed in the previous chapter (chapter 3.4.3), retrofitting of integrated combustion air supply control systems as well as external draught control devices are suitable technological measures to adapt the stove towards different draught conditions. Thereby, thermal efficiency performance could be increased and potentially emissions decreased. However, the systems have to be designed and adapted properly and specifically for the respective appliance. A closed-loop control system for combustion air supply in combination with a state-of-the-art appliance (primary optimized with an appropriate air staging concept) is regarded as a promising technology towards low emission and high thermal efficiency performance during real-life heating operation. For those appliances all technological measures could be designed already during the production process. Moreover, the influence of users during heating operation can be clearly reduced by installing a closed-loop control system. However, as shown in **Figure 34**, even such technologies have combustion conditions which are characterized by increased emission concentrations, especially during start-up and burn-out phases. Additionally, appliances which were evaluated in combination with a retrofitted combustion air supply control system or an external draught control device showed high emission concentrations (**Figure**

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[6] Segments of this section have already been published in [4].

30 – **Figure 33**). Especially compared to automatic combustion systems, e.g. pellet stoves or pellet boilers, the emissions of firewood stoves are clearly higher and therefore need further reduction.

Consequently, measures are needed in order to decrease gaseous and particulate emissions. To the best, those measures should support primary optimization, offer a high availability and work properly under all typical operating phases of batch-wise firewood combustion.

The utilization of integrated catalysts seems to be a suitable solution for stove manufacturers to decrease emissions significantly [108]. As already mentioned (chapter 1.1), in the United States or Canada integrated catalysts are commonly used. However, in Europe, stoves equipped with catalytic systems are rare, since the focus on technological development was set on the optimization of primary combustion conditions [52] [54]. However, research and development is ongoing to combine primary and secondary measures in order to use the synergies of both optimization approaches. Catalysts are featured with specific characteristics which make them more advantageous for the applicability in firewood stoves compared to other secondary abatement systems, like fabric filters or electrostatic precipitators (ESP). Catalysts are known to reduce both, gaseous and particulate emissions. Furthermore, compared to fabric filters they have a lower pressure drop which is important for a safe operation of stoves without a ventilator under natural draught conditions. Furthermore, catalysts do not need electrical current, like ESP devices.

Therefore, this thesis focuses on the applicability of oxidizing honeycomb catalysts which are already commercially available. The novel approach is the integrated application of catalytic converters in the post combustion chamber of the stoves without any bypass. Up to now, there are no serial-production stoves of European manufacturers in such a configuration on the European market. Before the presentation of the experimental combustion tests a short overview of the functionality and relevant characteristics of catalytic systems is provided.

4.1 State of the art

4.1.1 Catalytic principle

Many chemical reactions proceed only slowly or at high temperature levels. A certain amount of energy, the so called activation energy (E_a), is necessary to enhance the respective reactions. However, the utilization of catalysts reduces this activation energy (E_a) (**Figure 35**) [109]. Therefore, a catalyst enables chemical reactions which would normally proceed only at higher temperatures with a higher energy input. In addition, catalysts accelerate the chemical reaction processes and enhance the selectivity of the specific reactions in order to achieve the favored products. However, the catalyst is not consumed itself by the reactions and the thermodynamic of the overall chemical reaction is not changed. This means that the catalyst itself does not appear in the overall stoichiometry of the reaction and the net change of the reaction energy (ΔG) is equally with and without the presence of the catalyst (**Figure 35**) [109].

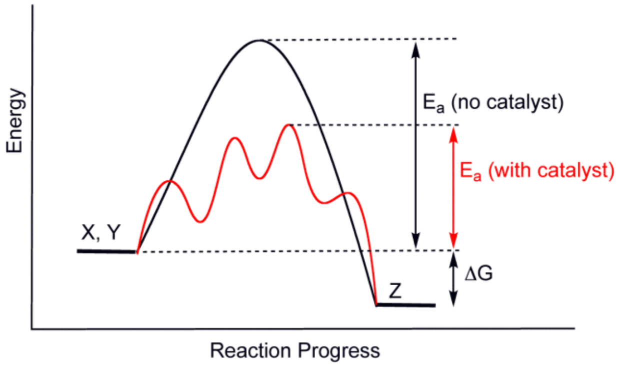


Figure 35: Generic potential energy diagram illustrating the principal of a hypothetical exothermic chemical reaction ($X + Y \rightarrow Z$) with (red) and without (black) the presence of a catalyst [110]

As illustrated in **Figure 35**, reactants or educts (X, Y) and products (Z) of the overall chemical reaction are the same with and without the impact of the catalyst. However, the elementary reactions of the overall reaction are differently under the presence of a catalyst since the lowering of the activation energy (E_a) opens alternative pathways for the chemical reactions.

4.1.2 Design of honeycomb catalysts

The use of honeycomb catalysts for oxidation of gaseous components of the flue gas is defined as heterogeneous catalysis. This means that gaseous educts react with a solid catalyst whereas homogeneous catalysis refers to the catalytic conversion of components consisting of the same chemical phase, e.g. liquid components with a liquid catalyst [109]. Honeycomb catalysts are typically used in the automotive industry to convert CO, OGC and NO_x emissions [111] [112].

Honeycomb catalysts consist of three main components:

- Carrier material
- Intermediate layer
- Catalytic layer

The carrier material forms the honeycomb geometry of the catalyst. The catalytic layer consists of a highly porous material, e.g. aluminum oxides (Al_2O_3), in order to increase the internal surface of the catalyst enormously compared to the sole geometric surface of the carrier material. The catalytic layer consists of the catalytically active components, e.g. platinum (Pt) and palladium (Pd). The intermediate layer is specified as "washcoat" of the catalyst whereas the catalytic layer are specified as "active phase" or "active sites" of the catalyst [113] (**Figure 36**).

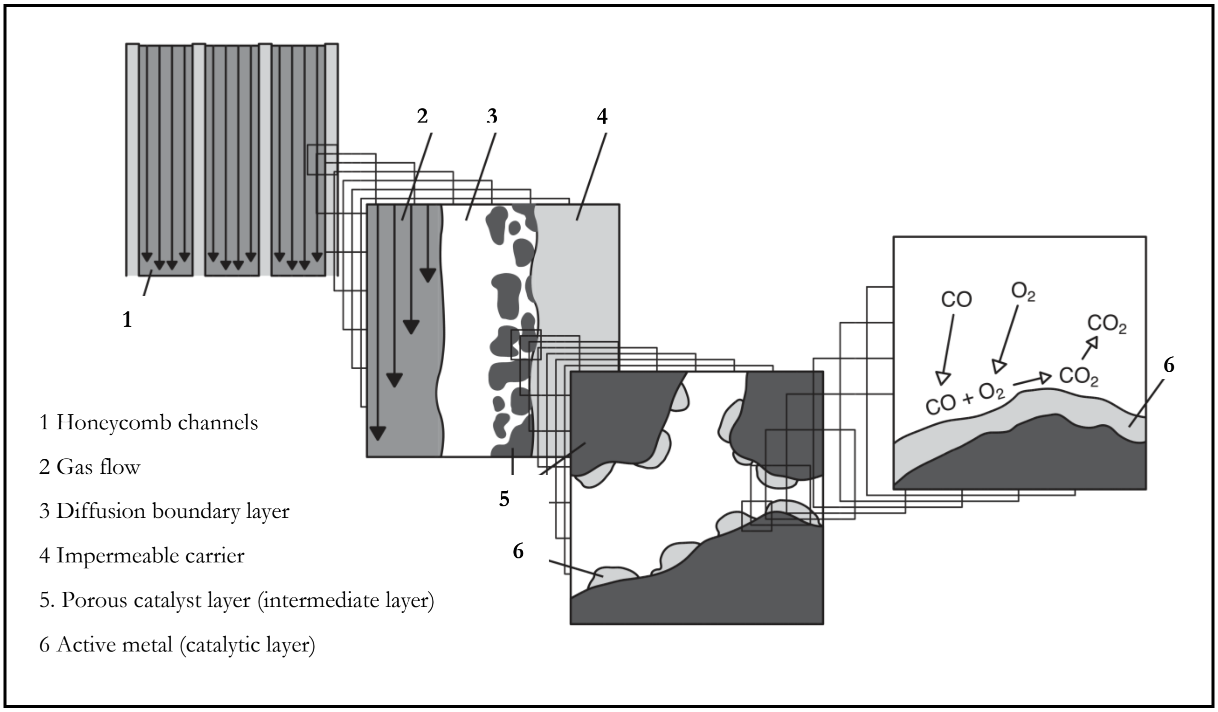


Figure 36: Characteristic design and reaction mechanisms of a honeycomb catalyst [113]

The chemical reactions referring to the catalytic conversion proceed at the active phase of the catalyst, i.e. at the active metals. For example, CO emissions are oxidized under the presence of O_2 to CO_2 with a Pt/ Pd catalyst (**Figure 36**).

For the constructive design of a catalyst or a catalytic reactor, including the carrier material, intermediate layer and the catalytic layer, the space velocity represents an important characteristic number. For honeycomb catalysts used in a gaseous fluid (e.g. flue gas) the space velocity is defined as the ratio of the volume flow (in m^3/h, wet conditions) of the flue gas and the volume of the catalyst (in m^3). Therefore, the unit is 1/h which is commonly used in practice [113]. The space velocity represents the reciprocal value of the residence time of the gases within the catalyst. Hence, a high space velocity indicates a low residence time of flue gases within the catalyst, whereas a high residence time of flue gases within the catalyst is indicated by a low space velocity number.

4.1.3 Catalytic reaction mechanisms

In general, the heterogeneous catalytic conversion at the honeycomb catalyst can be categorized in seven characteristic phases (**Figure 37**):

1. Flow of flue gas through the catalyst channels and diffusion of reactants (e.g. CO) through the boundary layer (boundary layer diffusion)
2. Diffusion of reactants into the pores of the washcoat (pore diffusion)
3. Chemisorption of educts at the active phases of the catalyst
4. Chemical reaction of reactants at the active phases of the catalyst
5. Desorption of products from the active phases
6. Diffusion of products out of the pores of the washcoat (pore diffusion)
7. Diffusion of products (e.g. CO_2) through the boundary layer into the flue gas flow through the honeycomb cells (b: boundary layer diffusion)

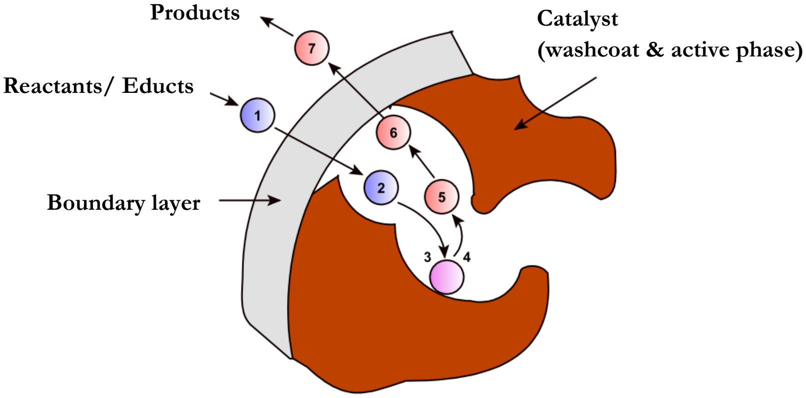


Figure 37: Characteristic phases of the heterogeneous catalytic conversion process [114] (translated in English)

The kinetics of the catalytic conversion depends on the catalyst activity, the concentration of reaction partners and the reaction temperature. The reaction rate depends exponentially on temperature conditions. However, the reaction rate is also influenced by transport mechanisms (e.g. diffusion) which are less temperature dependent. Therefore, three different areas can be distinguished for the kinetics of the reactions, especially for isothermal processes [113].

- Kinetic area
- Pore diffusion area
- Substance cross-over area (boundary layer diffusion area)

1) Kinetic area [113]

Compared to the diffusion rates of the reactants trough the boundary layer and pores the reaction rate in the kinetic area is slow. An increase of the temperature leads to a significant increase of the reaction rate (**Figure 39**). All available active sites (inner and outer surfaces) are used for the catalytic conversion. There is (almost) no decrease of the concentrations of reactants between the gas-filled cells and the catalyst interior (**Figure 38**).

2) Pore diffusion area [113]

The reaction rate in this area is high compared to the transport of reactants by pore diffusion, but low compared to boundary layer diffusion. An increase of temperature has less effect compared to the kinetic area (**Figure 39**). The concentration of reactants decreases towards the interior of the catalyst. However, on the outer surface of the catalyst the concentration of reactants is equally compared to the gas-filled channels (**Figure 38**).

3) Boundary layer diffusion area (Substance cross-over area) [113]

In the boundary layer diffusion area the reaction rate is high compared to the diffusion rate of reactants trough the boundary layer. Therefore, the boundary layer diffusion of reactants determines the speed of the conversion process. There is an abrupt decrease of reactants since they are immediately converted when they reach the outer surface of the catalyst (**Figure 38**). An increase of temperature would only lead to marginal or almost no increase of the conversion rates (**Figure 39**). In this area the effectiveness of the catalyst is highest.

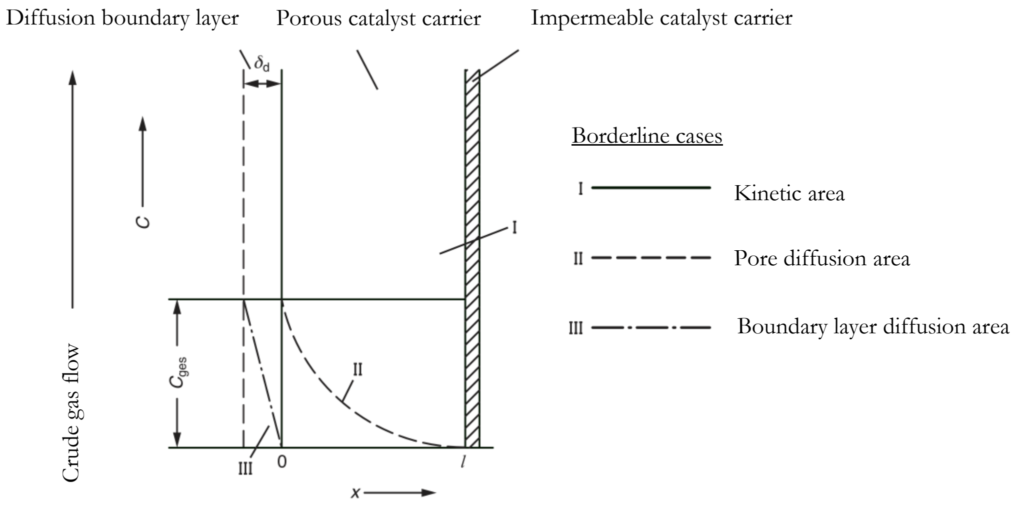


Figure 38: Concentration progression on a solid, porous catalyst (x=catalyst layer thickness, c=concentration of pollutant, δ_d=diffusion boundary layer, c_{ges}=concentration of pollutant in crude gas to be cleaned) [113]

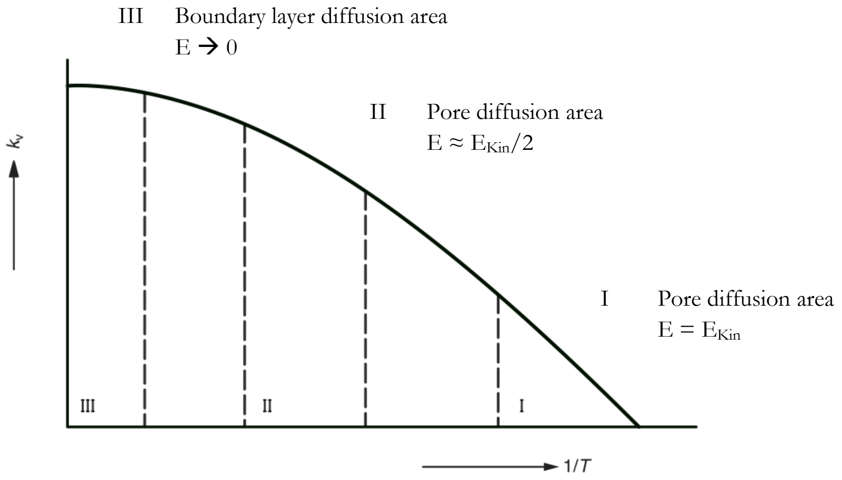


Figure 39: Influence of temperature of effective rate constant k_v (experimental activation energy E) [113]

4.1.4 Deactivation mechanisms

Catalyst deactivation means the decrease of the catalytic conversion performance over time [115] [116]. Catalytic systems have in general a certain service life due to natural ageing [113]. However, unsuitable operating conditions result in an acceleration of catalyst deactivation and consequently in a premature loss of function (**Figure 40**).

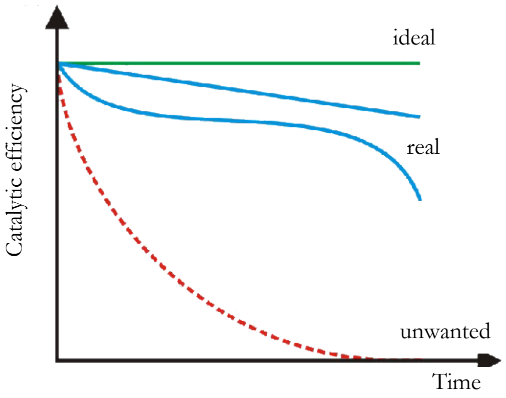


Figure 40: Examples of potential changes of catalytic efficiency in correlation to service time [117] (translated in English)

Deactivation mechanisms are in principal defined by three main mechanisms:

- Chemical deactivation
- Thermal deactivation
- Mechanical deactivation

1) Chemical deactivation

The most relevant type of chemical deactivation is poisoning of the catalyst. Poisoning is defined as strong chemisorption of unfavorable compounds, e.g. impurities, either on the intermediate and the catalytic layer (non-selective poisoning) or only on the active sites of the catalyst (selective poisoning). The poisoning compounds block the catalytic phase (**Figure 41**) [115]. Therefore, the catalytic surface decreases and the conversion rates can decrease rapidly. Typical compounds for poisoning of Pt, Pd-catalysts are lead (Pb), phosphorus (P), zinc (Zn), silica (SO_2) or iron (Fe) [116].

Another type of chemical deactivation is the formation of vapor compounds which are formed by reactions of certain gas species with the active phase of the catalyst [113]. Subsequently, the generated products are released into the crude gas. This process is defined as leaching and, for example, plays a relevant role when nickel (Ni) is used as active phase in catalysts (**Figure 41**) [115].

Poisoning mechanisms result most frequently in an irreversible deactivation of the catalyst [113]. Therefore, the operating conditions and the used type of catalyst have to be adjusted in technical applications in order to avoid poisoning.

2) Thermal deactivation

Thermal deactivation of the catalyst results from thermal degradation [116]. This means that the active sits are reduced by thermally induced mechanisms. For example, thermal sintering processes decrease the catalytic active surface area due migration of active sites (**Figure 41**) [115] or the loss of porosity of the intermediate layer. Thermal deactivation leads to an irreversible change of the catalytic efficiency [113]. Consequently, the maximum temperature conditions have to be considered for the application of a certain catalyst.

3) Mechanical deactivation

Mechanical deactivation is caused by fouling processes or by attrition [116]. Fouling is defined as the physical deposition of particles on the catalytic surface, i.e. onto the pores and/ or the active sites (**Figure 41**) [115]. Attrition means the loss of active phase material due to abrasion or due to mechanical-induced crushing of active sites of the catalyst. Both mechanisms lead to a decrease of the catalytic efficiency. However, deactivation by fouling is often (at least partially) reversible, e.g. oxidative regeneration of carbonaceous deposits [113].

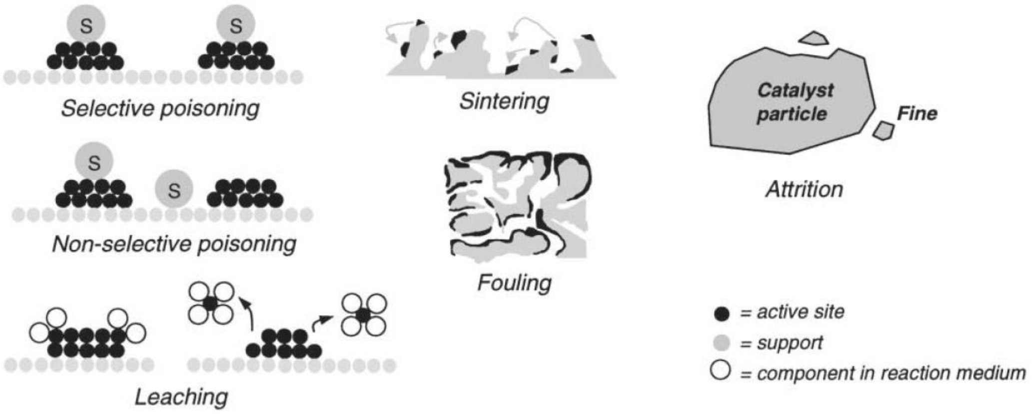


Figure 41: Major types of deactivation in heterogeneous catalysis [115]

4.2 Approach

In the following (chapter 4 – 6) the applicability of integrated honeycomb catalysts in firewood stoves is evaluated. They are regarded as potential secondary measure for manufacturers to significantly decrease gaseous and particulate emissions. The involved manufacturers selected two commercially available oxidizing honeycomb catalysts (ceramic and metallic – EnviCat®-Long Life Plus). Those selected catalysts should be evaluated regarding their catalytic efficiency (chapter 4), their impact on emissions in real applications as catalyst integrated solutions (chapter 5) and regarding long term durability and safety aspects during critical operating conditions (chapter 6).

In the first step, the catalytic efficiency of both types of platinum (Pt) and palladium (Pd) coated oxidizing honeycomb catalysts (ceramic and metallic carrier) was assessed. Since the evaluation of catalytic efficiency of integrated catalysts is difficult due to technical measurement reasons, a novel measuring methodology was applied. Therefore, a special test facility, called "DemoCat", was constructed which enabled parallel measurement in catalytically treated and untreated flue gas. With that DemoCat test facility it was possible to evaluate the conversion rates of integrated catalysts regarding carbon monoxide (CO), organic gaseous compounds (OGC) and particulate matter (PM) emissions under close to real-life operating conditions at the laboratory.

Thereby, testing close to real-life operating condition, as it is defined in this thesis, means that a whole test cycle is evaluated, including always ignition, preheating and several consecutive batches. For each evaluation all batches of the test cycle were considered. This is contrary to official-type-testing since it does not respect transient conditions, like ignition (batch 1) or preheating (batch 2) (see chapter 1.1 and chapter 7).

Potential correlations between catalytic conversion rates of CO, OGC and PM emissions, space velocity and coated area of honeycomb carriers were investigated and analyzed. Furthermore, catalytic conversion rates of CO and OGC emissions during the characteristic firewood combustion phases, i.e. start-up, intermediate and burn-out phase, were assessed.

The findings are important to identify how integrated catalytic systems could be used as synergetic solutions for primary optimized firewood stoves and provide profound knowledge of relevant criteria to develop and design catalyst integrated solutions.

4.3 Materials and methods

4.3.1 Used fuels

Beech ("*Fagus sylvatica*") and spruce ("*Picea abies*") firewood according to ÖNORM EN 14961-5:2011 standard [92] were used for all combustion tests (**Table 15**).

Table 15: Chemical properties of used firewood and kindling material

	Moisture content* (W) (kg/kg)	**Net calorific value** (H_u) (MJ/kg, d.b.)	**Ash content** (a) (g/kg, d.b.)	**Carbon** (C) (kg/kg, d.b.)	**Hydrogen** (H) (kg/kg, d.b.)	**Nitrogen** (N) (kg/kg, d.b.)	**Sulfur** (S) (mg/kg, d.b.)	**Chlorine** (Cl) (mg/kg, d.b.)
Analysis standard	EN 14774-1:2009 [93]	EN 14775:2009 [94]	EN 14775:2009 [95]	EN 15104:2011 [96]			EN 15289:2011 [97]	
Beech firewood ("*Fagus sylvatica*")	0.14 – 0.17	18.84	11	0.486	0.06036	< 0.001	93	36
Spruce kindling ("*Picea abies*")	0.12	18.29	4.5	0.488	0.0611	< 0.001	77	43

d.b. = dry base/ *as received

The firewood and kindling material derived from trees grown in the Austrian province "Lower Austria". It was provided by the local firewood producer HOFEGGER REINHARD (A-3250 Wieselburg) as ready to use products. The firewood was stored covered outside until the respective combustion tests were carried out.

4.3.2 Oxidizing honeycomb catalysts

A noble metal catalyst - platinum (Pt) and palladium (Pd) based on a washcoat of aluminum oxide (Al_2O_3) – on two different types of honeycomb carriers (ceramic and metallic) was used (**Table 16**).

Table 16: Physical properties of used types of ceramic and metallic honeycomb catalysts

Parameter	Unit	Ceramic honeycomb catalyst		Metallic honeycomb catalyst
Description	-	EnviCat®-LongLife Plus Ceramic		EnviCat®-LongLife Plus Metal
Material of carrier	-	mullite ceramic		brazed stainless steel
Cell shape	-	quadratic		trapezoid
Cell density	cells/cm^2	2.480		7.750
	cpsi	16		50
Shape	-	rectangular		rectangular
Dimensions (Length – l × Width – w)	m	0.59 × 0.158		0.063 × 0.168
Dimensions (Depth – d)	m	0.024	0.049	0.051
Abbreviation	-	cc-rec$_{24}$	cc-rec$_{49}$	mc-rec

Both types of honeycomb catalysts, called "EnviCat®-Long Life Plus", were specifically developed and adapted for firewood combustion in manually operated stoves. They are commercially available in different shapes and dimensions. In total three different types of catalytic devices were used (**Table 16**). All catalysts were not used before the combustion tests.

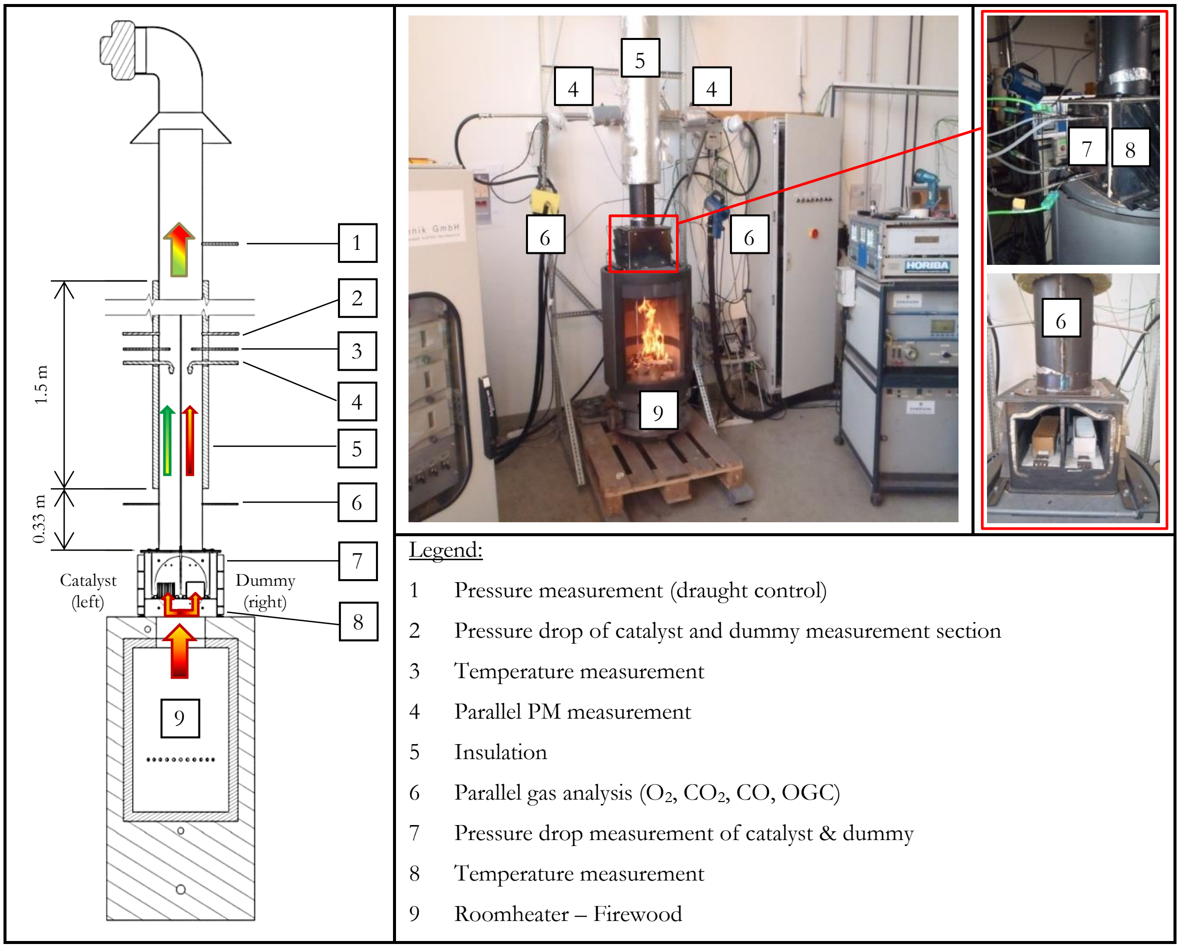
The main differences between the ceramic and metallic honeycomb catalysts were the shape of cells and the cell density. The ceramic catalyst had quadratic cell shapes and a cell density of 16 cpsi (cells per square inch) whereas the cell shapes of the metallic catalyst was trapezoid and the cell density 50 cpsi. The cell density correspond to 2.48 cells/cm^2 (ceramic) and 7.75 cells/cm^2 (metallic), respectively.

Also uncoated honeycomb carriers ("dummy") were used. These dummies had no catalytic effect, but enabled an equal pressure drop and therefore equal flow conditions. The physical data of these carriers were identical to the coated catalytic converters as it is given in **Table 16**. Catalysts and dummies were provided by the company CLARIANT.

4.3.3 DemoCat test facility

The DemoCat test facility was self-constructed and consisted of an adapted firewood stove and the subsequent measuring section (**Figure 42**).

The firewood stove was a roomheater classified according to EN 13240 [13] with a nominal thermal heat output of 10 kW. The round combustion chamber (diameter: 0.35 m, volume 0.0433 m^3) was lined with fire clay. The DemoCat test facility enabled integrated testing of catalytic honeycomb catalysts. Therefore, the post combustion chamber of the firewood stove and the downstream flue gas measuring section was split into two symmetric parts. In one part a catalyst, in the other part a dummy of the same dimensions were integrated (**Figure 42**). This ensured equal pressure conditions in both measurement sections. The tightness of both sections was proven by applying a blower door test with an overpressure of 10 Pa and measuring the leakage rate in both measuring sections. The leakage rate was below 1 m^3/h. According to pretests it was guaranteed that the total volume flow of the flue gas was equally distributed over both measuring sections. Due to the low flue gas velocities ($\leq$ 1 m/s) a continuous measurement, e.g. with a Prandtl or Pitot tube, was not possible. However, to identify potential deviations of volume flow conditions the static pressure drop of both measuring sections was continuously monitored. The inner dimensions of the DemoCat box were 0.25 m × 0.25 m × 0.21 m (length × width × height). The part of the box for the integrated dummy and the catalyst were 0.124 m × 0.25 m × 0.21 m (length × width × height). The diameter of the inlet cross section upstream the catalyst/dummy was 0.17 m. The cross section of the box outlet downstream of both measuring sections was 0.15 m in diameter.



Legend:

1 Pressure measurement (draught control)
2 Pressure drop of catalyst and dummy measurement section
3 Temperature measurement
4 Parallel PM measurement
5 Insulation
6 Parallel gas analysis (O_2, CO_2, CO, OGC)
7 Pressure drop measurement of catalyst & dummy
8 Temperature measurement
9 Roomheater – Firewood

Figure 42: Experimental setup of DemoCat test facility (scheme – left; pictures – right)

The catalysts and dummies were placed without any bypass in the respective part of the box by applying a drawer. In order to avoid any leakage also gasket material was placed around the catalyst and dummy (**Figure 43**; **Figure 44**). The maximum open cross section of drawers for catalyst integration was 0.156 m × 0.057 m (**Figure 43**).

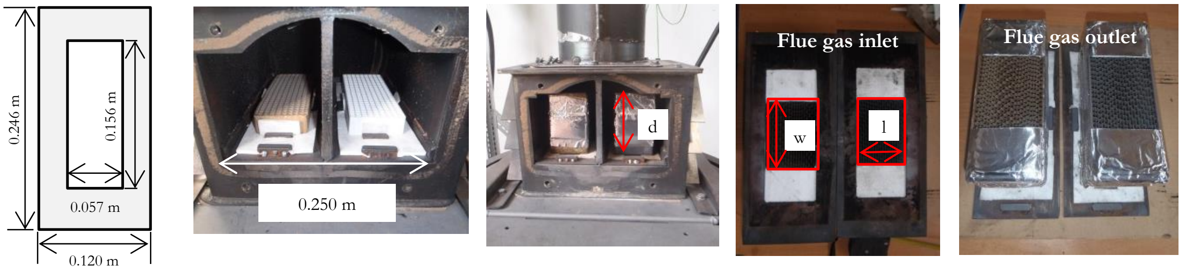


Figure 43: Scheme and example pictures of drawers for placement of catalyst and dummy in the DemoCat box. Variation of space velocity as well as coated area of honeycomb carrier by varying the effective catalytic volume using gasket material as well as alumina tape

Both measurement sections were equipped with measuring points for the gas analysis (O_2, CO_2, CO, OGC), PM measurement and for monitoring the pressure drop between both measurement sections. Measuring points at the back side of the DemoCat box were installed for temperature and pressure drop measurement (**Figure 42**). The complete measuring section with two separate flue gas sections was 1.83 m long. The first 0.33 m after the flue gas outlet was uninsulated. After the measurement section the flue gas of both sections was merged again and was conveyed in a chimney system (**Figure 42**). The draught in the chimney system was constantly controlled at 12 ± 2Pa for all test runs. The combustion air supply of the DemoCat was provided in three different streams, which were adjusted manually by three hand gears. Primary air was supplied just below the window of the combustion chamber door via a trench which was located around 0.05 m above the grate. Secondary air entered the combustion chamber via holes at the back wall of the combustion chamber. Thirdly, window purge air was supplied directly above the window of the combustion chamber door. This combustion air supply system represents a typical air staging concept of modern firewood stoves [52] [54].

4.3.4 Testing procedure

In order to investigate the performance of integrated ceramic and metallic honeycomb catalysts comparative combustion tests were carried out with the DemoCat test facility (**Figure 42**). The tests were conducted under typical real-life heating operating conditions including several consecutive batches as well as ignition and preheating [74]. The conversion rates regarding CO, OGC and PM emissions were assessed in correlation to different space velocities. Potential differences between ceramic and metallic honeycomb catalysts regarding conversion characteristics were investigated. In total nine different experiments were performed with the DemoCat as summarized in **Table 17**.

For the respective variations the catalytic volume and consequently also the space velocity and the coated areas of carriers (A_{cat}) of the ceramic and metallic catalytic honeycomb converters were varied in different steps. The space velocity variation was achieved by varying the cross-section area (blown area of catalyst) (**Figure 43**; **Figure 44**) or using different numbers of catalyst devices (**Table 16**) that were placed on each other. The cross-section area was adapted by covering part of it with inflammable gasket material. Additionally, these unused cells of the catalyst and dummy were masked with alumina tape (**Figure 43**; **Figure 44**).

Table 17: Variations tested for the ceramic and metallic catalytic honeycomb catalysts

Parameter	**Unit**	**Variation I**	**Variation II**	**Variation III**	**Variation IV**	**Variation V**
Ceramic honeycomb catalyst*	-	cc-rec$_{49}$	cc-rec$_{24}$	cc-rec$_{49}$	cc-rec$_{49}$	cc-rec$_{24}$ & cc-rec$_{49}$
Effective catalytic volume ($V_{c.eff}$)	cm³	145	213	290	436	649
Coated area of carrier (A_{cat})	m²	0.079	0.116	0.158	0.238	0.354
Blown area of catalyst	cm²	29.64	88.92	59.28	88.92	88.92
Metallic honeycomb catalyst*	-	mc-rec	mc-rec	mc-rec	2 × mc-rec	-
Effective catalytic volume ($V_{c.eff}$)	cm³	151	302	453	907	-
Coated area of carrier ($A_{cat.}$)	m²	0.185	0.369	0.554	1.108	-
Blown area of catalyst	cm²	29.64	59.28	88.92	88.92	

* Abbreviations and catalyst specifics defined in **Table 16**

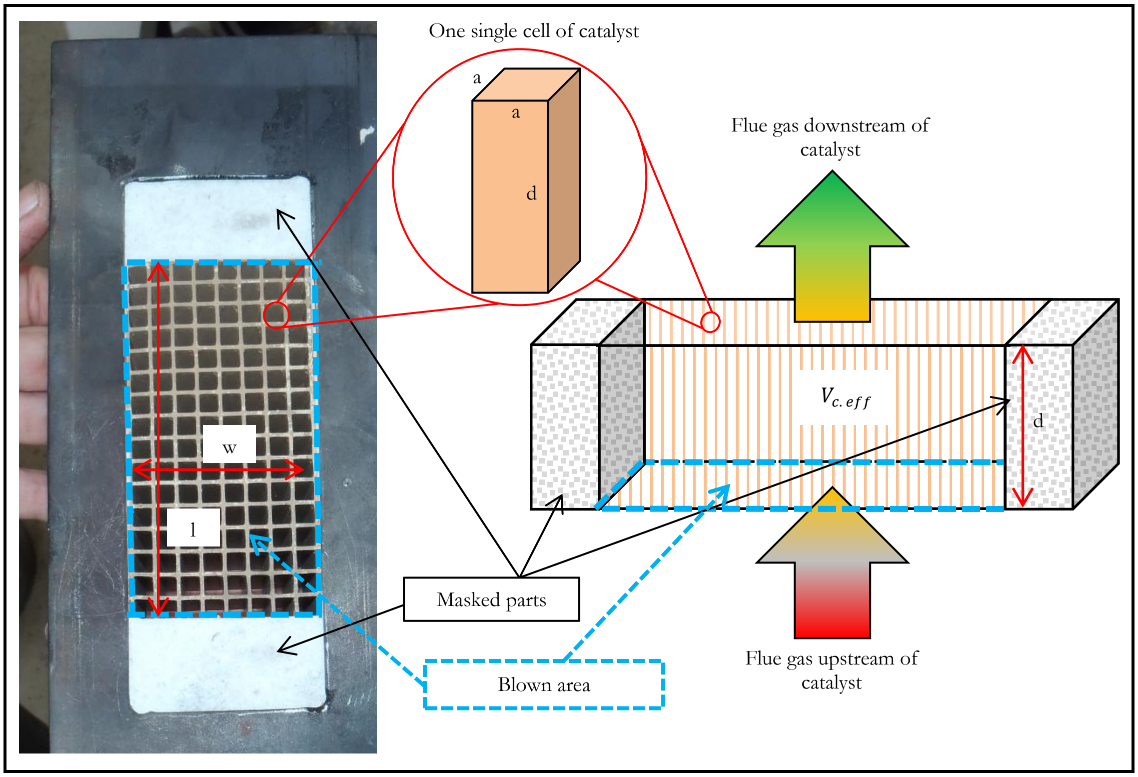


Figure 44: Illustration of the effective catalytic volume ($V_{c.eff}$) and the blown area (= w × l) of tested variations at the DemoCat (Example: Variation III of ceramic honeycomb catalyst)

For each variation a combustion test cycle comprised five consecutive batches and started from cold conditions. A batch started immediately after loading the appliance and closing the combustion chamber door. The batch was terminated when the CO_2 content of the flue gas reached 25% of the maximum CO_2 peak of the respective batch. This corresponded well with the recharging criteria of a maximum variation of firebed mass ±0.05 kg according to standard EN 13240 [13]. The test cycle was finalized when the recharging criteria after the fifth batch was reached. This test cycle should reflect real-life operation as described and discussed in REICHERT et al. [91].

Temperatures, pressure drops and the gaseous composition (concentrations of O_2, CO_2, CO, OGC as THC) were measured continuously and parallel in both measurement sections over the whole test cycle. In the dummy measurement section the gas analysis was conducted using the gas analyzer NGA 2000 – MLT4 for all test runs. For the test runs of different variations of the ceramic honeycomb converter flue gas analysis was done with the gas analyzer JCT Servopro 4900. For the test runs of metallic honeycomb converter the Horiba PG 350-E together with the Horiba VA-3000 CO analyzer was used. Gaseous organic carbon was measured by two equal FID measurement devices (M&A Thermo-FID PT63LT, M&A Thermo-FID ES). The different gas analysis devices were calibrated before each test cycle and were cross-calibrated in the same measurement section. The PM emissions were measured gravimetrically and parallel during each batch in both measurement sections. Therefore, two equal measuring equipment were used.

The heating operation was carried out according to the manual of the stove. For the ignition batch (batch 1) 0.5 kg of spruce kindling material (length of kindling material 0.25 m) and 6 pieces of beech firewood (length: 0.33 m) were used (complete mass: 3.0 kg). The placement of the firewood and kindling pieces in the combustion chamber was done cross-wise. The firewood pieces were placed directly on the grate, the kindling material was placed on top of the firewood pieces. Two pieces of commercial starting aids (wood fibers soaked with vegetable oil/FLAMAX Eco-Lighter [118]) on top of the kindling material were used for lighting the ignition batch according to the principle of top-down ignition technique [46]. For recharging a new fuel batch (batch 2 – batch 5) four pieces of beech firewood (length: 0.33 m) were used (complete mass: 3.0 kg). Also the firewood pieces for recharging were placed cross-wise on the grate in the combustion chamber. The air valve settings for window purge air and secondary air were fully open over the whole test duration. The air inlet flap settings for adjustment of primary air supply were fully open during the ignition and preheating batch (batch 1 and 2). During batch 3 to 5 the air valve setting for primary air supply was reduced from fully open (100%) to partly open (50%).

4.3.5 Instruments and measurements

Gaseous composition, flue gas temperatures and pressure drop were measured continuously using the devices specified in **Table 18**.

Table 18: Overview of measuring devices, measured components, measurement principle and accuracy for continuous measurements

Device and measured component	Range	Principle	Accuracy
NGA 2000 - MLT4 gas analyzer			
O_2	0 – 25 vol.-%	paramagnetic	< 0.5% of ultimate value of measurement range
CO_{low}	0 – 5000 ppm	NDIR	< 1% of ultimate value of measurement range
CO_{high}	0 – 10 vol.-%		
CO_2	0 – 20 vol.-%		
JCT Servopro 4900 gas analyzer			
O_2	0 – 100 vol.-%	paramagnetic	0.05% of measured value
CO_{low}	0 – 3000 ppm	NDIR	< 1% of measured value
CO_{high}	0 – 10000 ppm		< 1% of measured value
CO_2	0 – 25 vol.-%		< 1% of ultimate value of measurement range
Horiba PG-350 E			
O_2	0 – 25 vol.-%	paramagnetic	< 1% of ultimate value of measurement range
CO	0 – 5000 ppm	NDIR	< 0.5% of ultimate value of measurement range
CO_2	0 – 30 vol.-%		
Horiba VA-3000 CO Analyzer			
CO	0 – 10 vol.-%	NDIR	± 0.5% of measured value
FID M&A Thermo-FID PT63LT & MA Thermo-FID ES			
OGC (as THC)	0 – 100 ppm → 10000 ppm	flame ionization	< 4% of measured value
Thermokon DPT 2500 - R8			
Pressure drop	- 100 to + 100 Pa	piezoelectric effect	± 1.5% of ultimate value of measurement range
Thermocouple, Type K, class 1			
flue gas temperature	- 50 to + 1000 °C	thermoelectric effect (Seebeck)	± 1,5 °C of measured value

The fuel mass provided for each batch was measured with a balance (Type: Sartorius AW-8201, accuracy ± 0.1 g). Gravimetric PM measurement was carried out in the hot and undiluted flue gas according to the "Austrian and German method" described in CEN/ TS 15883:2009 [98]. Therefore, a partial flue gas stream was sampled during each batch for 30 min. The PM measurement started 3 min after the fuel batch started. The sampling rate was adjusted to 0.6 m^3/h (STP) and the diameter of the PM sampling nozzle was 12 mm. For retention of particles a stuffed quartz wool cartridge was used (3.5 g quartz wool per cartridge). The cartridge was placed in a heated filter head device (130 °C) which was located outside of the flue gas tract ("out-stack" measurement). Downstream of the filter the partial flue gas flow was conveyed into a gas drying unit and the volume flow was determined. Before and after measurement procedure all filter cartridges were dried in a drying oven at 130 °C for 4 h and subsequently cooled down in a desiccator equipped with silica gel for at least 8 h. The conditioned unloaded and loaded filters were weighed on a precision balance (Type: Satorius ME 235P, resolution ± 0.05 mg). PM emissions were determined according to the particle mass on the filter and the sampled partial volume flow of the flue gas.

4.3.6 Data evaluation and statistical evaluation

The analysis of gaseous CO emissions was done according to the standard EN 13240 [13], the analysis of OGC emissions according to CEN/TS 15883 standard [98]. Thereby, the average emission concentration of each batch of the test cycle was calculated and referred to 13 vol.-% oxygen level (mg/m^3, at 273.15 K and 101,325 Pa (= STP), dry, 13 vol.-% O_2). For determination of the catalytic emission conversion rate the values of the dummy measuring section were compared to the values of the catalyst measuring section. The catalytic conversion rate (CCR) was calculated based on emission concentrations of the flue gas in the DemoCat measurement sections according to **Eq. 5**.

$$CCR = \frac{\text{E dummy} - \text{E catalyst}}{\text{E dummy}} \times 100 \qquad \text{(Eq. 5)}$$

CCR	Catalytic conversion rate; in %
E catalyst	Emission concentration in catalytically treated flue gas; in mg/m^3, STP, dry, 13 vol.-% O_2
E dummy	Emission concentration in catalytically untreated flue gas in mg/m^3, STP dry,13 vol.-% O_2

The average catalytic emission conversion rate was calculated as arithmetic mean of all batches of a test cycle (batch 1 – 5).

The calculation of wet flue gas volume flow was carried out using the elemental composition of the fuel and flue gas measurements (i.e. O_2, CO_2, flue gas temperature) according to combustion calculations [119]. The total volume flow was equally distributed over both measuring sections of the DemoCat test facility. Therefore, 50% of the total volume flow was used to calculate the data for the measuring section with the integrated catalyst. The effective catalytic volume ($V_{c.eff}$) was defined as the volume of the catalyst limited by the external dimensions where the flue gas was led through (effective l × w × d) (**Figure 43**; **Figure 44**). This volume was used to calculate the space velocity which is defined as the ratio of wet flue gas volume flow at STP conditions and the effective catalytic volume ($V_{c.eff}$). For calculation of the geometric coated area of the honeycomb carrier (A_{cat}) the lateral area of one cell (e.g. for the ceramic honeycomb: A_{cat} of one cell = 4ad, see **Figure 44**) was calculated and multiplied by the cell density and by the cross section area of the honeycomb converter (w × l) of the respective combustion test (blown area of catalyst) (**Table 17**; **Figure 44**).

The catalytic conversion rate of CO and OGC emissions during the single characteristic combustion phases of a firewood combustion batch – as defined by PETTERSSON et al. [75] as start-up phase, intermediate phase and burn-out phase (**Figure 45**) – was assessed using the catalytic conversion rates of 5 min time intervals.

The characteristic combustion phases illustrated in **Figure 45** result from the batch-wise fueling of the stove with firewood and the processes of heating/drying, pyrolysis, gasification and oxidation of gaseous intermediate products. All processes occur simultaneously during batch-wise firewood combustion but their share on the whole combustion process varies during each batch phase. This results in different conditions, for example in respect to temperatures and air to fuel ratio [75]. This leads to the characteristic curves for flue gas temperature and O_2/ CO_2 as illustrated in **Figure 45**.

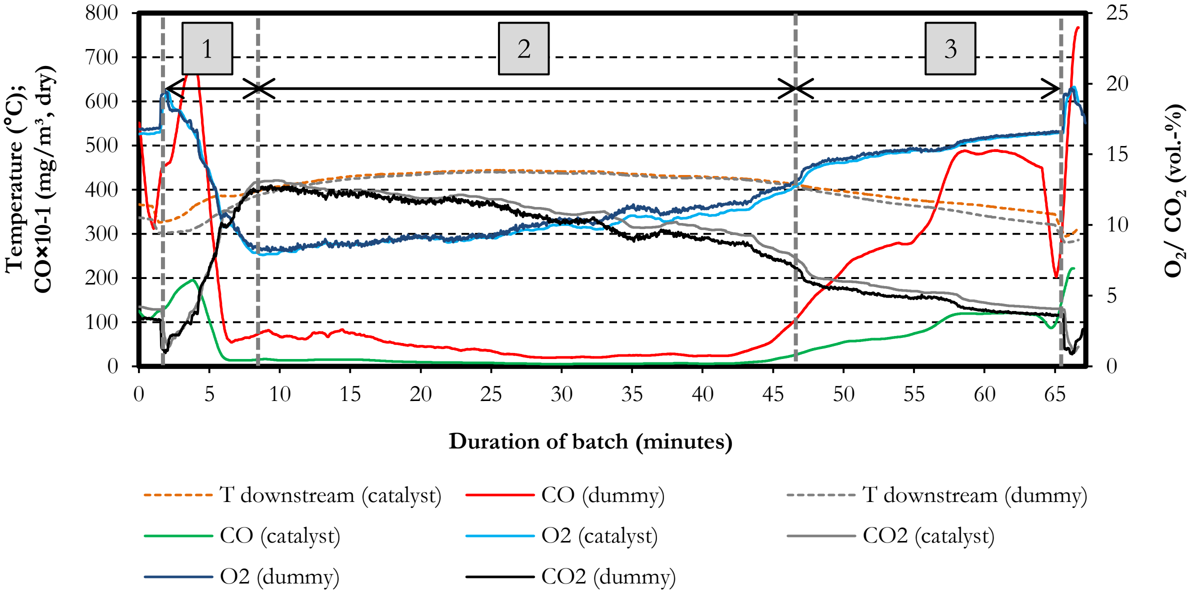


Figure 45: Illustration of characteristic combustion phases of a firewood batch after recharging a new fuel batch: start-up phase (1), intermediate phase (2) and burn-out phase (3). Measuring data of catalyst and dummy measuring section of the DemoCat are shown.

The classification of the respective conversion results to the single characteristic combustion phases was done according to following criteria:

1. Start-up phase – Duration from the beginning of a batch until the residual oxygen level reached its minimum. The start-up phase lasted around five to 10 min of the combustion batch.

2. Intermediate phase – Duration of the "quasi stationary" combustion phase of a firewood combustion batch with comparatively low CO emissions and nearly constant flue gas temperatures. This phase was finished when flames extinguished. The duration of the intermediate phase was typically around 40 min.

3. Burn-out phase – The burn-out phase started when flames extinguished and CO emissions as well as residual oxygen level increased during the charcoal burnout until recharging. The flue gas temperature level decreased. The burn-out phase lasted around 15 – 20 min.

To assess statistical validity the significance of conducted combustion tests was calculated by using a Student's t-Test. The correlation of space velocity, coated area and conversion rate was determined by regression analysis. The significance of correlation coefficients (Pearson's correlation coefficient: r_P) was evaluated by an f-test. The following interpretations are used for the p values resulting by Student's t- and f-tests:

- $p < 0.01$ highly significant difference or correlation
- $p < 0.05$ significant difference or correlation
- $0.32 \leq p \geq 0.05$ no significance, but a clear trend of difference or correlation
- $p > 0.32$ no significance and no trend of difference or correlation

In the following subchapter respective p values or Pearson's correlation coefficients are given in brackets for each statistical test.

4.4 Results and discussion

The curves of flue gas temperatures and residual oxygen (O_2) and carbon dioxide (CO_2) concentrations in the flue gas of both measurement sections showed a good conformity (**Figure 45**; **Figure 46**).

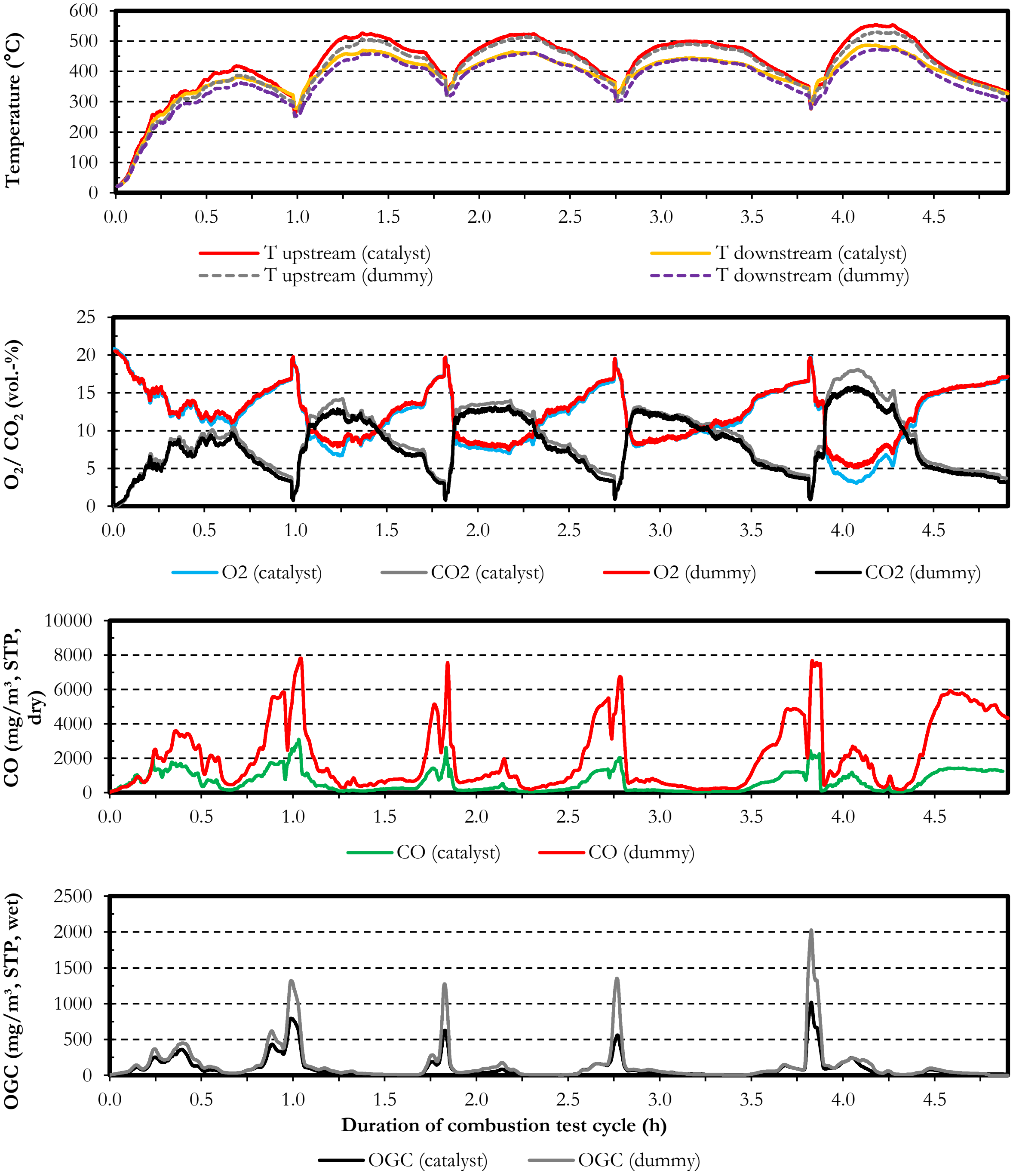


Figure 46: Example of a combustion test consisting of 5 batches in the DemoCat test facility (Variation IV, ceramic catalyst)

This revealed that the measuring methodology of the DemoCat worked properly and confirmed that the total volume flow was equally distributed over the catalyst and dummy measuring section during the combustion test. Anyway, to respect potential deviations of the different measuring devices (e.g. O_2/CO_2 of the 5th batch in **Figure 46**) the catalytic conversion rates were calculated based on average emission concentrations of each measuring section referred to 13 vol.-% O_2 (**Eq. 5**).

After lighting the ignition batch the flue gas temperature at the catalyst increased continuously. The catalytic conversion, indicated by different CO concentrations at the dummy and catalyst measurement section, started after 5 – 12 min. At that time the temperature at the catalyst reached around 250 °C (**Figure 46**). This response time for catalytic efficiency is considerably faster compared to response times of around 15 – 20 min for retrofitted catalysts without external heating reported by different studies [58] [59] [61]. The light-off temperature of the experiments corresponded well with previous studies where the light-off temperatures of catalytic CO conversion was observed at 250 °C – 300 °C [61]-[63]. During the ignition batch (batch 1) the flue gas temperature at the catalyst was lower compared to the following batches. From batch 2 to 5 the flue gas temperature at the catalyst was around 400 °C – 500 °C, except during the start-up phases after recharging a new fuel batch and during the burn-out phases of firewood batches (**Figure 46**). However, over the entire time of heating operation the temperature at the integrated catalysts was at least 300 °C which was identified as minimum temperature for an effective emission reduction [58].

Comparing the measurements of the dummy and catalyst section a slight increase of temperature due to the catalytic conversion is obvious at the beginning (start-up phase) and at the end of the batch (burn-out phase) (**Figure 45**). However, the increase of flue gas temperature was less than around 50 K for a short period (~5 minutes). The increased temperature downstream the catalyst correlates well with phases of increased CO emissions (>2500 mg/m^3, STP, dry, 13 vol.-% O_2) (**Figure 45**). However, the impact of catalytic temperature increase on the average batch results of flue gas temperature was low (**Figure 46**).

In the following the results of the complete test cycles (including batch 1 to 5, **Figure 46**) for both catalysts and all tested variations are evaluated and analyzed (**Table 19**).

Table 19: Average results of batch 1 – 5 of performance analysis tests regarding the pressure drop induced by integrated honeycomb catalysts (Pa), flue gas temperatures up- and downstream of honeycomb catalysts (°C), the flue gas volume flow through the catalytic converter (m^3/h, STP, w.b.) as well as space velocity (1/h)

Parameter	**Unit**	**Ceramic honeycomb catalyst**					**Metallic honeycomb catalyst**			
Variation	-	I	II	III	IV	V	I	II	III	IV
Pressure drop	Pa	12.8	1.6	5.6	2.4	4.7	14.3	7.8	4.2	7.7
$T_{flue\ gas}$ upstream catalyst	°C	418	456	465	428	453	403	421	420	403
$T_{flue\ gas}$ downstream catalyst	°C	370	409	415	393	408	363	389	395	385
Volume flow through catalyst	m^3/h, STP, w.b.	8.8	9.5	9.0	10.0	9.4	7.8	8.8	9.2	8.8
Blown area of catalyst	cm^2	29.64	88.92	59.28	88.92	88.92	29.64	59.28	88.92	88.92
Space velocity	1/h	60652	44322	31121	22915	14487	51736	28959	20246	9692
Coated area of carrier (A_{cat})	m^2	0.079	0.116	0.158	0.238	0.354	0.185	0.369	0.554	1.108

w.b. = wet based

The average temperature conditions of combustion tests (including all measurements of batch 1 to 5), measured just upstream the honeycomb catalyst, were equal to around 400 °C for the tested variations of both types of honeycomb catalysts. For the variations tested, average temperatures ranged between 418 °C (variation I) and 456 °C (variation II) for the ceramic honeycomb catalyst and between 403 °C (variation I & IV) and 421 °C (variation II) for the metallic honeycomb catalyst.

The measured pressure drop of both types of honeycomb catalysts was below 8 Pa except at variation I of both catalysts (**Table 19**). The pressure drop increased due to increased flue gas velocities, especially when the blown area of the catalyst at the respective test variation was reduced (ceramic: variation I & III/ metallic: variation I & II/ **Table 19**; **Figure 44**). This means, for example, that at variation I and III of the ceramic honeycomb catalyst the blown area was 29.64 cm^2 and 59.28 cm^2 compared to 88.92 cm^2 for variations II, IV and V.

Due to the higher cell density (**Table 16**) the pressure drop of the metallic catalyst was generally higher compared to the tested variations of the ceramic catalyst, even when the blown area and the space velocity were similar (**Table 19**). Consequently, the type of carrier, geometry and the design of integration conditions affect the volume flow and primary combustion conditions which can affect the catalytic efficiency. This has to be considered during the development process of catalyst integration and for assessing the catalytic efficiency.

In order to evaluate the comparability of the tested variations the average emissions of the different test variations at the dummy measurement section were statistically analyzed (see **Table A3**; **Table A4**). For the ceramic honeycomb catalyst the different pressure drop and volume flow conditions did not lead to significant differences of emission concentrations in catalytically untreated flue gas for (dummy measurement section) (**Table A3**). Therefore, variation I to V had similar initial conditions to evaluate the catalytic efficiency. In contrast, variation I of the metallic honeycomb catalyst with a pressure drop of 14.3 Pa revealed a significant primary effect due to changed combustion conditions. Accordingly, variation I showed significantly increased emissions of CO compared to variation II ($p = 0.017$) and variation III ($p < 0.01$) (**Table A4**). Thus, this has to be respected in the interpretation of results. For the remaining variations, where the induced pressure drop was below 8 Pa, the primary combustion conditions, indicated by CO and OGC emissions, were comparable.

4.4.1 Effect of space velocity and coated area of honeycomb carriers

There is a linear correlation of the effective catalytic volume ($V_{c.eff}$) and the coated area of honeycomb carrier (A_{cat}), whereas space velocity and coated area of honeycomb carrier are characterized by a power function (**Figure 47**).

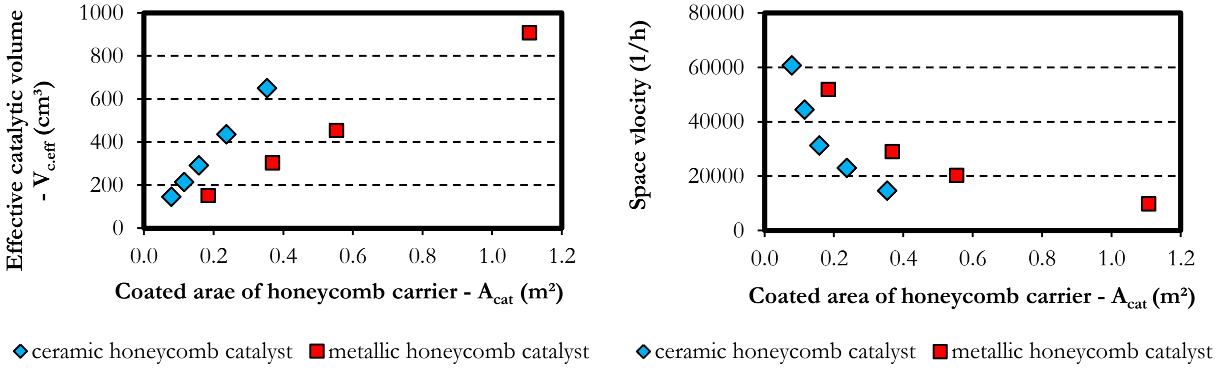


Figure 47: Correlations of effective catalytic volume, coated area of honeycomb catalyst and space velocity.

For dimensioning of integrated catalyst solutions both parameters can be used. Therefore, results of catalytic conversion rates are shown in correlation to space velocity and coated area of honeycomb converter (**Figure 48**).

Catalytic conversion rates in correlation with the space velocity and coated area of honeycomb converter (**Figure 48**) indicated a clear trend or even a highly significant correlation between CO (ceramic & metallic: $p < 0.01$) and OGC (ceramic: $p = 0.055$; metallic: $p < 0.01$) conversion and space velocity for both types of catalytic honeycomb converters.

For the ceramic honeycomb catalyst the average catalytic conversion rate of CO emissions ranged between 53% and 82% and for OGC emission conversion between 32% and 45% of the tested variations. The catalytic conversion rate of CO emissions decreased clearly and almost linearly

(r_P = -0.81) between space velocities of 14,000 1/h and 45,000 1/h from 80% to 50%. In contrast, the OGC emission conversion rate correlated only slightly in that range of space velocities (r_P = -0.39). The OGC conversion rates differed only of about 10% absolutely in that range.

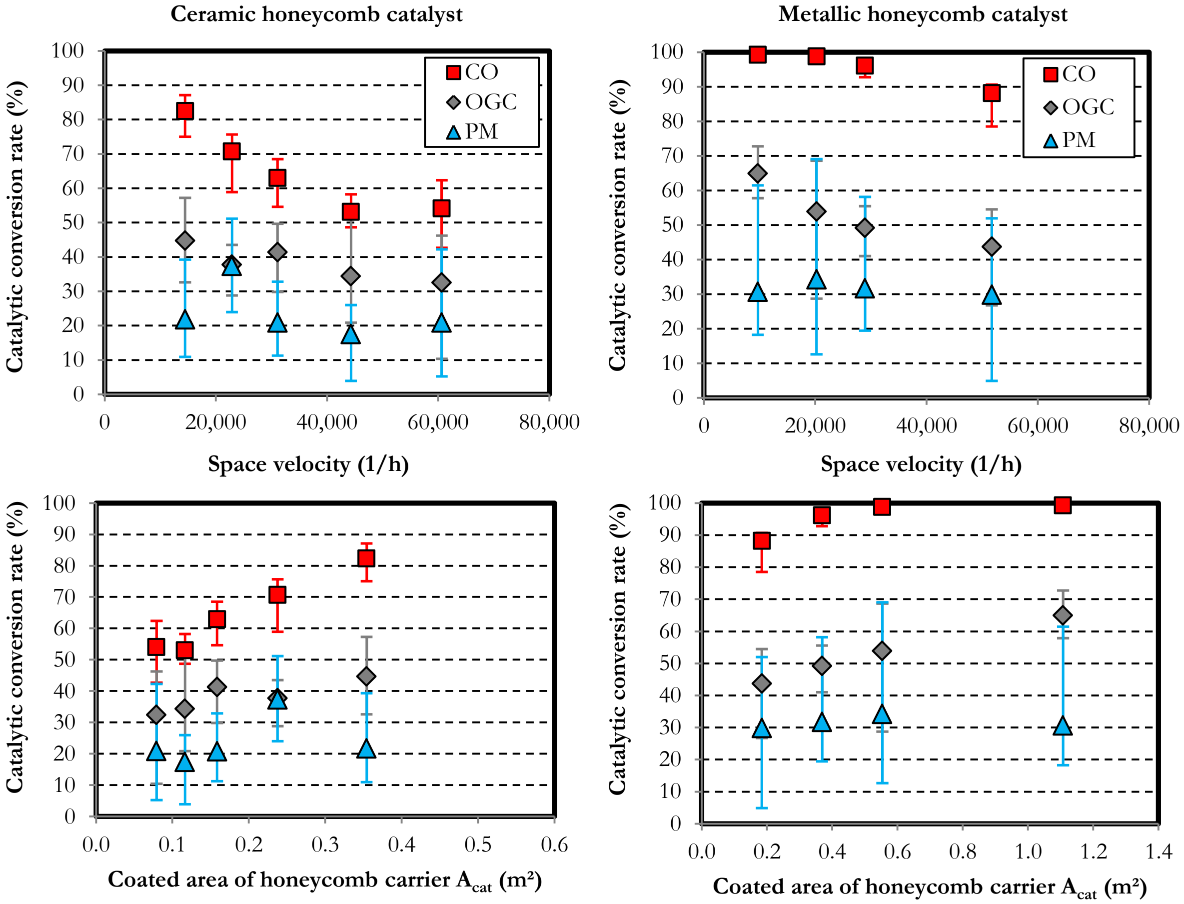


Figure 48: Test results of ceramic (left) and metallic (right) honeycomb catalysts regarding the correlation of space velocity (top) as well as coated area of honeycomb carrier (bottom) and conversion rate. Error bars represent the maximum and minimum values determined.

For the tested variations of metallic honeycomb catalyst CO emission conversion rates ranged from 88% to nearly 100%, OGC emission conversion from 44% to 65%. This type of honeycomb catalyst showed only a marginal decrease of catalytic conversion rates between space velocities of 10,000 1/h and 30,000 1/h which indicated that CO emission concentrations were too low for the available catalytic surface. Consequently, an increase of catalytic surface did not lead to an increased conversion rate. Even in the case that variation I with the very high space velocity of around 50,000 1/h was respected, the average conversion rates of OGC emissions showed a highly significant correlation in the tested range of space velocities ($p < 0.01$, r_P = -0.59).

The DemoCat tests confirmed the correlation of catalytic activity on gaseous emissions and space velocity, even if conversion rates of total heating cycles with varying conditions are respected. This is important to mention since previous studies investigated the effect of space velocity only under synthetic flue gas conditions and a controlled variation of temperatures [61] [62]. According to the findings of CARNÖ et al. [62] the catalytic conversion of CO differed by 55% comparing the conversion rates at space velocities of 20,000 1/h and 50,000 1/h at 180 °C. For representatives of OGC emissions, i.e. methane (CH_4) and naphthalene ($C_{10}H_8$) the characteristic conversion temperatures changed, but no effect on conversion rates was evident at 180 °C. However, as the highest catalytic OGC emissions conversion of noble metal catalysts were observed at temperatures higher than 350 °C [62] [63] an effect of space velocity on conversion rates of the DemoCat tests is explainable since the temperatures at the catalyst ranged around 400 °C – 500 °C (**Figure 46**). The change of characteristic conversion temperatures for OGC emissions was also reported by BENSAID et al. [61] according to between 3500 1/h and 10,000 1/h.

For PM emissions the DemoCat results did not confirm the same impact of space velocity as observed for gaseous emissions since no trend or significant correlation of conversion rate and space velocity was observed for both types of honeycomb catalysts. The average PM emission conversion rate ranged from around 20% to about 40% for all tested variations (**Figure 48**) which is in line with previous studies[59] [60] [65]. However, there are studies that showed a catalytic PM emission reduction potential up to 90% [61].

For the metallic catalytic converter the average PM emission conversion was higher by about 10%. But statistical analysis revealed no significant trend or difference for both types of catalytic converters. The high ranges of fluctuations in the conversion rates are explainable since all measurements of the heating cycle were considered (including ignition and preheating). Anyway, in all test runs the PM emissions at the catalyst measuring section were lower compared to the dummy measuring section. This confirmed that the catalytic process had a reductive effect on PM emissions. The test set-up of the DemoCat excluded any influence of potential primary effects of honeycomb carriers. It is assumed that the catalytic PM reduction bases on processes referred to impaction, thermophoresis and/or diffusiophoresis [120]. Especially processes of diffusiophoresis induced by the concentration gradient of CO and CO_2 in the flue gas due to catalytic conversion processes might be responsible for an enhanced deposition of particles on the catalyst's surface [121] [122]. Subsequently, the combustible share of agglomerated particles is oxidized when the temperature level is sufficiently high (**Figure 46**). This explanation is also supported by the study of BENSAID et al. [61] which reported a significant catalytic reduction effect on particle number concentrations. Even a catalytic effect on particle size distribution was observed in the study of HUKKANEN et al. [59] which assumes that the conversion of OGC emissions affect not only particle mass concentration, but also the particle size distribution.

Summarizing, the catalytic conversion rate under DemoCat test conditions was limited by the concentrations of emissions and the available catalytic active surface. Consequently, the catalytic CO conversion rate was limited by the diffusion processes of CO towards the catalyst's surface. Regarding OGC emission conversion the same characteristic as for CO emissions was observed. However, the catalytic oxidation potential of OGC was in general lower. This corresponds well to the findings of another study carried out with the same type of honeycomb catalysts [65]. Accordingly, it was shown that the conversion rate of OGC emissions is around 48%. The conversion rates for methane (CH_4) emissions were clearly lower at around 10% – 20% at a temperature of 300 °C. The catalytic effect on PM emission reduction showed no correlation with the space velocity or the coated area of the honeycomb carrier. A potential effect of different fractions of PM emissions, i.e. organic, inorganic and soot, on catalytic PM emission conversion need further investigations.

The differences in effectiveness of both types of honeycomb catalysts were evaluated using variation IV (ceramic) and variation III (metallic), since the average space velocity was quite similar (**Table 19**). The CO conversion rate of the metallic catalytic converter was highly significantly better ($p < 0.01$) and also OGC emission conversion was by trend better ($p = 0.08$) for the metallic honeycomb catalyst. Regarding PM emissions no trend of difference was observed ($p = 0.80$). The differences regarding CO and OGC conversion performance of both types of honeycomb catalysts reasoned in the higher cell density and the subsequent higher catalytic active surface of the metallic catalytic honeycomb converter.

4.4.2 Catalyst performance during characteristic combustion phases

Both types of catalytic honeycomb converters showed comparable conversion behaviors for CO and OGC emissions during characteristic firewood combustion phases. This is illustrated in **Figure 49** and **Figure 50** which represent an evaluation of 5 minutes intervals of flue gas measurements measured simultaneously in the catalyst and dummy measurement sections (**Figure 42**).

Figure 49 shows exemplarily the catalytic conversion rates of CO and OGC emissions of the ceramic (left) and the metallic (right) honeycomb catalysts during one batch. The catalytic conversion rates are represented by the bulks and referred to the single characteristic combustion phases (**Figure 45**). The bullets of both diagrams represent the raw gas emissions of both species measured in the dummy measurement section.

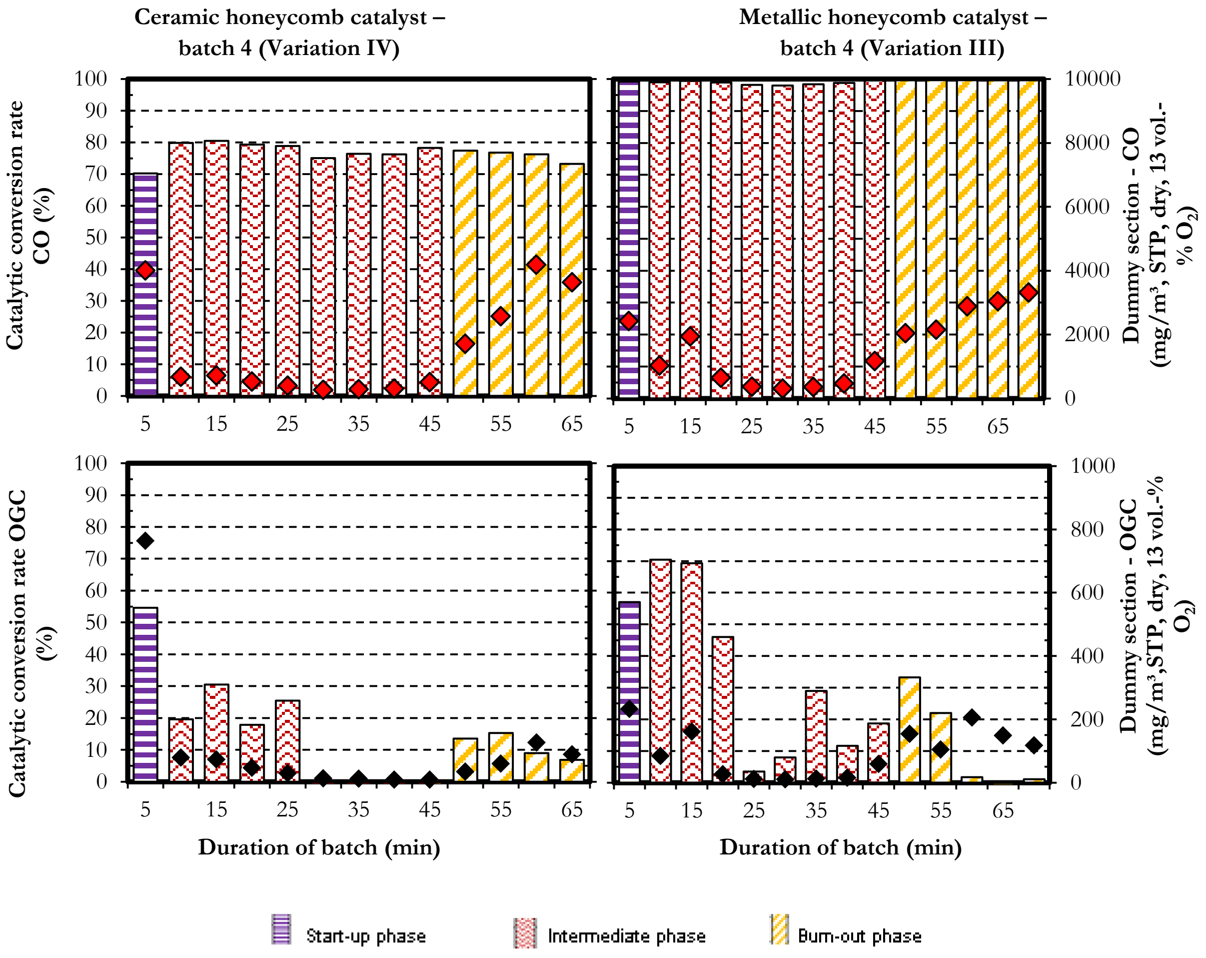


Figure 49: Catalytic conversion rate of CO and OGC emissions during characteristic combustion phases – start-up phase, intermediate phase and burn-out phase (Bulks represent the catalytic conversion rate in %). The bullets represent the emission concentrations measured at the dummy measurement section for the respective interval (in mg/m³, STP, dry, 13 vol.-% O_2). The left diagrams illustrate the results for the ceramic catalyst, the right diagrams show the results for the metallic catalyst.

The catalytic CO conversion rates varied only marginally by around ±5% (**Figure 49**) although emission concentrations varied between 200 mg/m^3 and 4000 mg/m^3 (STP, dry, 13 vol.-% O_2) of the complete batch. This confirms the theory that the catalytic conversion rate was limited by diffusion mechanisms of molecules within the cells towards the catalytic active surface and not by the kinetics of the catalytic process itself [113]. For the ceramic honeycomb catalyst the CO conversion rates ranged between 70% and 80%. The metallic honeycomb catalyst showed catalytic conversion rates higher than 95% during the characteristic combustion phases. However, since the absolute CO emission level was generally much higher during the start-up as well as burn-out phase (2000 mg/m^3 – 4000 mg/m^3) the most effective CO emission reduction was achieved during these phases. Hence, the effectiveness in terms of emission reduction needs to be assessed by looking at the percentage of total converted CO and OGC emissions of the respective batch. This is shown in **Figure 50** where the share of catalytic emission conversion of total converted batch emissions is illustrated in combination with the temperature curves up- and downstream of the catalysts. Thereby, in relation to **Figure 49** the effectiveness of the catalysts during the different characteristic combustion phases (**Figure 45**) as well as potential influences of the temperature level is analyzed.

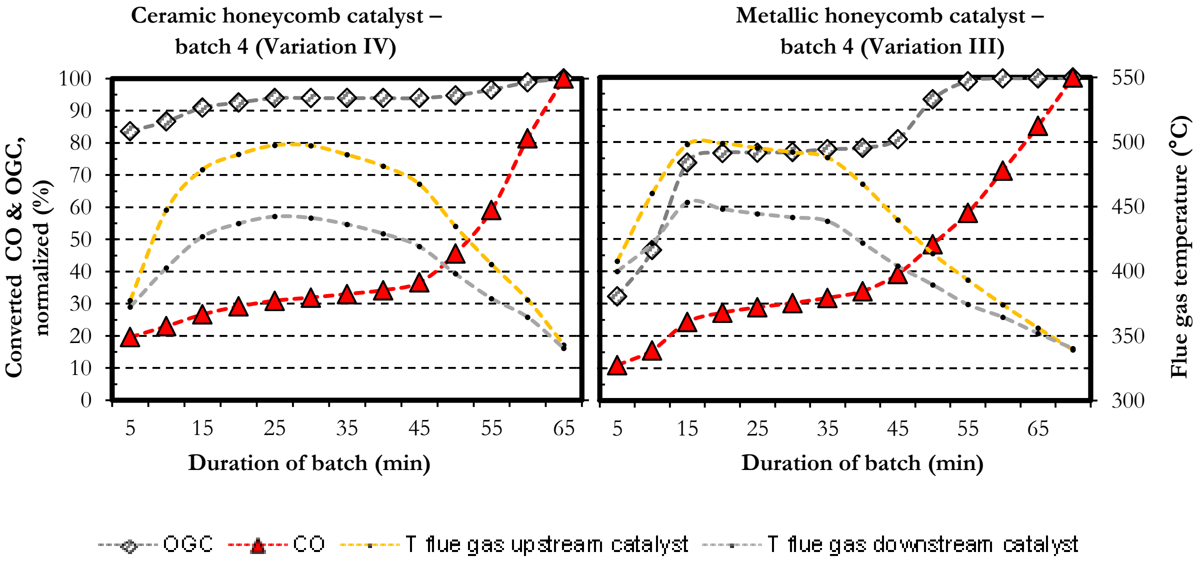


Figure 50: Relative cumulated converted CO and OGC emissions during characteristic combustion phases – start-up phase, intermediate phase and burn-out phase of ceramic (left) and metallic (right) honeycomb catalysts.

The percentage of cumulatively converted emissions for each interval based on the absolute mass of converted CO and OGC emissions of the respective batch. Regarding the catalytic OGC conversion rates the results were different. The relative catalytic OGC emission conversion was by trend highest during the start-up phase and found its minimum during the first half of the intermediate phase (~first

25 min of respective batch) (**Figure 49**). Respectively, the most effective OGC emission reduction of about 75% to more than 80% was achieved during the start-up phase and first part of the intermediate phase (**Figure 50**), when the emission concentrations were at the highest level (around 100 mg/m^3 up to more than 700 mg/m^3) (**Figure 49**).

The reason for the different OGC conversion rates during the characteristic combustion phases is most probably the share of methane (CH_4) of total OGC emission measurement, especially during the burn-out phase [123]. In general, CH_4 is a very stable molecule [124] which needs high temperatures for oxidation, even with catalytic systems [123]. It was shown that methane in general is poorly catalytically oxidized by this type of honeycomb catalyst (10% – 20% at 300 °C temperature) [65]. Another study, where a platinum catalyst was used at a space velocity of 20,000 1/h, showed that CH_4 conversion did not start before a minimum temperature of 400 °C. At least 500 °C were necessary to achieve catalytic CH_4 conversion rates of 10% – 15% [62].

Since OGC emissions during the burn-out phase of firewood combustion are dominated by CH_4 emissions [123] and the flue gas temperature at the catalyst decreases (**Figure 50**), the low conversion rates of OGC emissions during the burn-out phases can be explained.

4.5 Summary – Catalytic efficiency of integrated oxidizing honeycomb catalysts

In the previous chapter a novel methodology and a specific test-setup (DemoCat) were presented to evaluate the catalytic efficiency of integrated catalytic converters in firewood stoves. Thereby, the effect of ceramic and metallic honeycomb catalysts on gaseous and particulate emissions were analyzed under real firewood combustion conditions at different space velocities.

The results revealed high emission reduction potentials of more than 95% conversion rate for CO, 60% for OGC and about 30% for PM emissions. The conversion rates of CO and OGC emissions correlated with the space velocity and the coated area of honeycomb carriers. Beside temperature conditions, these both parameters were identified as key parameters for the integration design. For PM emission conversion no correlation with the space velocity was evident.

Furthermore, the measurements showed a quick response time of the catalytic activity of around 5 – 12 min due to a fast increase of temperatures at the catalyst to more than 250 °C. The elevated temperature level at the catalyst due to the integration design promotes better catalytic conversion rates, especially for catalytically convertible OGC emissions and deposited soot or organic compounds on the catalyst surface.

Primary optimization measures focuses particularly on the stretched intermediate phase of a combustion batch. As it was shown by the measurements the percentage of catalytic conversion rates differed only marginal between the characteristic combustion phases for CO emissions. For OGC emissions the highest conversion rates were observed during the start-up phase and the first part of the intermediate phase. However, since CO and OGC emission concentrations are highest during the start-up and burn-out phase of a firewood batch, the most effective catalytic emission reduction in terms of absolute emission reduction was observed during these phases. Consequently, the use of integrated honeycomb converters and implemented primary measures revealed highly synergetic effects.

The metallic honeycomb catalyst showed significantly better CO emission conversion rates and by trend also higher OGC conversion rates compared to the ceramic catalyst. This was due to the increased cell density and consequently the increased amount of coated area of the metallic honeycomb carrier.

Concluding, it appears that catalyst integrated firewood stoves represent an effective option for stove manufacturers in order to decrease emissions significantly and to meet future emission limit values. This becomes even more relevant if future test methods focus on real-life heating evaluation and also take into account transient combustion conditions, like the ignition and preheating phase as illustrated in the following chapter.

5 APPLICABILITY OF OXIDIZING HONEYCOMB CATALYSTS – EMISSION IMPACT[7]

5.1 Approach

Following the evaluation of the catalytic efficiency at the DemoCat test facility the effect of the integrated catalysts on CO, OGC and PM emissions was assessed at five different serial-production firewood stoves. Thereby, the impact of catalyst carrier geometries on primary combustion conditions was specifically quantified. This so called primary effect of catalyst integration results from an additional pressure drop in the post combustion chamber which influences volume flow conditions of combustion air supply. Consequently, combustion conditions and combustion processes are affected resulting in different emissions. Considering the primary effect is important for further development of catalyst integrated solutions. The primary effect should be also respected when distinguishing between the exclusive catalytic emission conversion (catalytic coating) and the total effect of the catalytic system (catalyst carrier geometry and catalytic coating) on emissions. Finally, exemplary data about the catalytic effect on the PM emission composition are presented in order to identify a potential selectivity of PM reduction effects.

5.2 Materials and methods

5.2.1 Used fuels

Beech ("*Fagus sylvatica*") firewood according to ÖNORM EN 14961-5:2011 standard [92] was used for all combustion tests (**Table 20**).

The kindling material for the ignition batch was spruce ("*Picea abies*"). The firewood as well as kindling material was provided by the local firewood producer HOFEGGER REINHARD (A-3250 Wieselburg) as ready-to-use products. The firewood derived from trees grown in the Austrian province "Lower Austria" and was dried after splitting for at least two years under ambient conditions in covered

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[7] Segments of this section have already been published in [6].

piles. It was delivered in boxes with a volume of about 1 m^3. The boxes were stored in an outside cabin until the combustion tests were carried out.

Table 20: Chemical properties of used firewood and kindling material

	Moisture content* (W) (kg/kg)	**Net calorific value** (H_u) (MJ/kg, d.b.)	**Ash content** (a) (g/kg, d.b.)	**Carbon** (C) (kg/kg, d.b.)	**Hydrogen** (H) (kg/kg, d.b.)	**Nitrogen** (N) (kg/kg, d.b.)	**Sulfur** (S) (mg/kg, d.b.)	**Chlorine** (Cl) (mg/kg, d.b.)
Analysis standard	EN 14774-1:2009 [93]	EN 14775:2009 [94]	EN 14775:2009 [95]	EN 15104:2011 [96]			EN 15289:2011 [97]	
Beech firewood (“*Fagus sylvatica*”)	0.14 – 0.17	18.84	11	0.486	0.06036	< 0.001	93	36
Spruce kindling (“*Picea abies*”)	0.12	18.29	4.5	0.488	0.0611	< 0.001	77	43

d.b. = dry base/ *as received

5.2.2 Oxidizing honeycomb catalysts

The same types of honeycomb catalysts as described in the previous chapter were used (ceramic and metallic carriers) and integrated in different firewood roomheaters. The honeycomb carriers were coated with a washcoat of aluminum oxide (Al_2O_3). The washcoat contained the catalytic active noble metals platinum (Pt) and palladium (Pd). Both types of catalysts (ceramic and metallic) were specifically developed for the use in manually operated firewood stoves. They are commercially available and are sold under the brand name “EnviCat®-Long Life Plus” by the company CLARIANT.

The catalyst based on a ceramic carrier material was round shaped with a diameter of 0.144 m and a cell density of 3.875 cells/cm² or 25 cpsi (cc-round). Compared to the rectangular ceramic catalysts used for the DemoCat tests the cell density of the round shaped ceramic catalysts was higher (see **Table 16**). The metallic honeycomb catalyst based on a carrier of brazed stainless steel. The metallic catalyst was also round shaped with a diameter of 0.149 m and a cell density of 7.750 cells/cm² or 50 cpsi (mc-round). The depth of both types of catalyst was 0.051 m. The shape of single cells of the ceramic catalyst was rectangular, the shape of single cells of the metallic catalyst was trapezoid (see **Table 16**).

Also uncoated honeycomb carriers (“dummy”) were used which had no catalytic effect, but enabled an equal pressure drop and therefore equal primary combustion conditions. The physical data of these carriers were identical to the coated catalytic converters (cc-round, mc-round). Catalysts and dummies were unused before the combustion experiments and were provided by the company CLARIANT.

5.2.3 Combustion appliances and test-setup

The ceramic and metallic catalysts were integrated in five commercial firewood stoves tested according to EN 13240 standard [13]. All stoves represent state-of-the art technologies and were selected and supplied by the manufactures. They were not used for combustion experiments described in previous chapters. The main technical characteristics of the roomheaters are summarized in **Table 21**.

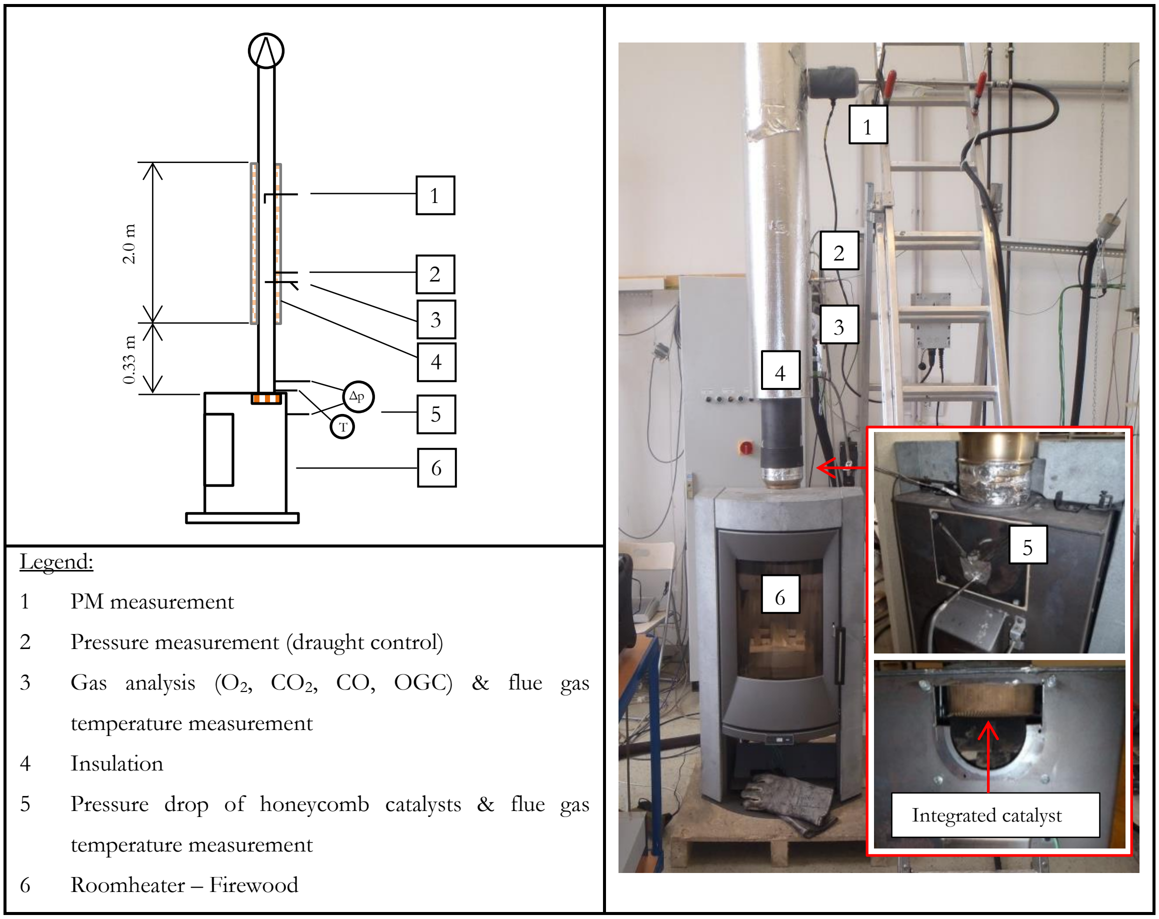
Table 21: Overview of relevant data of used firewood stoves (EN 13240) for catalyst integrated testing

Parameter	Stove A	Stove B	Stove C	Stove D		Stove E	
Nominal heat output	8 kW	10 kW	8 kW	8 kW		6.5 kW	
Mass	265 kg	215 kg	105 kg	280 kg		90 kg	
Flue gas outlet diameter	0.13 m	0.15 m	0.18 m	0.13 m		0.15 m	
Year of production	2012	2012	2012	2013		2014	
Adjustment of air inlet flap settings	manually – one stageless adjustable air valve	manually – one stageless adjustable air valve	manually – one stageless adjustable air valve	automatically controlled		manually – one stageless adjustable air valve	
Primary air	through the grate	from the front	from the front	through the grate		through the grate	
Secondary air	from the back & window flushing air	from the back & window flushing air	from the back & window flushing air	only as window flushing air		from the back & window flushing air	
Volume and dimensions of combustion chamber	0.0312 m³ (0.33 m width, 0.27 m depth, 0.35 m height)	0.0433 m³ (0.35 m diameter*, 0.45 m height	0.0616 m³ (0.44 m width, 0.35 m depth, 0.40 m height)	0.0312 m³ (0.33 m width, 0.27 m depth, 0.35 m height)		0.0270 m³ (0.30 m width, 0.30 m depth, 0.30 m height)	
Material of combustion chamber	Grey cast iron and fire clay	Grey cast iron and fire clay	Grey cast iron and vermiculite	Grey cast iron and fire clay		Grey cast iron and fire clay	
Integrated honeycomb catalyst	cc-round	cc-round	mc-round	cc-round	mc-round	cc-round	mc-round
Integration position	before flue outlet	in post combustion chamber	before flue outlet	before flue outlet		before flue outlet	
Effective catalytic volume ($V_{c.eff}$)	677 cm³	831 cm³	889 cm³	677 cm³	677 cm³	677 cm³	677 cm³
Coated area of carrier ($A_{cat.}$)	0.420 m²	0.515 m²	1.086 m²	0.420 m²	0.827 m²	0.420 m²	0.827 m²

* round combustion chamber

Each stove was adapted by integration of a ceramic or metallic oxidizing honeycomb catalyst in post combustion chamber or just before the flue outlet (**Table 21**, **Figure 51**). Stove A, B and C were tested with only one type of honeycomb catalyst integrated. Stove D and E were tested with both types of honeycomb catalysts integrated.

In each case, the catalyst integration was done without any flue gas bypass (**Figure 51**). The pressure drop induced by the integrated catalyst was continuously monitored. According to EN 13240 [13], the diameter of the measurement section was 150 mm for all stoves. After the flue outlet 0.33 m of the flue gas pipe was not insulated. Gas analysis was carried out 0.60 m downstream the flue outlet. Flue gas temperature was monitored by a thermocouple, type K, centrally placed in the flue gas pipe. The flue gas draught was measured 0.1 m downstream the gas analysis. The measuring point for PM measurement was located 1.5 m downstream the flue gas outlet.



Legend:

1 PM measurement
2 Pressure measurement (draught control)
3 Gas analysis (O_2, CO_2, CO, OGC) & flue gas temperature measurement
4 Insulation
5 Pressure drop of honeycomb catalysts & flue gas temperature measurement
6 Roomheater – Firewood

Figure 51: Scheme of test set-up (left) and example picture of stove D (right)

5.2.4 Testing procedure and measurements

For each combination of firewood stove and the respective type catalyst three different test cycles were conducted:

- Test cycle 1: Test run without an integrated catalyst or dummy.
- Test cycle 2: Test run with an integrated dummy in order to guarantee equal pressure drop conditions compared to the integrated catalyst solution.
- Test cycle 3: Test run with an integrated catalytic converter.

The testing procedure was in principal equally as described for the DemoCat tests (see chapter 4/ [4]). Therefore, only the most relevant characteristics of the testing procedure and measurements are mentioned. Each tests cycle consisted of five consecutive batches, starting from cold conditions with the ignition batch (batch 1). The draught in the chimney system was constantly controlled at 12 ± 2 Pa during the whole test cycle. The fuel mass provided for each batch was measured by a scale (Type: Sartorius AW-8201, accuracy ± 0.1g). Gaseous composition was monitored continuously during the whole test duration. For measuring O_2, CO_2, and CO the gas analyzer NGA 2000-MLT4 was used. Organic gaseous compounds (OGC) were measured as total hydrocarbons (THC) based on CH_4 equivalents at 180 °C using a FID (M&A Thermo-FID PT63LT). The gas analysis equipment was calibrated before the combustion experiments of each stove. The pressure drop induced by the catalyst or dummy was monitored with the measuring device Thermokon DPT 2500-R8. Details about the measuring range, measuring principle and accuracy of measuring devices can be found in **Table 18** (see chapter 4.3.5). PM sampling was conducted gravimetrically by out-stack measurement in each batch as described in chapter 4.3.5.

For stove B quartz plane filters instead of stuffed filter cartridges were used for retention of particulate matter emissions in test cycle 2 and test cycle 3. These plane filters were used for chemical analysis in order to assess potential catalytic effects on the chemical composition of PM emissions. Before the preconditioning at 130 °C the plane filters were heated at 400 °C for 4 h and subsequently cooled down in a desiccator. In the desiccator a sample of distilled water was located in order to prevent the adsorption of organic material of ambient air on active points of the quartz filters. The share of elemental carbon (EC), organic carbon (OC) and total carbon (TC) was determined by thermo-optical analysis using a carbon/ hydrogen analyzer (Leco RC-612). Ions (Cl, NO_3, SO_4, Na, NH_4, K, Mg, Ca) were analysed by aqueous extraction and IC and ICP-MS.

The heating operation of the stoves was performed according to the specifications given in the user manual of the respective stove. The first fuel batch was lighted by top-down ignition technique (stove

A, B, C, D) or bottom-up ignition technique (stove E). As kindling material small pieces of spruce and two pieces of specific starting aids (wood fibers soaked with vegetable oil/ FLAMAX Eco-Lighter [118]) were used. Firewood pieces and kindling material was adjusted crosswise in the combustion chamber. The subsequent fuel batches (batch 2 – 5) were placed directly on the firebed. The adjustment of damper settings for combustion air supply was done manually, except for stove D which was operated by an automatically controlled combustion air supply device. During the ignition batch the air inlet flap settings of primary and secondary air were fully open (100%). After the ignition batch the air inlet flap settings were adapted to constant settings according to the specifications of the manual.

For stove A the fuel mass of one batch was 2.1 kg, for stove B 2.6 kg, for stove C 2.3 kg, for stove D 2.1 kg and for stove E 1.5 kg. For stove A, B, D and E the fuel mass of one batch was evenly distributed on three firewood pieces (length of firewood pieces: A, D and E: 0.25 m/ B: 0.33 m), for stove C the fuel mass of one batch was evenly distributed on two firewood pieces (length: 0.33 m). In addition to 0.5 kg of spruce kindling material (length: 0.25 m) the ignition batches were performed with four smaller pieces of firewood (stove A, C, D and E) or six smaller firewood pieces (stove B) with equal lengths as used in the consecutive batches.

5.2.5 Data evaluation and statistical analysis

The analysis of CO emissions was done according to EN 13240 standard [13], the analysis of OGC emissions according to CEN/TS 15883 standard [98]. The average emission concentration of test cycle (batch 1 to 5) was calculated and referred to 13 vol.-% O_2 (mg/m^3, STP, dry, 13 vol.-% O_2). For PM emissions a time-weighted average of 5 measurements, each of them lasted 30 minutes, of one test cycle was calculated.

Three different emission impacts on CO, OGC and PM emissions were determined based on the results of test cycle 1, 2 and 3. The first emission impact represent the primary effect, which was determined by comparing test cycle 1 (without catalyst/dummy) and test cycle 2 (integrated dummy) (**Eq. 6**).

$$PE = \frac{\text{E without catalyst or dummy} - \text{E dummy}}{\text{E without catalyst or dummy}} \times 100\% \qquad \text{(Eq. 6)}$$

PE	Primary effect; in %
E without catalyst or dummy	Emission concentration of test cycle 1 (batch 1 to 5); in mg/m^3, STP, dry, 13 vol.-% O_2

E dummy	Emission concentration of test cycle 2 (batch 1 to 5); in mg/m³, STP dry,13 vol.-% O_2

For determination of the emission impact due to catalytic emission conversion the average emission concentrations of batch 1 to 5 of test cycle 2 (integrated dummy – E dummy) and test cycle 3 (integrated honeycomb catalyst – E catalyst) were calculated according to **Eq. 5** (see chapter 4).

As third emission impact the net emission reduction rate was calculated which represents a combination of the catalytic conversion and the primary effect. The net emission reduction rate was calculated based on test cycle 1 and test cycle 3 (**Eq. 7**).

$$NERR = \frac{\text{E without catalyst or dummy} - \text{E Catalyst}}{\text{E without catalyst or dummy}} \times 100\% \qquad (\textbf{Eq. 7})$$

$NERR$	Net emission reduction rate; in %
E without catalyst or dummy	Emission concentration of test cycle 1 (batch 1 to 5); in mg/m³, STP, dry, 13 vol.-% O_2
E Catalyst	Emission concentration of test cycle 3 (batch 1 to 5); in mg/m³, STP dry,13 vol.-% O_2

The statistical significance of test results was evaluated by a Student´s t-test. The level of significance was defined at p values of < 0.05. Results were highly significant if p values of < 0.01 resulted by the statistical analysis. For further interpretations p values between 0.05 and 0.32 were defined as a trend for those tests. This approach corresponds to the range of one standard error which is a typical indicator for estimation of the validity of physical measurements.

In chapter 5.3 respective p values are given in brackets for each result of statistical analysis.

5.3 Results and discussion

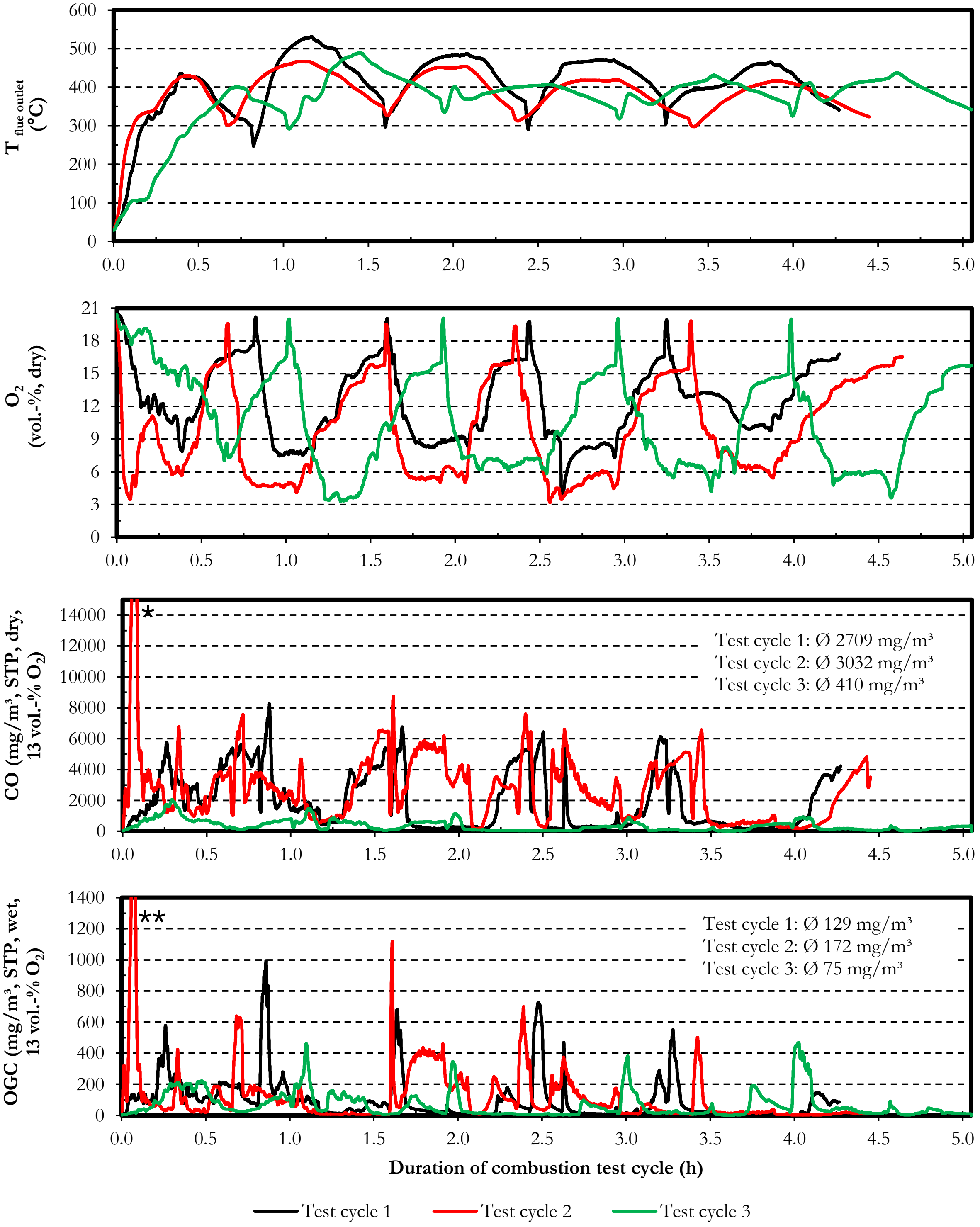


Figure 52: Example of measurements of flue gas temperature (downstream catalyst), O_2, CO, and OGC emissions of test cycle 1, 2 and 3 (Stove B). * CO peak: 22,000 mg/m³, STP, dry, 13 vol.-% O_2/ ** OGC peak: 1,600 mg/m³, STP, wet, 13 vol.-% O_2

Figure 52 illustrates an example of measurements of stove B for all three test cycles. It is obvious that the durations for the whole testing differed between the three tests. Test cycle 1 (without catalyst/ dummy) lasted 255 min, test cycle 2 (with integrated dummy) 267 min and test cycle 3 (with integrated catalyst) 299 min. (**Table 22**). Also the curves of O$_2$ were different, especially when comparing test cycle 1 (without dummy/catalyst) with test cycle 2 and 3 (with integrated dummy and catalyst). During test cycle 1 the O$_2$ values were in general higher compared to test cycle 2 and 3. As shown in **Table 22** the average O$_2$ concentration of test cycle 1 was higher at 12.3 vol.-% compared to 9.6 vol-% and 10.6 vol.-% of test cycle 2 and 3. The minimum O$_2$ values of the single batches of test cycle 1 were clearly higher at around 9 vol.-% (except batch 4) compared to test cycle 2 and 3 with minimum O$_2$ values at about 5 vol.-% (**Figure 52**). Flue gas temperatures measured downstream the catalyst/ dummy were highest for test cycle 1 for both, in the single batches as well as in average for the whole test cycle (**Figure 52**; **Table 22**). The highest CO and OGC emission concentrations were observed for test cycle 2 (with integrated dummy) at the start-up phases of single test batches. Moreover, test cycle 2 showed highest average CO and OGC emissions (CO: Ø 3032 mg/m^3; OGC: Ø 172 mg/m^3) compared to test cycle 1 (CO: Ø 2709 mg/m^3; OGC: Ø 129 mg/m^3) and test cycle 3 (CO: Ø 410 mg/m^3; OGC: Ø 75 mg/m^3).

The characteristics described for stove B (**Figure 52**) were typically for all tested stoves (stove A – E). In the following the main results for all stoves are given.

For all stoves, the average flue gas temperatures were different between test cycle 1, 2 and 3 (**Table 22**). The average oxygen concentrations were higher without integrated dummy or catalyst. Furthermore, the test durations with integrated dummies and catalysts were longer (**Table 22**, **Figure 52**).

Table 22: Relevant parameters indicating the primary combustion conditions of validation and evaluation test cycles

	Stove	**O$_2$ (vol.-%)**			**$T_{flue\ gas}$ upstream catalyst (°C)**			**$T_{flue\ gas}$ downstream catalyst (°C)**			**Duration of test cycle (min)**		
Test cycle		1	2	3	1	2	3	1	2	3	1	2	3
Ceramic honeycomb catalyst	A	12.2	11.4	11.7	n.d.	n.d.	n.d.	347	383	380	285	298	314
	B	12.3	9.6	10.6	n.d.	n.d.	n.d.	407	384	369	255	267	299
	D	13.1	12.4	12.3	329	353	346	319	334	343	322	333	377
	E	11.7	10.6	11.3	407	431	406	403	413	400	173	179	205
Metallic honeycomb catalyst	C	11.3	11.2	9.5	n.d.	n.d.	n.d.	420	392	396	255	260	290
	D	13.8	13.4	13.5	334	353	349	344	328	340	290	330	326
	E	11.7	11.3	11.2	407	406	407	403	388	400	173	205	216

n.d. – not determined

Highest CO and OGC emissions were observed for test cycle 2 at heating operation with the integrated dummy. These are indicators that primary combustion conditions were affected negatively by the integrated catalyst´s carrier geometry. However, the emission concentrations of CO and OGC of test cycle 3 were lower over the whole test duration with integrated honeycomb catalysts compared to test cycle 1 and 2 (**Figure 52**).

The average pressure drop of catalysts (test cycle 3) ranged between 4.5 Pa and 6.4 Pa. The pressure drops for the metallic honeycomb converter with a higher cell density of 7.750 cells/cm² showed slightly higher pressure drops compared to the ceramic catalyst (3.875 cells/cm²) (**Table 23**). The same results of pressure drops at similar space velocities and also the same trend of difference between the ceramic and metallic honeycomb converters were found in the previous DemoCat tests (**Table 19**/ [4]).

Table 23: Average results of batch 1 – 5 of test cycle 3 regarding the pressure drop induced by integrated honeycomb catalysts (Pa), flue gas temperatures up- and downstream of catalytic honeycomb converters (°C), the flue gas volume flow through the catalytic converter (m³/h, STP, w.b.) as well as space velocity (1/h)

Parameter	**Unit**	**Ceramic honeycomb catalyst**				**Metallic honeycomb catalyst**		
Stove	-	A	B	D	E	C	D	E
Pressure drop	Pa	4.5	n.d.	4.5	5.0	4.8	6.4	5.6
$T_{flue\ gas}$ upstream catalyst		n.d.	n.d.	346	406	n.d.	449	407
$T_{flue\ gas}$ downstream catalyst/ flue outlet	°C	380	369	343	400	396	340	400
Volume flow through catalyst	m³/h, STP, w.b.	20.0	24.4	20.9	19.1	21.2	21.1	18.2
Space velocity	1/h	29542	29085	30931	28142	23813	31167	26922
Coated area of carrier ($A_{cat.}$)	m²	0.420	0.515	0.420	0.420	1.086	0.420	0.827

w.b. = wet based/ n.d. – not determined

The average flue gas temperatures measured downstream the catalytic converters ranged from around 340 °C to 400 °C (**Table 23**). This temperature range was found as sufficient for catalytic conversion of CO and long-chained hydrocarbons with a Pt, Pd catalyst [62] [63]. Furthermore, the temperature conditions were similar compared to the previous presented DemoCat tests with the same types of catalysts (**Table 19**/ [4]) which consequently allow a comparison of conversion rates.

The emission impact comparing test results of test cycle 1, 2 and 3 of catalyst integrated solutions are illustrated in **Figure 53** which illustrates the evaluation of all tested stoves.

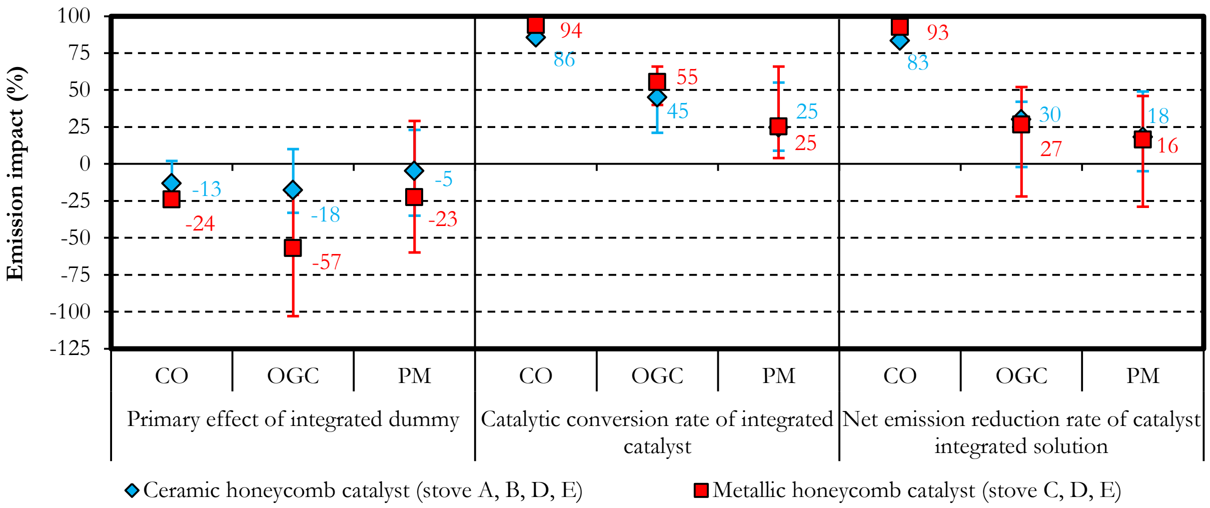


Figure 53: Average test results of tested stoves regarding the primary effect (left), the catalytic conversion rate (middle) and the net emission reduction rate (right). Calculations result from **Eq. 5** to **Eq. 7**. Error bars represent minimum and maximum values determined.

5.3.1 Primary effect

The primary effect (see **Eq. 6**) led in most cases to higher CO, OGC and PM emissions as illustrated by a negative emission impact (**Figure 53**, left section). For the integrated ceramic dummy the primary effect resulted in an emission impact in the range of -18% and -5%. For the metallic dummy the emission impact of the primary effect ranged from -57% to -23%. In both cases, the highest emission impact was observed for PM emissions, the lowest emission impact for OGC emissions. Consequently, lowest increase of average emissions due to the dummy integration was observed for PM, the highest for OGC emissions. Only in single cases the primary effect led to a decrease in emissions of test cycles with integrated dummies (error bars with positive emission impact > 0) (**Figure 53**). The primary effect is caused by the pressure drop which was induced by the honeycomb carrier geometry. This pressure drop influenced primary combustion conditions and the formation process of the emissions, i.e. combustion air supply conditions, turbulence and temperature. This was also confirmed by generally lower oxygen concentrations and longer test cycle durations for test cycle 2 and 3 with integrated dummy and catalyst (**Table 22**).

The statistical analysis using all batch results of test cycle 1 and 2 of all stoves with integrated ceramic or metallic honeycomb carriers showed a clear trend regarding a primary effect for CO (ceramic: $p = 0.21$; metallic: $p = 0.31$) and OGC (ceramic: $p = 0.31$; metallic: $p = 0.18$) emissions, but no trend for PM emissions (ceramic: $p = 0.88$; metallic: $p = 0.72$). However, it has to be mentioned that statistical analysis for single stoves revealed only a few trends for the primary effect for stove D (ceramic: CO: $p = 0.30$/ metallic: CO: $p = 0.25$; OGC: $p = 0.23$) and E (ceramic: PM: $p = 0.28$, metallic: CO: $p = 0.11$; PM: $p = 0.14$).

The results showed that the average primary effect is more pronounced for the metallic honeycomb carrier (**Figure 53**). This may be correlated with the tendency of a higher pressure drop of the metallic honeycomb catalyst (**Table 23**). Comparing all results of primary effects of the metallic and ceramic honeycomb carriers statistically, the primary effect for CO ($p = 0.14$) and OGC ($p = 0.24$) emissions was by trend higher for the metallic honeycomb carriers whereas no difference was found regarding the primary effect on PM emissions ($p = 0.58$).

The high variance of determined primary effects resulted most probably from appliance specific differences of the technology for combustion air supply and heat exchanger. This is similar compared to the draught tests described in chapter 3 where the tested appliances reacted differently towards increased draught conditions regarding gaseous emissions (**Figure 24**). Both, increased draught conditions or the integration of the dummy material are linked with different pressure conditions in the post combustion chamber which influence primary combustion conditions. Consequently, the primary effect of different stoves might be strongly deviating and is difficult to predict.

5.3.2 Catalytic conversion rate

The catalytic conversion rates (see **Eq. 5**) were by trend equally for both types of honeycomb converters (**Figure 53**, middle section). The highest average conversion rate was observed for CO (ceramic: 86%, metallic: 94%) followed by OGC (ceramic: 45%, metallic: 55%) and PM emissions (ceramic and metallic: 25%).

The results of average catalytic CO conversion rates were statistically significant for all tested stoves with integrated metallic or ceramic honeycomb catalysts. Except stove C ($p = 0.04$) the results achieved for CO conversion rates were even statistically highly significant ($p < 0.01$). A clear trend of a catalytic OGC emission conversion was observed for roomheater A ($p = 0.11$), B ($p = 0.10$) and E (ceramic: $p = 0.05$). For stove E, the OGC emission conversion rate for the integrated metallic honeycomb catalyst was even statistically highly significant ($p < 0.01$). For stove D, the only stove with automatically controlled combustion air supply, no trend of catalytic OGC emission conversion rate was found for both types of catalytic honeycomb converters. Except for stove A ($p = 0.05$) and C ($p = 0.28$), there was no trend of a catalytic PM emission conversion evident. But it has to be mentioned that average conversion rates of PM emissions were in a positive range up to more than 50% for both types of honeycomb catalysts (**Figure 53**).

The catalytic conversion results are in line with studies which were done with the same type of catalytic honeycomb converters [45] [65]. The catalytic conversion rates achieved for CO and OGC were similar [28] or even higher [57]-[59] compared to other studies where Pt and Pd supported catalytic systems were used as retrofit applications. Further, it has to be mentioned that in those studies potential primary effects of the carrier material were not respected separately and thus may reflect a net reduction rate of the respected emissions. Comparing both types of used honeycomb converters the CO emission conversion rate for the metallic honeycomb converter was in average 94 % for the 3 tested stoves. This result was significantly higher ($p = 0.02$) compared to the ceramic honeycomb converter with an average conversion rate of about 86% (**Figure 53**). Compared to the ceramic catalyst, the integrated metallic catalytic converter showed higher average OGC and PM emission conversion rates. However, statistical analysis showed neither significance nor a trend for a better conversion rate of the metallic catalytic honeycomb catalyst regarding OGC ($p = 0.41$) and PM ($p = 0.98$) emission conversion.

In general, the results of catalyst integrated systems of the metallic and ceramic catalytic converters fit very well to the results of the previous DemoCat combustion tests with similar space velocities of around 22,000 1/h (see **Figure 48** of chapter 4). However, the catalytic conversion rates observed during these tests compared to the catalytic conversion rates of the ceramic honeycomb catalysts (Stove A, B, D, E) were slightly lower for OGC emissions and clearly lower for CO emissions. This resulted from the higher cell density of the round ceramic honeycomb converter (3.875 cells/cm²/ 25 cpsi) in comparison to the rectangular ceramic honeycomb converters (2.480 cells/cm²/ 16 cpsi) used at these tests. This is confirmed by similar CO conversion rates (82%) when combustion tests with a similar amount of coated area of honeycomb carrier geometry (0.354 m²) are compared with conversion rates (86%) for the ceramic catalysts of stove A, B, D and E (coated honeycomb area: 0.420 m² – 0.515 m²) (**Figure 48**, **Figure 53**).

5.3.3 Net emission reduction rate

The net emission reduction rate (see **Eq. 7**) resulted from a combination of the primary and the catalytic effect. The net emission reduction effect is relevant for an assessment of real-life impact since potential negative primary effects towards emissions are respected. The average net emission reduction rate of stove A, B, D and E with integrated ceramic honeycomb catalyst was 83% (CO), 30% (OGC) and 18% (PM). For the metallic honeycomb catalyst the average net emission reduction rate for stove C, D and E was 93% (CO), 26% (OGC) and 16% (PM) (**Figure 53**, right section). For both types of honeycomb catalysts the CO net emission reduction was significant ($p < 0.05$) or even highly significant ($p < 0.01$) for all tested stoves. Except for stove D, a clear trend towards OGC net emissions reduction was observed. The net emission reduction of PM emissions was significant for stove A and by trend also for stove C (p = 0.17) and E (ceramic: p = 0.08; metallic: p = 0.1). The difference of net emission reduction between both types of honeycombs catalysts was significant for CO emission reduction (p = 0.02) but there was no trend for better OGC (p = 0.91) and PM (p = 0.95) emission reduction.

Overall, the experiments showed that catalyst integrated solutions of firewood stoves can achieve Ecodesign ELV of CO, OGC and PM emissions, not only considering the best operating batches but even the total heating cycle including also the ignition and preheating batch. This is pointed out in **Figure 54** where the average emission concentrations of test cycle 3 (with integrated catalyst) of different stoves are compared with Ecodesign ELV (CO: 1500 mg/m³, OGC: 120 mg/m³, PM: 40 mg/m³, all ELV referred to dry flue gases, STP conditions and 13 vol.-% O_2) [37]. As illustrated in **Figure 54** some appliances meet all the ELV clearly (e.g. stove A, B and C) whereas other appliances marginally exceed OGC and PM ELV (e.g. stove D and E). However, Ecodesign ELV refer to official-type-testing conditions which evaluate the appliance performance in heated-up and well-defined operating conditions (see chapter 7.3.1). Therefore, transient conditions, e.g ignition and preheating (batch 1 and 2) are principally not respected by official-type-testing. However, those transient conditions were considered as close-to-real-life evaluations in **Figure 54**. Consequently, the performance of catalyst integrated solutions regarding low emissions is higher rated compared to official-type-test results.

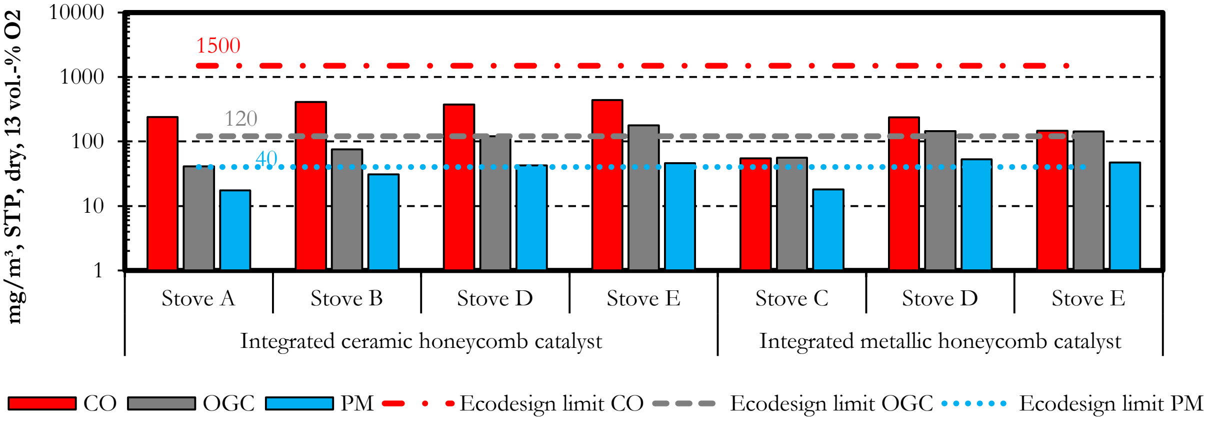


Figure 54: Average emissions of test cycle 3 measured with catalyst integrated solutions of integrated ceramic and metallic catalytic honeycomb catalysts. Dashed lines represent ELV of Ecodesign requirements for firewood stoves coming into force in 2022.

5.3.4 Catalytic effect on PM emission composition

As mentioned in chapter 5.2.4 quartz plane filters instead of stuffed filter cartridges were used for PM measurements. These measurements were carried out during test cycle 2 and 3 (stove B, cc-round). For a first assessment of potential catalytic effects on PM emission composition a chemical analysis of PM samples of the three test cycles was conducted. The filters were analyzed regarding the share of EC, OC, and ions.

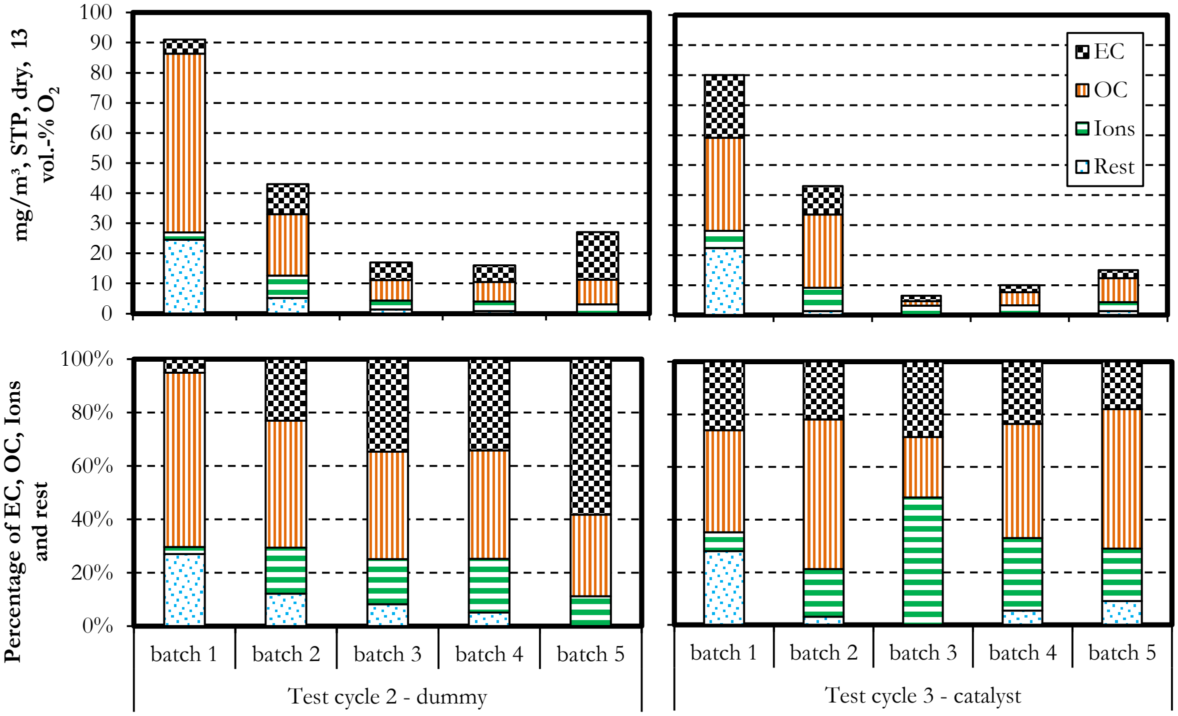


Figure 55: Comparison of PM measurement results and the PM composition of EC, OC, ions and rest with integrated dummy (left) and integrated catalyst (right) (Stove B).

The results are illustrated in **Figure 55** and showed that PM emissions with integrated ceramic catalyst were generally lower. A share of EC and OC of around 80% was observed for test cycle 2, whereas for test cycle 3 the share of EC and OC was lower at around 65%. But a clear catalytic effect on the PM composition was not evident, especially regarding the ratio of EC ($p = 0.86$) and OC ($p = 0.47$). Comparing the mass fraction on the filter also ions were not influenced by a catalytic effect. According to statistical analysis of comparison of the percentages of EC, OC and ions there was a trend towards a higher share of ions due to catalyst integration ($p = 0.22$). This confirmed that EC and OC were reduced in test cycle 3 compared to test cycle 2. However, based on these results, there was no clear selectivity found out towards either more catalytic OC or EC conversion. As the numbers of analysis are low there should be more measurements to clarify the catalytic effect on PM emission composition (see chapter 9).

5.4 Summary – Emission impact of integrated oxidizing honeycomb catalysts

The DemoCat tests (chapter 4) identified the space velocity and the coated area of honeycomb carriers together with the temperature conditions as key parameters for the integration design and the emission conversion potential. Furthermore, the synergetic effect of primary optimization and catalyst integration was illustrated by the high catalytic conversion rates during the start-up and burn-out phases of a firewood batch.

In chapter 5 a clear emission reduction potential for firewood stoves by integrating ceramic or metallic honeycomb converters (EnviCat® – Long Life Plus) was confirmed by close-to-real-life test cycles. Depending on the type of honeycomb catalyst, CO emissions are reduced by 93% (metallic) or 83% (ceramic), OGC emissions by about 30% and PM emissions by about 20%. Consequently, the ELV for CO, OGC and PM emissions which were set during the Ecodesign and Energy labeling process of the European Commission (CO: 1500 mg/m³, OGC: 120 mg/m³, PM: 40 mg/m³, all ELV referred to dry flue gases, STP conditions and 13 vol.-% O_2) were met by most of the tested catalyst integrated solutions even when the ignition and preheating batch were taken into account.

The integration design of the honeycomb carrier geometry resulted in changed pressure conditions at the combustion chamber and influenced combustion conditions. In most cases, this so called primary effect had a negative impact on emissions and therefore needs to be considered during the development process of integrated catalytic solutions. Accordingly, it is suggested to develop catalyst integrated solutions in three steps – 1. Integrate a dummy, 2. Doing the primary optimization and defining the operational heating aspects (e.g. adjustment of air valve settings) and 3. Change the dummy against the catalyst (see also chapter 1.4).

Compared to other studies using catalytic systems as retrofit applications, catalyst integrated solutions, showed higher CO and OGC conversion rates. Beside the possibility to consider primary effects, a further advantage of catalyst integration compared to catalyst retrofit applications is the elevated temperature level at the catalyst surface. This leads to a higher catalytic conversion rate, especially for catalytically convertible OGC emissions.

Further, a sufficient temperature level is guaranteed for an oxidation of deposited soot (EC) and organic compounds (OC) on the catalyst surface. However, no selectivity towards either more reduction of EC or OC was indicated.

Concluding, the potential applicability of catalyst integrated solutions for emission reduction of firewood stoves at real-life operating conditions was confirmed. Still open topics are the long term stability of the catalytic conversion and the sensitivity of catalyst integrated solutions against off-specification heating operation which is evaluated in the following chapter.

6 APPLICABILITY OF OXIDIZING HONEYCOMB CATALYSTS – LONG TERM DURABILITY AND SAFETY ASPECTS[8]

Ceramic and metallic types of honeycomb catalysts convert gaseous and particulate emissions as shown in the two previous chapters. However, the applicability of catalyst integrated solutions in real-life operation highly depends on aspects of safety and long term durability. Those topics were addressed by two different test series which are presented in this chapter.

In general, there are several safety aspects referring to the heating operation of firewood stoves. The most important aspect, which has to be guaranteed, is that no harmful flue gases can enter the installation room. Therefore, a minimum flue gas draught is necessary to enable a reliable conveyance of flue gases by the chimney system (**Figure 11**). Additionally, the risk of fire due to hot surface temperatures, either at the envelope of the stove or at the connecting flue gas pipe to the chimney has to be considered during installation, especially when there are adjacent inflammable materials, e.g. wooden furniture or ground floor.

For that general safety risks, the EN standards for firewood room heating appliances, for example for firewood roomheaters (EN 13240, [13]) or residential biomass cookers (EN 12815, [15]), require specific aspects as well as specific safety tests to guarantee safe and reliable operation. Accordingly, the general premise of EN standards is that during normal operation no flue gases escape into the room and no embers fall out of the combustion chamber into the room. Furthermore, the materials which are used for the stove design shall not be harmful in any case and their types as well as thickness of materials shall meet certain minimum requirements. According to the EN standards (e.g. [13]) there are three different types of safety tests defined:

- Safety test for open operation
- Temperature safety test
- Natural draught safety test

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[8] Segments of this section have already been published in [3].

1) <u>Safety test for open operation</u> [13]

The "safety test for open operation" is only conducted for stoves which are intended for an operation without any combustion chamber door or with open doors. The test has to be carried out in heated-up conditions, typically after the performance analysis test (see **Figure 72**, chapter 7.2.4). Therefore, the draught settings are changed from 12 ± 2 Pa to 6 ± 1 Pa and a fuel batch mass corresponding to nominal load is fueled in the combustion chamber. During operation of at least 1 h no flue gases or burning fuel from the firebed shall leave the combustion chamber. In addition it has to be checked if there is a sufficient suction effect at the upper end of the combustion chamber, for example by using smoke cartridges.

2) <u>Temperature safety test</u> [13]

The "temperature safety test" is an obligatory test for each type of appliance. The test is conducted under increased flue gas draught conditions (+3 Pa). This means, for example, 15 Pa (tolerance +2/ -0) for a firewood roomheater with a nominal load of 10 kW. The stove is heated-up with the ignition and preheating batch. Subsequently the "temperature safety" test is performed. Therefore, a fuel batch of squared timber of fir (size: 4×6 cm or 5×5 cm) has to be used. The length of the squared timber is defined by the dimensions of the combustion chamber. However, the grate should be entirely covered by the fuel. The total mass of fuel is calculated based on the surface of the combustion chamber floor and the net calorific value of the test fuel. The air valve settings have to be adjusted to maximum settings, e.g. fully open. Then the fuel is burnt off and subsequently a new batch with the same amount of squared fir timber is conducted. This procedure is repeated until steady state is reached and the maximum temperatures do not increase any more. According to this safety test the stove shall not be damaged or deconstructed.

3) <u>Natural draught safety test</u> [13]

The "natural draught safety test" applies only to continuous burning appliances that are specified for operation at a chimney connected with more than one appliance. The appliance has to be installed on a balance and connected to a defined natural draught chimney system. Using an ignition and preheating batch the stove is heated-up. The preheating batch, which is defined as pretest, shall be carried out with a specific burning rate (for wood: 33±5%) of nominal load. The pretest at the specific burning rate is carried out until basic firebed is reached (at least 2 h of operation). Subsequently, a batch mass representing nominal load is fueled in the combustion chamber. The air valve settings for primary air supply shall be adjusted according to the minimum settings and for secondary air supply according to the manufacturer's instructions. The "natural draught safety test" is successfully passed by the appliance if the basic firebed is reached and the flue gas draught has not dropped below 3 Pa. If that is the case before basic firebed is reached the test is continued for further 10 h. The "natural safety test" is also successfully passed, if the total volume of CO emissions in the flue gas during the 10 h is below 250 dm³.

Based on the above mentioned safety tests the manufacturer has to define the minimum distances of the stove to adjacent combustible materials in the installation manual. Furthermore, in many European countries, e.g. Austria, Germany, Switzerland, the chimney sweeper has to approve each newly installed firewood room heating appliance before the initial operation. The chimney sweeper approves the installation of the heating system by checking, if the stove is classified according to the respective EN standard and meets potential further national requirements, e.g. testing of firewood roomheaters at part load for approval according to the Austrian 15a B-VG. Furthermore, the distances to adjacent combustible materials are checked as well as the leakage rate of the chimney system.

6.1 Approach

In the context of this chapter specific safety aspects which solely referred to the integration of the catalysts are analyzed. In detail, the risk of total blocking and subsequent impossible operation ability or operational problems due to particle agglomeration on the catalyst's surface were evaluated within the so called "safety tests". It is expected that the blocking of the integrated catalysts is principally a time consuming process. Therefore, an evaluation by the standardized safety tests which are defined by the above mentioned tests of the EN standard is not possible.

Furthermore, the potential decrease of catalytic conversion rate due to deactivation processes, e.g. thermal, chemical or mechanical deactivation (see chapter 4.1.4, **Figure 41**), resulting from long term operation was assessed by the "long term durability tests".

6.2 Materials and methods

6.2.1 Used fuel

Beech (“*Fagus sylvatica*”) firewood with an average length of 0.25 m according to ÖNORM EN 14961-5:2011 [92] was used for all combustion tests (**Table 24**). The firewood pieces and spruce kindling material (“*Picea abies*”) were derived from trees grown in the Austrian Province “Lower Austria”.

Table 24: Chemical properties of used firewood and kindling material

	Moisture* **(W)** (kg/kg)	**Net calorific value** **(H_u)** (MJ/kg, d.b.)	**Ash** **(a)** (g/kg, d.b.)	**Carbon** **(C)** (kg/kg, d.b.)	**Hydrogen** **(H)** (kg/kg, d.b.)	**Nitrogen (N)** (g/kg, d.b.)	**Sulfur** **(S)** (mg/kg, d.b.)	**Chlorine** **(Cl)** (mg/kg, d.b.)
Analysis standard	EN 14774-1:2009 [93]	EN 14775:2009 [94]	EN 14775:2009 [95]	EN 15104:2011 [96]			EN 1515289:2011 [97]	
Beech firewood (“*Fagus sylvatica*”)	0.12 – 0.15	17.73	8.6	0.472	0.0616	< 1.0	93	36
Spruce kindling (“*Picea abies*”)	0.095	18.29	8.6	0.487	0.0631	< 1.0	50	31

d.b. = dry base/ *as received

Both, firewood and kindling material was bought as ready-to-use products from the local firewood producer HOFEGGER REINHARD (A-3250 Wieselburg). It was stored covered outside until the respective combustion tests were conducted.

6.2.2 Oxidizing honeycomb catalysts

The same types catalysts as already described in chapter 5 were used (see chapter 5.2.2). Accordingly, two different types of oxidizing honeycomb catalysts, both of the product line “EnviCat®-Long Life Plus”, supplied by the company CLARIANT were used [125] (**Table 25**).

Table 25: Physical properties of used types of honeycomb catalysts

Parameter	**Unit**	**EnviCat®-LongLife Plus Ceramic**	**EnviCat®-LongLife Plus Metal**
Cell density	c/cm²	3.875	7.750
	cpsi	25	50
Diameter	cm	14.4	14.9
Dimensions (Depth – d)	cm	5.1	5.1
Abbreviation	-	cc-round	mc-round

The first catalyst based on a ceramic carrier material with quadratic cells, the second catalyst based on a metallic carrier with trapezoid cells. The coating was equal for both types of catalysts and based on platinum (Pt) and palladium (Pd) on aluminum oxide (Al_2O_3). Two ceramic and two metallic catalysts were used (**Table 25**).

6.2.3 Stoves

Two different firewood stoves classified according to the standard EN 13240 [13] were used (**Table 26**).

Table 26: Overview of relevant data of used stoves and type of integrated honeycomb catalysts

Parameter	**Stove A**		**Stove B**	
Nominal heat output	8 kW		6.5 kW	
Mass	280 kg		90 kg	
Flue outlet	13 cm		15 cm	
Year of production	2013		2014	
Adjustment of air inlet flap settings	automatically controlled		manually – two stageless adjustable air valves	
Primary air	through the grate		through the grate	
Secondary air	only as window flushing air		from the back & window flushing air	
Volume and dimensions of combustion chamber	0.0312 m^3 (0.33 m width, 0.27 m depth, 0.35 m height)		0.0270 m^3 (0.30 m width, 0.30 m depth, 0.30 m height)	
Material of combustion chamber	Grey cast iron and vermiculite		Grey cast iron and vermiculite	
Integrated catalytic converter	cc-round	mc-round	cc-round	mc-round

Both roomheaters were also used for the evaluation of the emission impact of both types of honeycomb converters (see chapter 5: **Table 21** – Stove D and E). They are commercially available and represent commonly used stoves in terms of air staging and combustion chamber design. Stove A was a heavy stove providing heat storage stones on the top and at both sides outside of the combustion chamber whereas stove B was a light stove consisting of a steel envelope. Stove A was equipped with an automatic control device for combustion air supply. Hence, the combustion air supply is adapted by a control mechanism according to a temperature measurement in the combustion chamber. Thereby, the total combustion air flow is divided in two parts, primary air and secondary air. The automatic control system adjusts the primary and secondary air supply using an actuator connected to two dampers. The combustion air supply of stove B was manually controlled by two dampers, one for primary air supply and the second for secondary air supply. Window flushing air of stove B was provided by two holes above the combustion chamber door. The amount of window flushing air is not controllable by the user. The honeycomb catalysts were integrated in the post combustion chamber, just right upstream the flue outlet of both stoves (**Figure 56**; **Figure 58**; **Figure 60**).

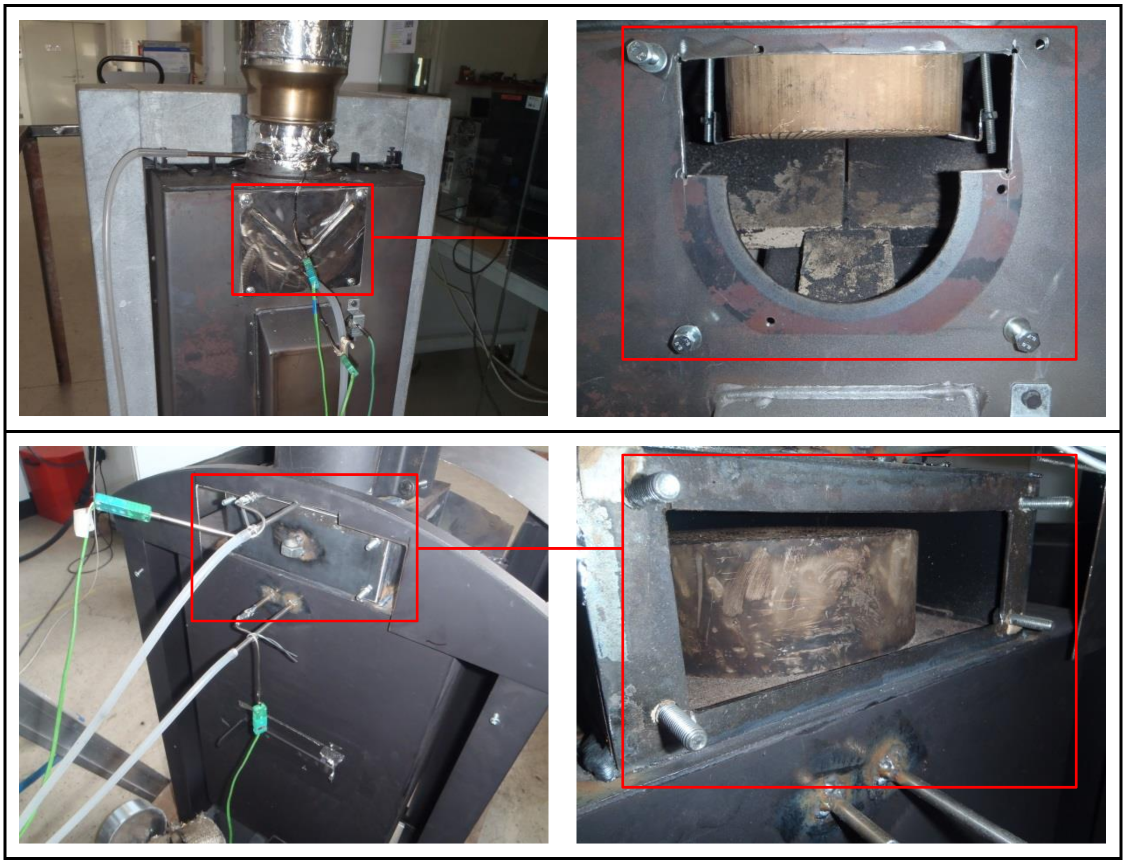


Figure 56: Integration positions of honeycomb catalysts for stove A (top 1 and 2) and stove B (bottom 1 and 2)

Therefore, two mounts were used for stove A in order to clamp the catalyst just before the flue outlet (**Figure 56**, top). For stove B a small box was constructed directly downstream the original flue outlet and the honeycomb converter was placed in this box (**Figure 56**, bottom). Between the honeycomb converters and the steel body of the stove a heating resistant gasket material was used. For both stoves there was no bypass for the flue gas for the total heating operation times. The open diameter for the flue gas passing through the honeycomb catalysts was 13 cm (diameter of flow cross-section area). Consequently, the effective catalytic volume was 677 cm^3 for both types of honeycomb converters which was identical to the integration design described in the previous chapter (**Figure 51**, see chapter 5.2.3).

6.2.4 Test procedure, experimental set-up and measurements

Safety tests

For assessing the effect of critical heating operation, 20 single ignition batches were carried out under natural draught conditions for each type of honeycomb catalyst integrated in stove A and B (**Figure 56**, **Figure 57**).

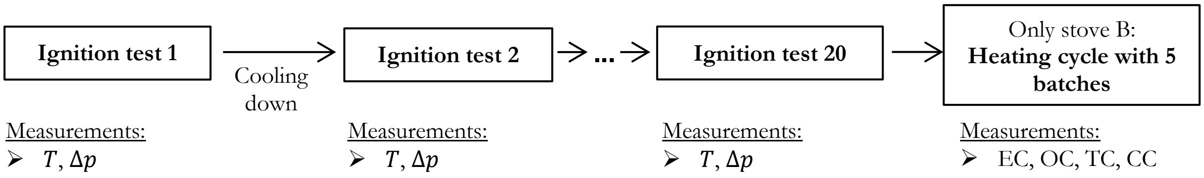


Figure 57: Test procedure of safety tests (stove A & B)

Since only one batch per heating cycle was performed, the flue gas temperatures were comparatively low and the stove itself was not at steady state and still heating up. Consequently, potential agglomerations on the catalyst's surface are not completely burnt-off and might eventually block the catalyst. This testing procedure emulates a critical operation mode which can occur in real-life, especially when the stove is used in summer times for only short burn [74].

For chemical analysis of deposited material a sample of agglomerated particles on the catalyst's surface was taken after the 20th batch (only for stove B). Subsequently, a heating cycle of five consecutive batches was performed and followed by a second sampling of agglomerated particles on the catalyst's surface. The samples were taken manually from the clogged cross-section area of the honeycomb catalyst. Thereby, the deposits were scraped onto a quartz plane filters which were conditioned before the sampling at 130 °C for 4 h and subsequently cooled down in a desiccator. The samples were analyzed regarding elemental carbon (EC), organic carbon (OC), total carbon (TC), and carbonate carbon (CC). The analysis was carried out by thermo-optical method using a carbon/ hydrogen analyzer (Leco RC-612). Thereby, sample is inserted into a heated quartz tube. Released carbon containing compounds are oxidized to CO_2, which is selectively detected by infrared cells. By choosing appropriate temperatures and carrier gases in the quartz tube TC as well as the fractions of OC, EC and CC can be distinguished. Carbon released in a temperature window from 200 °C to 600 °C under inert atmosphere is assigned to OC, carbon released between 600 °C and 900 °C is assigned to CC, carbon after switching to oxidizing conditions to EC [126].

The stoves were connected to a double walled masonry chimney of fireclay (Ahrens W3G). The flue gas was conveyed by a ceramic flue gas pipe with a diameter of 0.15 m. The total height of the chimney was 9.5 m, the effective height was 6.2 m (**Figure 58**).

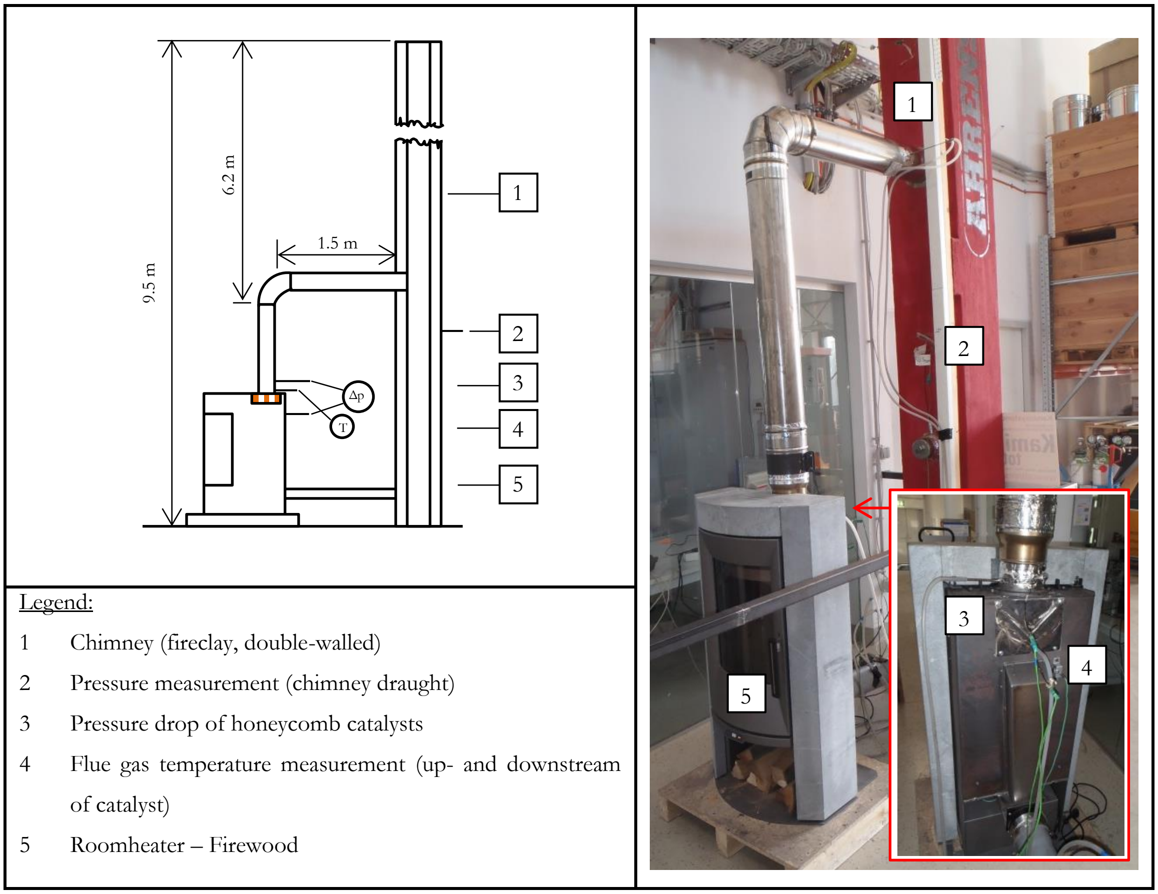


Figure 58: Experimental set-up of safety tests and example picture of stove A

The chimney system offers the possibility to intake combustion air for the stoves from outside via the annular gap between the chimney wall and the flue gas pipe. This is essential for using roomsealed appliances in modern and airtight houses. For stove A, classified as roomsealed by manufacturer, the combustion air supply was provided by the annular gap. For stove B, which was no roomsealed appliance, the combustion air supply was provided directly from the installation room. Hence, the connection to the annular gap was sealed for the experiments with stove B. Potential particle agglomeration and blocking was assessed by monitoring the pressure drop (Thermokon DPT 2500-R8) induced by the catalyst over the whole test duration and by visual inspection of the catalyst's surface at the end of the test cycle after the last ignition batch. Additionally, the flue gas temperatures before and after the catalyst were measured (Thermocouple, type K, class 1).

Between the single ignition tests at least five hours for cooling down were maintained to enable cold conditions for each ignition test. The ignition batch was lighted according to the user manual using the top-down ignition technique. For stove A three pieces of firewood (beech, total mass of 3.0 kg) were placed on the grate and several pieces of kindling material (spruce, total mass of 0.4 kg) were placed cross-wise on top of the firewood pieces. Two specific starting aids (wood fibers soaked with vegetable oil/FLAMAX Eco-Lighter [118]) located on the top of the kindling material were lit. The ignition batch for stove B was carried out according to the same procedure as described for stove A. For stove B three pieces of firewood (beech, 1.5 kg) and several pieces of kindling material (spruce, 0.4 kg) were used. The mass of fuel was determined by a precision balance (Sartorius AW-8201, accuracy ±0.1 g). Before each ignition test the ash of the test before was removed from the combustion chamber. The combustion air supply for stove A was adjusted according to the automatically controlled air supply, for stove B primary and secondary air inlet flap settings were fully open.

Long term durability tests

The intention of the safety tests was the evaluation of the robustness and applicability of integrated catalysts under off-specification heating operating conditions. Thereby, especially the risk of blocking the catalysts under "cold" conditions (only the ignition batch per heating operation) was evaluated. In contrast, the long term durability tests evaluated the stability of the catalytic conversion rate under long term operating conditions as well as the risk of blocking the integrated catalysts under "heated-up" conditions (5 – 11 batches per heating operation).

As illustrated in **Figure 59** the long term durability of both types of integrated catalysts was evaluated by 110 combustion batches at stove A.

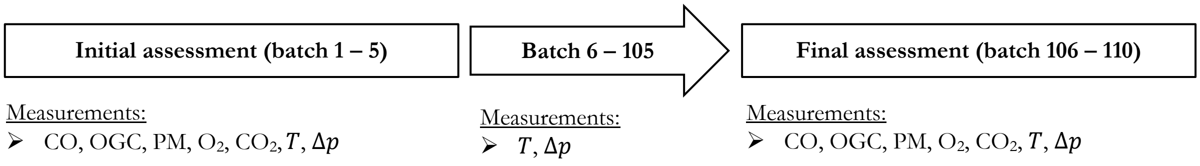
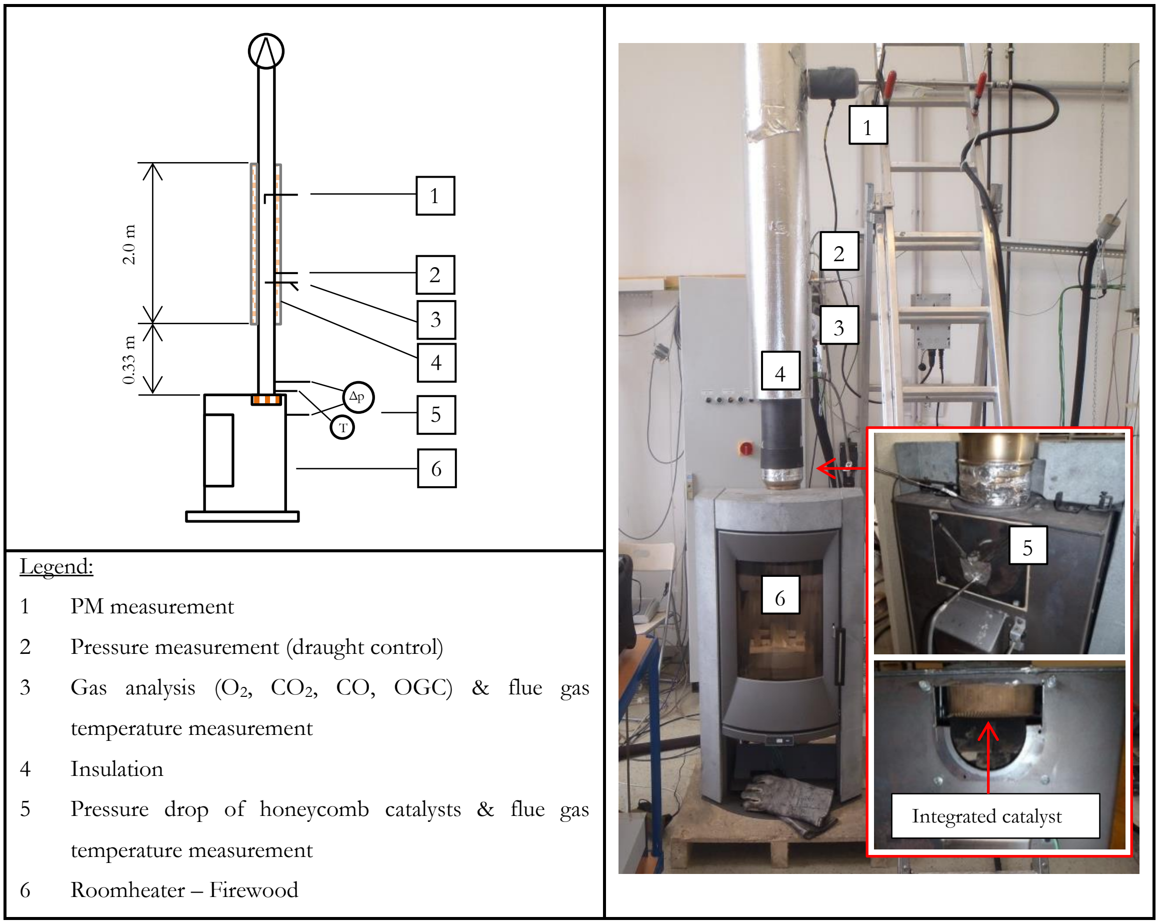


Figure 59: Experimental set-up of safety tests and example picture of stove A

By comparing the initial emissions (initial assessment test: batch 1 – 5) and the final emissions (final assessment test: batch 106 – 110) the effect of long term use on catalytic emission reduction regarding gaseous (CO, OGC) and PM emissions was evaluated. Furthermore, the risk of blocking due to long term use was assessed by continuous monitoring of pressure drop induced by the honeycomb converter and by visual inspection of the catalyst before and after the long term tests.

The tests were performed under controlled draught conditions at 12 ± 2 Pa. The experimental setup followed in principle the design of EN 13240 [13] (**Figure 60**).



Legend:

1 PM measurement
2 Pressure measurement (draught control)
3 Gas analysis (O_2, CO_2, CO, OGC) & flue gas temperature measurement
4 Insulation
5 Pressure drop of honeycomb catalysts & flue gas temperature measurement
6 Roomheater – Firewood

Figure 60: Experimental set-up of long term durability tests and example picture of stove A

The diameter of the measurement section was 0.15 m. After the flue outlet 0.33 m of the flue gas pipe was not insulated. The gas analysis was carried out 0.60 m downstream the flue outlet. The flue gas temperature downstream the honeycomb catalyst was monitored by thermocouples (type K, class 1). The flue gas draught was measured 0.1 m downstream the gas analysis (Thermokon DPT 2500-R8). The measuring point for PM measurement was located 1.5 m downstream the flue gas outlet (**Figure 60**). The pressure drop induced by the catalyst was monitored over the whole test duration using a measurement interval of 1 min for both types of honeycomb converters (Thermokon DPT 2500-R8).

A typical operation day of the 100 batch time period (batch 6 – 105) covered seven to eleven successive batches. For the initial (batch 1 – 5) and final (batch 106 – 110) assessment tests a test cycle consisting of five consecutive batches were carried out. Gaseous flue gas compositions were measured continuously over the whole test duration. CO, O_2 and CO_2 was measured by a multi gas analyzer

(NGA 2000-MLT4), organic gaseous compounds (OGC) were measured by a FID (M&A Thermo-FID PT63LT). Particulate matter (PM) emissions were measured in each batch according to the "German and Austrian method" of CEN/TS 15883:2009 [98]. The PM sampling started three minutes after recharging a new fuel batch and was terminated after 30 min of sampling. The diameter of the PM sampling nozzle was 12 mm and a constant sampling rate of 0.6 m^3/h (STP: 273.15 K/0 °C; 101,325 Pa) was adjusted. For retention a stuffed quartz wool cartridge was used (3.5 g quartz wool per cartridge). The sampling system outside of the flue gas ("out-stack" measurement) was heated to a constant temperature of 130 °C in order to avoid condensation. For conditioning before and after measurements all samples were dried in a drying oven at 130 °C for 4 h and subsequently cooled down in a desiccator equipped with silica gel for at least 8 h. The conditioned unloaded and loaded filter cartridges were weighed on a precision balance (Type: Satorius ME 235P, accuracy ± 0.01 mg).

A combustion batch started right after loading the appliance and closing the door. The batch was terminated when the CO_2 content of the flue gas reached 25% of the maximum CO_2 peak of the respective batch. According to pretests this corresponded well with the recharging criteria of a maximum variation of firebed mass of ±0.05 kg according to EN 13240 [13]. Since there was no flue gas monitoring during the 100 batch heating operation recharging was conducted when the flames of the respective batch extinguished. This moment is typical for recharging in real-life heating operation as investigated by an user survey of Austria (see chapter 2: Question 4.6 of **Table 8**) and Europe [74].

The ignition batch was performed in the same way as it was done for the safety tests of stove A (see above). In contrast, the air valve settings were adjusted manually and were fully open during the ignition batch. For the other batches three pieces of beech firewood with a total mass of 2.1 kg were used respectively. The adjustment of combustion air supply was done manually at constant damper settings. The valve for primary air supply was around 10% of cross-section area of air intake open, the valve for secondary air supply was fully open (100%). According to the user manual the grate was fully open during the ignition batch. At recharging the second fuel batch the grate was closed manually and remained closed for the rest of the day of the heating operation.

6.2.5 Data evaluation and statistical analysis

Safety tests

Figure 61 shows exemplarily the measurements of an ignition test batch of the safety test series. The measurements covered the flue gas temperature up- and downstream the integrated catalyst, the chimney draught and the pressure drop induced by the integrated catalyst.

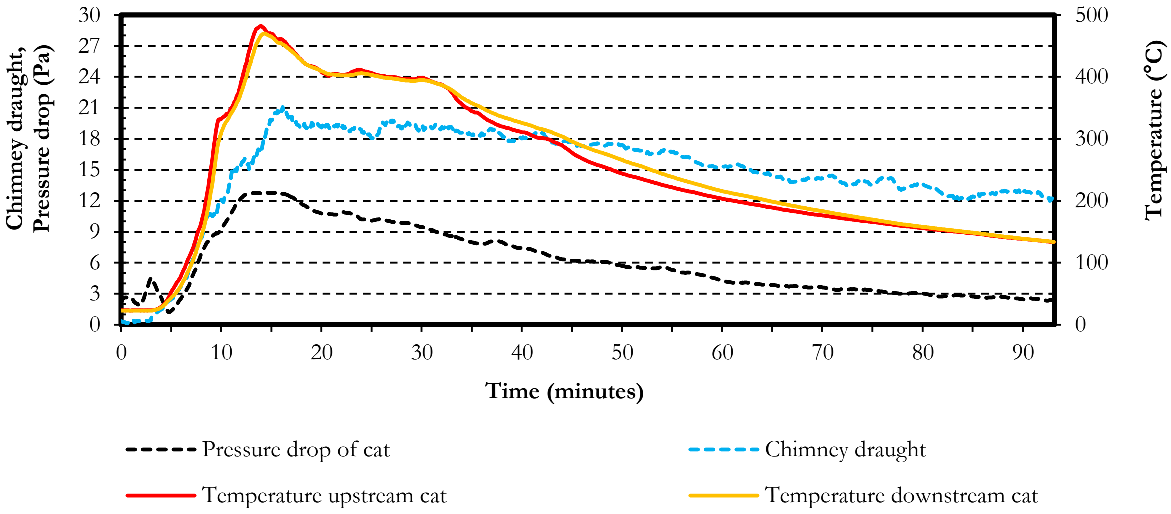


Figure 61: Example of safety tests of stove A with integrated ceramic honeycomb catalyst (cat)

As illustrated in **Figure 61** the chimney draught, flue gas temperatures and the pressure drop induced by the honeycomb catalyst increased after lighting the ignition batch. After a peak of temperatures at about 10 to 15 minutes after lighting, chimney draught and pressure drop curves decreased slowly. In general, the flames of the ignition batch extinguished at about 60 min after lighting. Due to the retained heat in the chimney system the chimney draught and the catalyst pressure drop decreased slowly after flames extinguished. Therefore it was decided to include a part of the cooling phase (~30 min) in the data analysis interval of 90 min. Following data were investigated for each ignition test (90 min):

- Maximum (T_{max}) and average ($\emptyset T$) temperature up- and downstream of the honeycomb catalyst
- Maximum ($\Delta p_{cat.\,max}$) and average ($\emptyset \Delta p_{cat}$) pressure drop induced by the honeycomb catalyst
- Maximum ($\Delta p_{max.\,flue\ gas}$) and average ($\emptyset \Delta p_{flue\ gas}$) chimney draught

The monotony of the course of pressure drop measurements and ignition tests was evaluated by the correlation coefficient Kendall's Tau (τ_k). The Kendall's Tau represents a statistical rank correlation coefficient which is used to analyze the ordinal association between two measured quantities. The statistical test for the Kendall's Tau determination refers to a non-parametric hypothesis test for statistical dependence. The Kendall's Tau and the significance of the correlation were statistically evaluated with the software SPSS (version number: 22). The confidence level for statistical significance was defined at 95% ($p < 0.05$). Highly significant results are characterized by p values < 0.01. A trend was defined for p values between 0.05 and 0.32, which is in the range of one standard error. In chapter 6.3 “Results and discussion” respective p values of Kendall's correlation coefficients are given in brackets for each statistical test.

For evaluation of the potential blocking of the catalyst by agglomeration of unburnt particles photographs of the catalyst were compared before and after the safety tests. Furthermore, for stove B the samples of deposited material on the catalyst surface were taken before and after the five batches heating cycle and analyzed regarding TC, EC, OC and CC.

Long term durability tests

The evaluation of CO emissions was done according to EN 13240 [13], the analysis of OGC and PM emissions according to the specification CEN/TS 15883 [98]. The average gaseous and particulate emission concentration of each assessment test cycle (batch 1 – 5 & batch 106 – 110) was calculated and transferred to 13 vol.-% O_2 (mg/m^3, STP, dry, referred to 13 vol.-% O_2). The effect of long term heating operation was assessed by comparing emissions of initial and final assessment tests. For evaluating the significance of differences of emissions measured at the initial and final assessment tests a Student's t-test was used. For statistical interpretation the same approach as described for the safety tests was applied.

In order to do a qualitative evaluation of the blocking of the catalyst by agglomeration of unburnt particles photographs of the catalyst before and after the long term test series were compared. Furthermore, the pressure drop induced by the integrated catalysts ($\Delta p_{cat}/\ \emptyset\Delta p_{cat}$) of the initial and final assessment tests were analyzed and compared.

6.3 Results and discussion

Safety tests

The safety tests evaluated the robustness and applicability of integrated catalysts under off-specification heating operating conditions. Thereby, especially the risk of blocking the integrated catalysts under "cold" conditions (only the ignition batch per heating operation) was evaluated. No interruption of the combustion tests, of the 20 ignition batches, was necessary for both stoves integrated with ceramic and metallic honeycomb catalysts. The complete firewood of the respective ignition test batch was combusted so that no unburnt residues remained on the grate. However, combustion conditions were insufficient for complete gas phase burn-out indicated by agglomerations of soot and tar at the vermiculite plates of the combustion chamber. Furthermore, the window of the combustion chamber was black after the safety test series. As illustrated in **Table 27** the temperatures up- and downstream the integrated honeycomb catalysts were similar for each stove. The average temperatures of safety tests of stove A for the ceramic and metallic honeycomb catalysts ranged between 256 °C and 260 °C (upstream) and between 257 °C and 263 °C (downstream). For stove B, the temperature level was slightly lower at average temperatures in the range of 218 °C – 228 °C (upstream) and 214 °C – 223 °C (downstream). The average chimney draught of the 90 min evaluation periods (**Figure 61**) ranged between 14 Pa and 16 Pa for stove A and between 19 Pa and 24 Pa for stove B. Concluding, the data of **Table 27** indicate in general similar operating conditions during the safety test series for each stove.

Table 27: Average results and standard deviations of 20 ignition test batches for stove A and B regarding characteristic operating conditions of safety tests

Stove	T_{max} (upstream cat)	$\emptyset T$ (upstream cat)	T_{max} (downstream cat)	$\emptyset T$ downstream cat)	$\Delta p_{cat.\,max}$	$\emptyset \Delta p_{cat}$	$\Delta p_{max.\,flue\,gas}$	$\emptyset \Delta p_{flue\,gas}$
	°C				Pa			
Stove A - ceramic honeycomb catalyst	396 ± 38	256 ± 13	379 ± 39	257 ± 11	16 ± 5	8 ± 2	30 ± 11	14 ± 1
Stove A - metallic honeycomb catalyst	409 ± 41	260 ± 11	403 ± 52	263 ± 11	19 ± 5	10 ± 3	26 ± 5	16 ± 3
Stove B - ceramic honeycomb catalyst	413 ± 28	228 ± 9	385 ± 19	223 ± 9	15 ± 5	7 ± 1	38 ± 15	19 ± 4
Stove B - metallic honeycomb catalyst	401 ± 36	218 ± 14	381 ± 31	214 ± 12	20 ± 10	8 ± 2	53 ± 31	24 ± 12

The chimney draught conditions for stove A which are mainly influenced by the flue gas temperatures [46], weather conditions [101] and the chimney design [49] were on average comparable at around 15 Pa (**Table 27**, $\emptyset \Delta p_{flue\,gas}$). For stove B a higher range of flue gas draught was measured due to windy weather conditions at single test days. In general, the pressure drop of the catalyst depends on the volume flow and the total open cross-section area of the catalytic system.

Consequently, a higher volume flow due to increased chimney draught results in a slightly increased pressure drop (**Figure 62**).

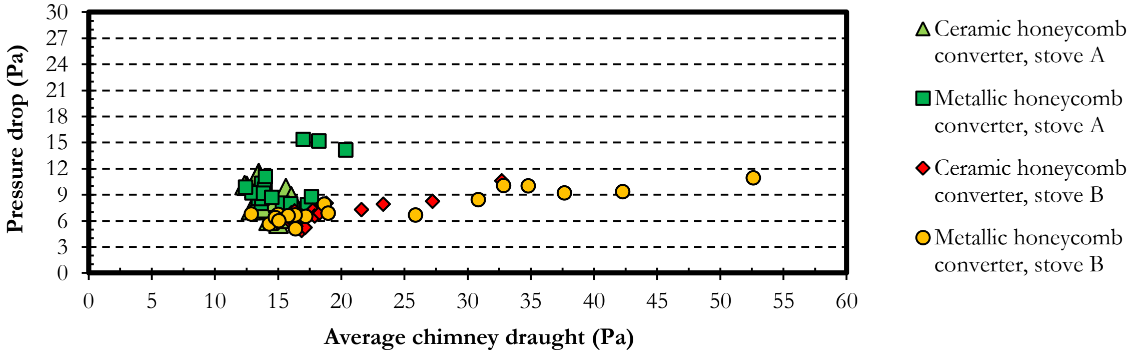


Figure 62: Correlation of average pressure drop and average chimney draught of analysis interval

Deposited material on the catalyst's surface are indicated, when the pressure drop is increased although draught conditions are equal. As illustrated in **Figure 63** the average pressure drop of combustion tests increased slightly with the number of batches performed for both types of honeycomb catalysts and both stoves during the safety tests.

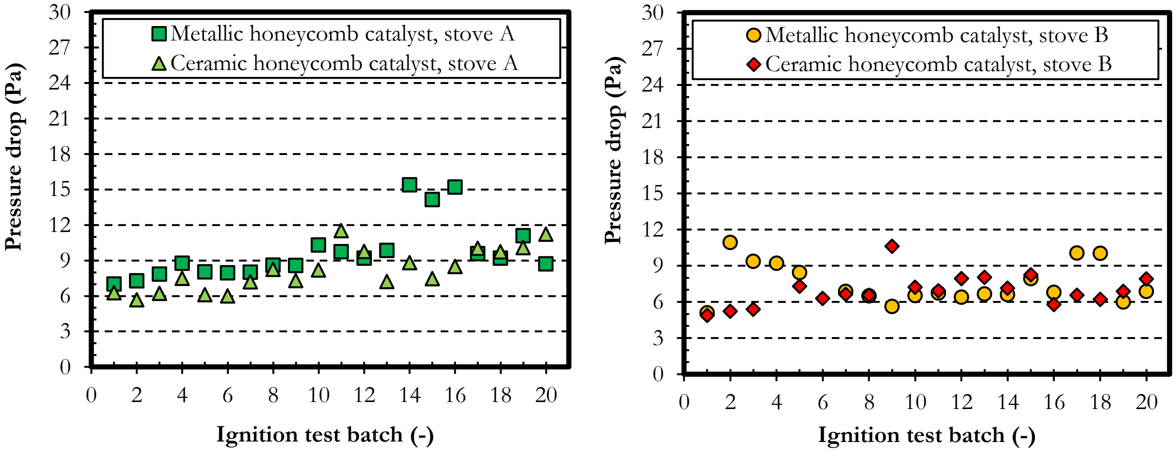


Figure 63: Average pressure drops induced by honeycomb catalysts integrated in stove A (left) and B (right).

For stove A, pressure drops of integrated ceramic and metallic honeycomb catalysts clearly increased over time of the safety test series from about 6 Pa to 10 Pa. This trend of increasing pressure drop over the number of ignition tests was also confirmed by statistical analysis which revealed a highly significant increase of average pressure drop for the ceramic ($p < 0.01$, $\tau_k = 0.61$) and metallic ($p < 0.01$, $\tau_k = 0.56$) honeycomb catalyst. The increased values of test batches 14 to 16 (**Figure 63**) are explainable by higher average flue gas draught compared to the remaining tests (**Figure 62**; **Table A8**).

The same behavior was observed for stove B (see **Table A9**; **Table A10**). For the ceramic honeycomb catalyst integrated at stove B, by trend, an increased average pressure drop at progressive ignition tests were observed ($p = 0.09$, $\tau_k = 0.28$). However, statistical analysis for stove B revealed neither significance nor a trend of correlation of increased pressure drop at progressive ignition tests for integrated metallic ($p = 0.89$, $\tau_k = -0.02$) honeycomb catalyst. The pressure drop was different at the beginning of safety tests (stove A: 6 Pa – 7 Pa, stove B: 5 Pa). This can be explained by the higher thermal heat output and consequently higher volume flow during combustion tests of stove A. Generally, the pressure drop measurements indicated deposited material on the catalyst's surface which was confirmed by visual analysis after the 20th batch of the safety tests (**Figure 64**; **Figure 65**).

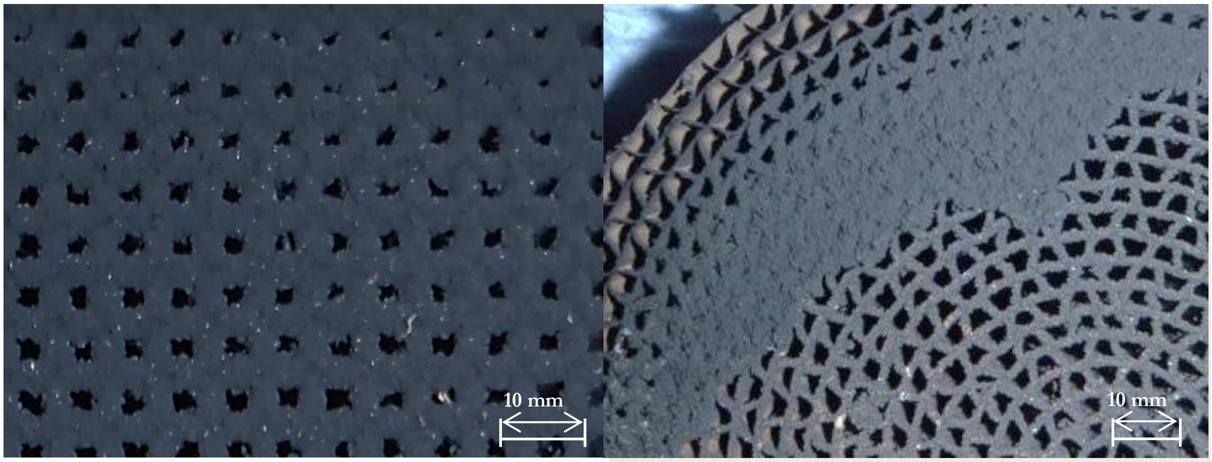


Figure 64: Close-up view of agglomerations deposited on the catalyst´s surface of the ceramic (left) and metallic (right) honeycomb converters after the 20th combustion test (stove A)

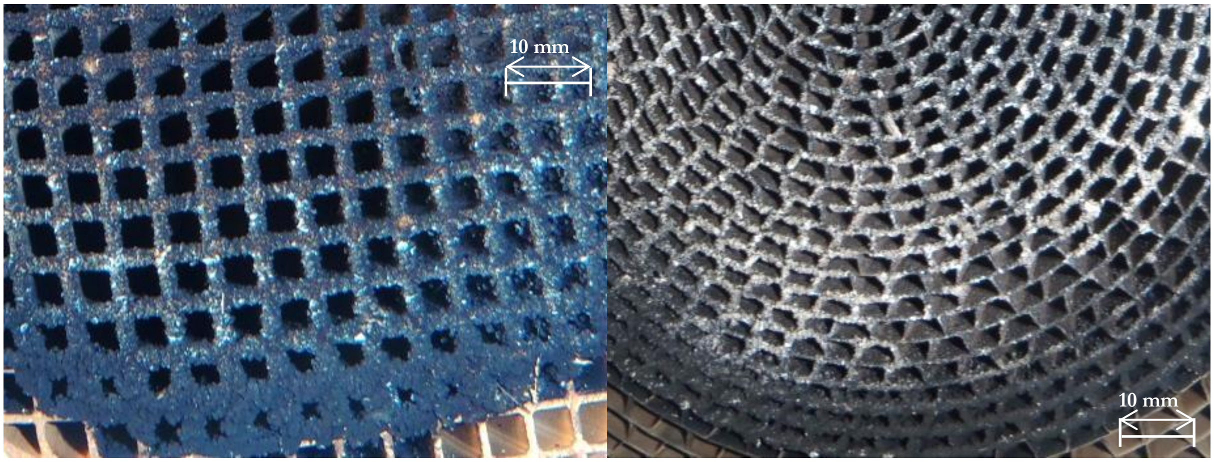


Figure 65: Close-up view of agglomerations deposited on the catalyst´s surface of ceramic (left) and metallic (right) honeycomb converters after the 20th combustion test (stove B).

For both stoves and integrated catalysts visual evaluation showed deposited agglomerations on the flow cross-section area of both catalysts after finishing the safety tests (**Figure 64**; **Figure 65**). However, in all cases the flow cross-section area of the honeycomb catalysts was not completely blocked after the ignition tests.

For stove A, more comprehensive agglomerations on the surface of the ceramic catalyst were evident compared to the metallic catalyst (**Figure 64**). For stove B, no clear difference in the amount of deposited material was observed for different types of honeycomb catalysts (**Figure 65**). Comparing the visual impression of both stoves it is obvious that stove A generally had more deposited unburnt material on the flow cross-section area of the catalysts. For stove A, totally blocked cells for both types of honeycomb catalysts were observed, whereas for stove B no cells were totally blocked. The blocking resulted most probably at areas where flue gas velocities were low due to the integration design of stove A. Beside low flow conditions enabling impaction and subsequent agglomeration of particles on the flow cross-section area of the honeycomb catalyst, thermophoresis and diffusiophoresis might be further mechanisms enhancing deposition and agglomeration of particles on the catalyst's surface [120] [121].

For stove B, the visual evaluation indicated that deposited material consisted of mineral (grey color) and carbonaceous particles (black color) [126] (**Figure 65**). The elemental analysis showed that after the safety tests the elemental composition of deposited particles consisted predominantly of EC and OC, whereas after the heating operation using five successive batches the combustible fractions of deposited agglomerations were burnt-off (**Table 28**). This was confirmed by visual evaluation which indicated significantly less agglomerations as well as only mineral agglomerations after the five batch heating cycle (**Figure 66**; **Table 28**).

Table 28: Results of chemical analysis of total carbon (TC), organic carbon (OC), elemental carbon (EC), carbonate carbon (CC) of deposited agglomerations after the safety tests and after a five batch heating cycle

Sample	**TC** (g/kg)	**OC** (g/kg)	**EC** (g/kg)	**CC** (g/kg)
Ceramic honeycomb catalyst after safety tests	400	120	280	< 30
Ceramic honeycomb catalyst after five batch heating cycle	< 50	< 50	< 50	< 50
Metallic honeycomb catalyst after safety tests	580	120	450	< 30
Metallic honeycomb catalyst after five batch heating cycle	< 50	< 50	< 50	< 50

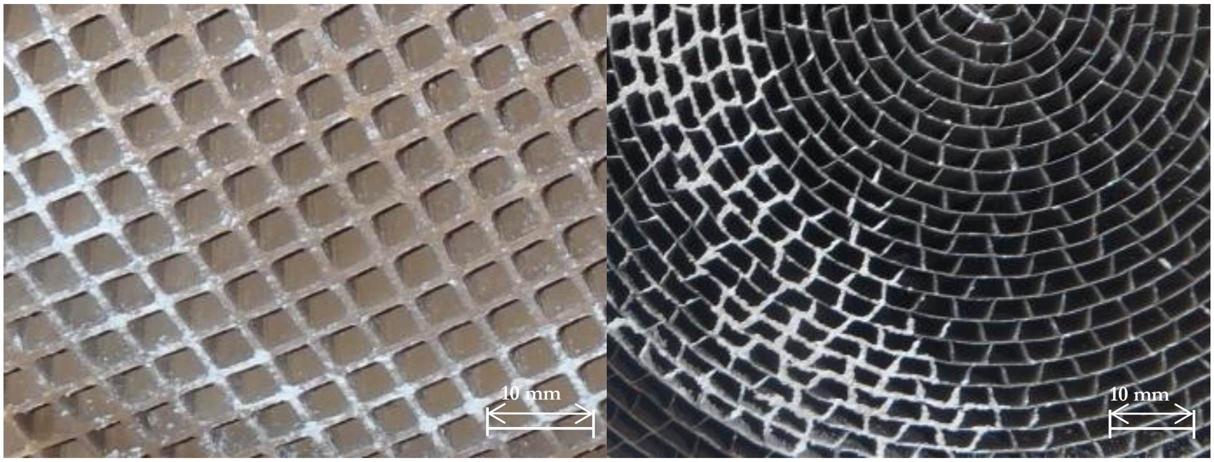


Figure 66: Close-up view of agglomerations deposited on the catalyst´s surface of ceramic (left) and metallic (right) honeycomb converters after the five batch heating cycle subsequent to the 20th batch of the safety test series (stove B).

In summary, the safety tests which covered in total 20 ignition batches showed that heating operation using only the ignition batch did not lead to a total blocking of both types of integrated honeycomb catalysts and did not cause any operational problems. Further, chemical analysis of deposited material confirmed that the combustible fractions can be burnt-off when a heating operation with five batches is performed.

6.3.1 Long term durability tests

The long term durability tests evaluated the stability of the catalytic conversion rate under long term operating conditions as well as the risk of blocking the integrated catalysts under "heated-up" conditions (5 – 11 batches per heating operation). The effect of long term heating operation on catalytic conversion rates was assessed by comparing emissions of initial and final assessment tests.

Table 29 shows relevant operating parameters of the initial and final assessment tests. Comparing the relevant operating characteristics of the initial and final assessment tests it was observed that the duration of the final assessment test was longer for both types of integrated honeycomb catalysts. Additionally, the average oxygen level as well as average flue gas temperature measured downstream of the catalyst was lower during the final assessment test (**Table 29**).

Table 29: Relevant operating parameters of initial and final assessment tests

Combustion test	**Initial assessment (batch 1 – 5)**			**Final assessment (batch 106 – 110)**		
	Duration (minutes)	O_2 (vol.-%)	T_{cat} (°C)	Duration (minutes)	O_2 (vol.-%)	T_{cat} (°C)
Ceramic honeycomb catalyst	268	12.4	376	292	12.2	369
Metallic honeycomb catalyst	261	11.6	398	270	10.3	381

The average flue gas temperatures just downstream the honeycomb catalysts ranged between 370 °C to nearly 400 °C. However, after the ignition batch there were phases of around 15 – 30 min in each batch when the flue temperature was higher than 400 °C [127]. Therefore, in contrast to the safety tests, the combustible fractions of deposited material can be oxidized. For the integrated ceramic honeycomb catalyst statistical analysis of batch results showed by trend longer batch durations ($p = 0.19$) for the final assessment test. For the metallic honeycomb catalyst the same trend was observed ($p = 0.12$). Additionally, there was a trend of lower oxygen values ($p = 0.07$) of the flue gas during the final assessment test. These aspects indicated deposited material on the catalyst's surface which resulted in different primary combustion conditions at the initial and final assessment tests.

This was further confirmed by comparison of the pressure drop measurements of both assessment tests. **Figure 67** illustrates the curve of pressure drop measurements during the initial and final assessment tests for both types of integrated catalysts (left: ceramic, right: metallic).

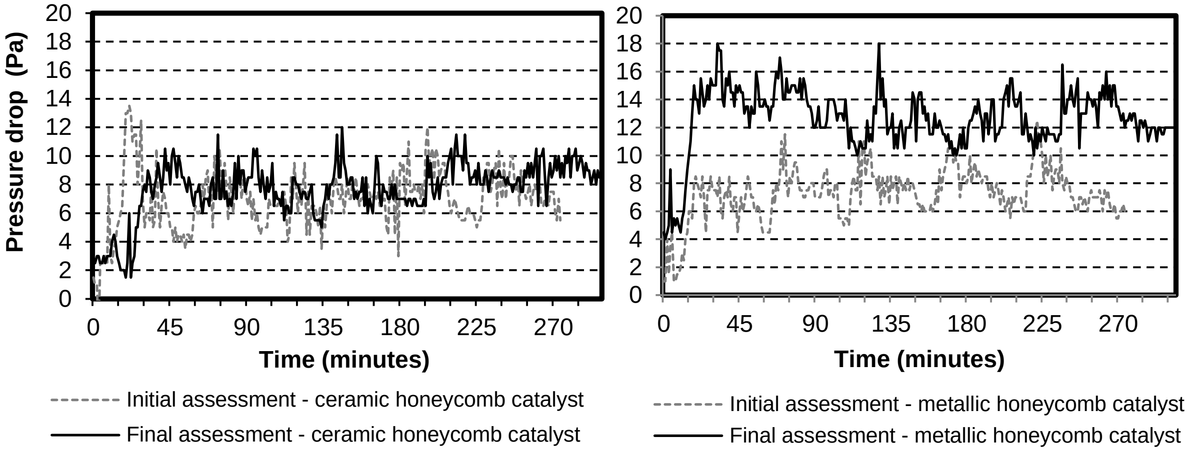


Figure 67: Pressure drop (Δp_{cat}) of integrated ceramic (left) and metallic (right) honeycomb catalysts at initial and final assessment tests of long term durability tests

The average pressure drop of the integrated ceramic honeycomb catalyst was 7.0 Pa at the initial assessment test and slightly increased to 7.7 Pa at the final assessment test. In contrast, there was a clear increase of pressure drop measured for the metallic honeycomb catalyst during the final assessment test compared to the initial assessment test from 7.3 Pa to 12.6 Pa (**Table 30**). The pressure drop measurements indicated a clear difference in the amount of deposited material on the catalyst's surface. The visual evaluation revealed deposited agglomerations for both types of catalysts which were equally distributed over the whole flow cross-section area (**Figure 68**).

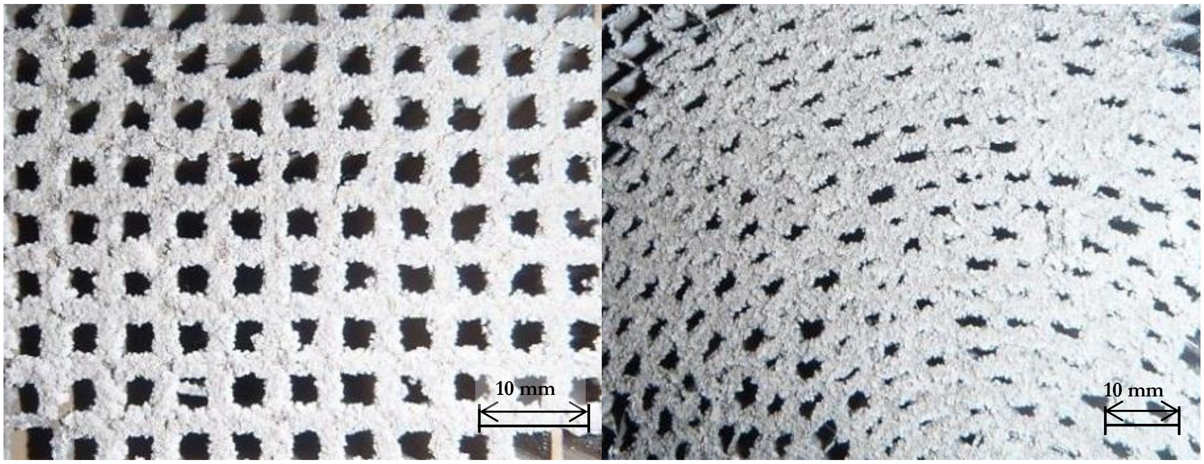


Figure 68: Close-up view of deposited agglomerations on integrated ceramic (left) and metallic (right) honeycomb catalysts after the final assessment tests

For the ceramic catalyst no totally blocked cells were observed whereas for the metallic honeycomb catalysts several cells were totally blocked by the deposited agglomerations. The color of the deposited material was grey instead of black as it was observed for the safety tests (**Figure 64**; **Figure 65**; **Figure 68**). This indicated that the agglomerations consisted predominantly of mineral particles [126]. The main reason for the higher sensitivity towards blocking is most probably the higher cell density of the metallic honeycomb catalyst and consequently smaller open cross-section area of single cells. Consequently, the free length of path for diffusion processes towards the cell walls is shorter which results in higher agglomeration rates and subsequently in an increased deposition. Although visual evaluation showed a significant amount of unburnt particles on the flow cross-section area of both integrated honeycomb catalysts no operating failure occurred during the long term durability tests although all experiments were performed without bypass.

Table 30 summarizes the average emission concentrations and pressure drop values of the initial and final assessment tests for both types of integrated honeycomb catalysts. Since the pressure drop for the metallic catalyst increased significantly during the long term operation the metallic catalyst was dismounted, cleaned with water and pressured air and integrated again. Subsequently, the final assessment test was repeated. The results of this test cycle (final assessment test – after cleaning) are also presented in **Table 30**.

The comparison of the initial and final assessment test cycle results regarding CO, OGC and PM emissions showed generally a different effect for gaseous and particulate emissions. Average CO emissions measured for the ceramic catalyst increased from 412 mg/m³ at the initial assessment test to 457 mg/m³ at the final assessment test. For the metallic catalyst CO emissions increased from

167 mg/m³ to 513 mg/m³, respectively. The same behavior was observed for OGC emissions. They increased from 106 mg/m³ to 120 mg/m³ for the ceramic catalyst and from 84 mg/m³ to 122 mg/m³ for the metallic catalyst. However, for PM emissions the trend was contrary. For both types of catalysts average PM emissions were decreased at the final assessment test. For the ceramic catalyst they decreased from 71 mg/m³ at the initial assessment test to 35 mg/m³ at the final assessment test, for the metallic catalyst they decreased from 54 mg/m³ to 34 mg/m³ (**Table 30**).

Table 30: Test cycle results of initial and final assessment tests for each integrated honeycomb converter. Effect of cleaning of metallic honeycomb catalyst when repeating the final assessment test (= Final assessment – after cleaning, batch 111-115)

Combustion test	**Initial assessment (batch 1-5)**				**Final assessment (batch 106-110)**				**Final assessment – after cleaning (batch 111-115)**			
	CO	OGC	PM	$\emptyset\Delta p_{cat}$	CO	OGC	PM	Δp_{cat}	CO	OGC	PM	$\emptyset\Delta p_{cat}$
	(mg/m³, dry, STP, 13 vol.-% O_2)			Pa	(mg/m³, dry, STP, 13 vol.-% O_2)			Pa	(mg/m³, dry, STP, 13 vol.-% O_2)			Pa
Ceramic honeycomb catalyst	412	106	71	7.0	457	120	35	7.7	-	-	-	-
Metallic honeycomb catalyst	167	84	54	7.3	513	122	34	12.6	227	82	42	7.7

For the ceramic honeycomb catalyst statistical analysis of differences of initial and final emissions showed neither a trend nor significance, even for PM emissions which were in average 50% lower compared to the initial assessment test. However, for the metallic catalyst the difference of CO emissions was a factor of three which was highly significant ($p < 0.01$). Furthermore, for OGC (increased by a factor of 1.45) and PM emissions (decreased by a factor of 0.63) a clear trend was observed (OGC: $p = 0.32$; PM: $p = 0.10$) for the differences between the assessment tests. Interestingly, PM emissions were lowest for both types of catalysts at the final assessment test although CO and OGC emissions were higher.

The reasons for the differences of emissions between the initial and final assessment test can be explained as follows. At the final assessment test primary combustion conditions were different and the catalytically active volume or the coated area of honeycomb carriers was reduced by mechanical deactivation due to deposited material [116]. Both aspects in combination led most probably to the increase of CO and OGC emissions. The lower PM emissions at the final assessment tests can be explained by an increased filtering effect which resulted from a smaller open cross-section area of single cells due to the deposited agglomerations (**Figure 68**). Consequently, the mineral fraction of particles seems to play a major role regarding the long term durability of integrated honeycomb catalysts although they are only of minor importance of total PM emission concentrations of firewood combustion in stoves [128].

In order to investigate the reversibility of potential deactivation mechanisms of long term operation the metallic honeycomb catalyst was cleaned with water and pressured air. Subsequently, the catalyst was reintegrated into the firewood stove and the final assessment was repeated (final assessment – after cleaning). The comparison of CO, OGC and PM emissions of the final assessment with the final assessment – after cleaning showed that CO and OGC emissions decreased form 513 mg/m^3 to 227 mg/m^3 and 122 mg/m^3 to 82 mg/m^3, and PM emissions increased from 34 mg/m^3 to 42 mg/m^3 after the cleaning process. Furthermore, the pressure drop measurements reached the initial level of 7.7 Pa (**Table 30**). For CO emissions statistical analysis still showed a trend of increased emissions ($p = 0.13$) whereas for OGC and PM no trend was observed comparing initial and final assessment test – after cleaning. Although it has to be mentioned that CO emissions of the final assessment test were in range of initial assessment test plus standard deviation of batch results, the increased CO emissions might be also an indicator for irreversible deactivation mechanisms. A possible decay of reversible catalytic conversion potential might result from mechanical deactivation by the loss of catalytic material or internal surface area. This results from attrition or crushing mechanisms (e.g. caused by the mineral particles) and the subsequent loss of catalytic material or internal surface area [116]. Further potential reasons for irreversible catalyst deactivation are poisoning of the catalyst or thermal degradation. However, the risk of poisoning was low since only natural wood with a low content of sulfur was used (**Table 24**), which is one of the most relevant poisoning elements for catalysts based on Pt and Pd [116]. Additionally, previous studies showed no thermal degradation for this type of catalyst even for temperatures up to 900 °C [63].

The 105 batches performed until the final assessment test, correspond to around 75 h – 100 h of heating operation. For example, in Austria a typical average use of firewood roomheaters was estimated at around 500 operating hours per year [83]. Consequently, a cleaning of the catalyst at least three to four times per heating season seems necessary to enable optimal conversion rates and guarantee safe operating conditions.

6.4 Summary – Long term durability and safety aspects of integrated oxidizing honeycomb catalysts

As presented in chapter 4 and 5 oxidizing honeycomb catalysts integrated in firewood stoves represent a step further towards low emission technologies. However, catalyst integrated solutions without bypass bear a safety risk due to blocking of the catalyst surface. This can occur either due to maloperating conditions by incorrect installation and user behavior or due to long term operation.

These aspects were addressed in this chapter (chapter 6) for both types of honeycomb catalysts (ceramic and metallic). The focus was on the evaluation of safety issues, e.g. pressure drop increase due to blocking by critical operating conditions (safety tests) and on the evaluation of the long term operation performance, e.g pressure drop increase due to blocking by long term operation (long term durability tests).

In general, the experiments showed that long term operation and critical operating conditions did neither lead to operational problems nor to break-offs of test series due to safety aspects. However, they clearly confirmed that unburnt particles can deposit and agglomerate on the catalyst surface and influence emission levels.

Deposited agglomerations at the blown cross-section of the catalyst surface were observed according to the safety test series. Therefore, the comparative low temperatures of less than 400 °C at the catalyst represent critical operating conditions and enhance particle agglomerations and the risk of an increased pressure drop. As shown by the chemical analysis, the agglomerated deposits sampled after the safety tests consisted predominantly of carbonaceous components, i.e. EC (120 g/kg) and OC (280 – 450 g/kg). The major share of agglomerated carbonaceous deposits was effectively oxidized with a heating cycle consisting of five consecutive batches. After that heating cycle the share of sampled deposited material on the catalyst surface of EC and OC was less than 50 g/kg. Consequently, it was confirmed that a sufficient heating-up of the integrated catalyst is needed to avoid deposition of carbonaceous agglomerations.

The long term durability of both types of catalytic converters was evaluated by totally 110 batches which correspond to around 75 h – 100 h heating operation. For both catalysts (ceramic and metallic) mineral agglomerations on the catalyst´s surface were observed. For the metallic catalyst the agglomerated deposits resulted in a significant increase of pressure drop (+5.3 Pa), CO (~300%) and OGC (~45%) emissions whereas PM emissions were decreased by 63%. However, cleaning the metallic catalyst with water and compressed air enabled the initial catalytic conversion rate and pressure drop.

For the ceramic catalyst long term operation did not result in significantly increased gaseous CO and OGC emissions but also led to decreased PM emissions. This is explainable by an increased filtering effect induced by the smaller open cross-section area of single cells.

Finally, the experiments showed that the metallic catalyst is in general better for emission reduction (see chapter 4 and 5) but more sensitive regarding blocking, especially by long term operation. Therefore, in terms of real-life applicability the ceramic honeycomb catalyst seems to be more suitable compared to the metallic honeycomb catalyst. But also for the ceramic catalyst a regular cleaning of about three to five times per year in real-life operation will be necessary to enable safe and efficient operating conditions. For the cleaning of the catalyst the use of water is suggested in order to remove water soluble components.

Technological add-ons monitoring the increase of pressure drop in combination with chimney draught conditions appear as a possible solution to signalize a cleaning demand. Further investigations on irreversible catalyst deactivation mechanisms, e.g. poisoning due to the use of sulfur rich material like litter etc. and quantification of lifetime as integrated solution are still necessary.

7 A NOVEL REAL-LIFE ORIENTED TEST PROTOCOL (*“beReal”*) VERSUS THE OFFICIAL-TYPE-TEST PROTOCOL (EN 13240)[9]

So far, it was shown how user behavior in real-life operation looks like, what are the effects of single operational aspects, e.g. the ignition technique, on emissions and thermal efficiency and the potential of integrated catalysts towards a significant emission reduction. However, a sustainable improvement of real-life heating operation is only achievable, if the user behavior and technological development is linked together. Therefore, it is necessary to evaluate technologies as close as possible to the realistic utilization at end-customers.

Consequently, as mentioned in the introduction of this thesis, new testing concepts – that better reflect the real-life behavior and are less endangered by manipulation and rooms for interpretation – are required by the European public administration [73]. It is expected, that such a real-life oriented test protocol would be capable to push technological innovation and development further. Moreover, it would enable a better differentiation of good and poor products for the end-customer. Another important aspect represents the discussions which are currently ongoing within the car industry (“diesel gate”) and which raise the probability that biomass combustion systems are faced with similar issues.

7.1 Approach

In a European R&D project, called *“beReal”* [129] such a new test protocol for firewood roomheaters (EN 13240 [13]) was developed under collaboration of R&D institutes, stove manufacturers, and industry associations. The new test protocol aims at an evaluation of the combustion performance regarding emissions and thermal efficiency under testing conditions close to real-life [130]. The “*beReal*” test procedure is based on the findings of different user surveys (chapter 2, [74]) and long-term field tests [88] to consider typical aspects of user behavior in the test concept. Specific operational aspects, for example, the ignition mode, the effect of flue gas draught (chapter 3) [2], air valve settings, and fuel characteristics [131], were assessed by laboratory tests. For firewood roomheaters a one-page user manual, called “Quick-User-Guide” (QUG) (**Figure 9**), was developed, which defines the specific

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[9] Segments of this section have already been published in [5].

operation mode for each appliance [88]. The QUG is the basis for heating operation during testing. Moreover, it should act as a user manual in real-life. The reproducibility and real-life relevance were demonstrated by a round robin test [132] and during a field test campaign [133], respectively.

In this chapter a comparative evaluation of the testing procedure and the emission and efficiency results of the novel test protocol *"beReal"* and the existing standard type test protocol is presented exemplarily with one firewood roomheater (EN 13240). Therefore, comparative combustion experiments according to both test protocols were conducted. EN test results were compared with official-type-test (ott) results. The results of CO, OGC and PM emissions were compared with future Ecodesign requirements which will come into force in 2022 in whole Europe. Fundamental differences of test procedures and data evaluation and their effect on the test results were identified. The repeatability of both methods was assessed and compared by calculating the coefficient of variation (CV). Finally, potentials and limitations of the *"beReal"* test protocol toward the effect on real-life evaluation are presented.

7.2 Materials and methods

7.2.1 Used fuel

Beech ("*Fagus sylvatica*") firewood with an average length of 0.33 m was used for all combustion tests (**Table 31**).

Table 31: Chemical properties of used fuel (chemical analysis included also bark)

	Moisture content* (W) (kg/kg)	**Net calorific value** (H_u) (MJ/kg, d.b.)	**Ash content** (a) (g/kg, d.b.)	**Carbon** (C) (kg/kg, d.b.)	**Hydrogen** (H) (kg/kg, d.b.)	**Nitrogen** (N) (mg/kg, d.b.)	**Sulfur** (S) (mg/kg, d.b.)	**Chlorine** (Cl) (mg/kg, d.b.)
Analysis standard	EN 14774-1:2009 [93]	EN 14775:2009 [94]	EN 14775:2009 [95]	EN 15104:2011 [96]			EN 15289:2011 [97]	
Beech firewood ("*Fagus sylvatica*")	0.16	18.18	8.3	0.485	0.0607	1110	95	9

*as received/ d.b. = dry base

The beech firewood of the ignition batch was split in small pieces for producing the kindling material. The firewood pieces derived from trees grown in the Austrian Province "Lower Austria". It was bought as ready-to-use products from the local firewood producer HOFEGGER REINHARD (A-3250 Wieselburg) and was stored covered outside until the respective combustion tests were conducted.

7.2.2 Combustion appliance

A commercial firewood roomheater, classified according to EN 13240 [13] and not used for any tests described in the previous chapters of this thesis, was used for the combustion tests [134]. **Figure 69** shows the used stove on the test bench. The combustion air supply is conducted to the stove by a central pipe and is delivered into the combustion chamber as primary, secondary, and tertiary air. Primary air is supplied via two vertical cleavages at the left and right corner of the combustion chamber. Secondary air is supplied via holes at the back wall of the combustion chamber and tertiary air enters the combustion chamber as window flushing air. An adaption of combustion air supply is possible manually with one leaver. Therefore, the proportion of primary, secondary, and tertiary air cannot be influenced by the user separately. The stove represents a modern state-of-the-art technology and was provided by the manufacturer. According to the manufacturers information this type of stove is very popular and one of the most frequently sold type of appliance of the product portfolio. The most relevant ott results are summarized in **Table 32**.

Table 32: Performance characteristics of the used stove model assessed during standard type testing according to EN 13240

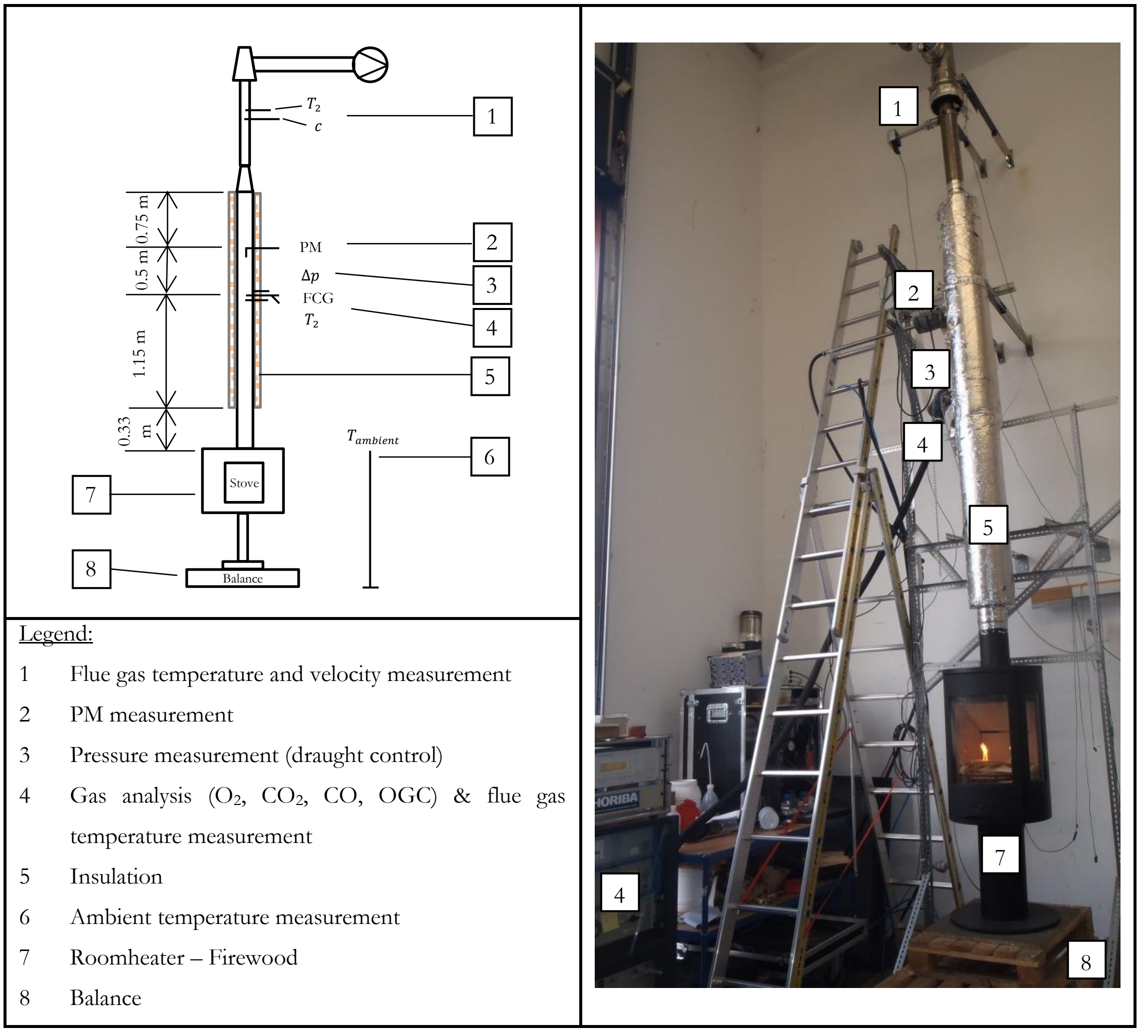
Parameter	Unit	Official-type-test (ott) results
Nominal thermal heat output	kW	5.5
Thermal efficiency	%	80
Carbon monoxide (CO)	vol.-%	0.09
	*mg/m³	1125
Nitrogen oxides (NO_x)	*mg/m³	94
Organic gaseous compounds (OGC)	*mg/m³	73
Particulate matter (PM)	*mg/m³	12
Temperature at flue outlet	°C	299

* based on standard temperature (273.15 K) and pressure (101,325 Pa) (STP), dry, referred to 13 vol.-% O_2

7.2.3 Experimental test setup and measurements

For the combustion experiments the firewood stove was placed on a balance (Mettler Toledo PTA 455-600, accuracy ±50 g) at the laboratory test stand (**Figure 69**). A portion (0.33 m) of the flue gas pipe was not insulated. The following part of the measuring section (0.15 m diameter) was insulated with a layer of 0.05 m glass wool. The combustion tests were carried out under controlled draft conditions of 12 ± 2 Pa. The flue gas draft (Δp) was monitored 50 mm downstream the gas analysis using a differential pressure manometer (Thermokon DPT 2500-R8). Gaseous composition of the flue gas (FGC) was measured continuously over the entire test duration. CO_2, O_2, and CO concentrations were measured with a multigas analyzer (NGA 2000-MLT4). OGC was measured with an FID (M&A Thermo-FID PT63LT) as total hydrocarbons (THC) based on methane (CH_4) equivalents. A thermocouple (type K) centrally placed in the flue gas pipe was used for measuring the flue gas temperature (T_1). The volume flow conditions were assessed by measuring the flue gas velocity (c) with a vane wheel anemometer (Höntzsch ZS25: accuracy ±0.1 m/s) and the flue gas temperature (T_2)

(thermocouple, type K) after reducing the inner diameter of the flue gas pipe to 99 mm. Ambient air temperature ($T_{ambient}$) was measured with a thermocouple (type K) at the distance of around 2 m next to the firewood stove on the level of the flue outlet (1.5 m above the floor).



Legend:
1 Flue gas temperature and velocity measurement
2 PM measurement
3 Pressure measurement (draught control)
4 Gas analysis (O_2, CO_2, CO, OGC) & flue gas temperature measurement
5 Insulation
6 Ambient temperature measurement
7 Roomheater – Firewood
8 Balance

Figure 69: Scheme and picture of experimental test setup (all dimensions in m)

PM emissions were measured gravimetrically by out-stack measurement according to VDI 2066-1 guidelines [135]. Isokinetic sampling was adjusted based on the flue gas velocity measurements. The retention of the particles was carried out with a stuffed quartz wool cartridge and a downstream plane filter, both of which were heated up to 160 °C during the measurements. Rinsing of the sampling probe was carried out with acetone after each combustion experiment. The total mass of rinsing was distributed mass-weighted to all PM measurements of the experiment. For pre- and post-conditioning the stuffed filter cartridges, the plane filters, and the rinsing pots were dried at 180 °C for 1 h and subsequently cooled down in a desiccator for at least 8 h until the thermal equilibrium was reached. The collected mass of particles was determined by weighing the filters before and after the measurements on a precision balance (Type: Sartorius ME 235P, accuracy ±0.01 mg).

7.2.4 Combustion experiments

Real-Life Test Protocol: *"beReal"*

Three combustion experiments according to the *"beReal"* test protocol were carried out [136]. Each combustion experiment consisted of a heating cycle with eight consecutive batches and a subsequent cooling down phase (= 9 test phases). The heating cycle started with the ignition batch followed by four batches at nominal load (batch 2 – 5) and three batches at partial load (batch 6 – 8). The cooling-down phase is defined as the duration when the refilling criteria of the eighth batch is reached until the flue gas temperature T_1 decreases to 50 °C (**Figure 70**).

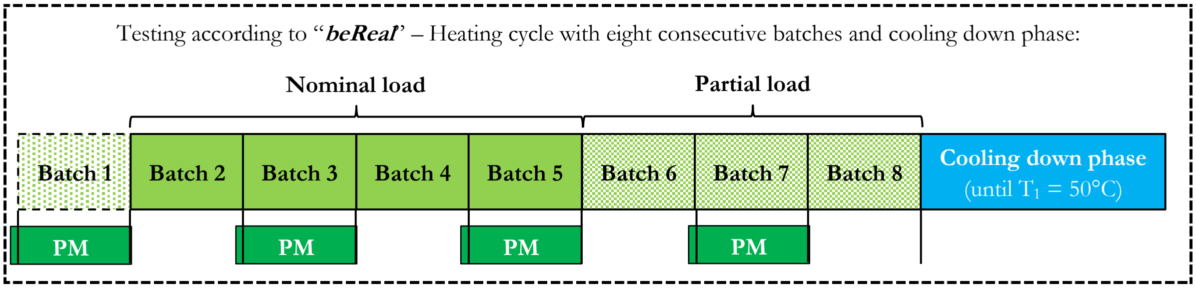


Figure 70: Test and PM sampling phases according to the "*beReal*" test protocol

Nominal load represents 100% of the fuel mass (1.5 kg) per batch whereas partial load is defined as 50% of the nominal load batch mass (0.75 kg). For nominal and partial load the total batch mass was equally divided on two firewood pieces without bark. The length of the firewood pieces was 0.33 m. The firewood pieces were placed crosswise in the combustion chamber as defined in the manual and the QUG. The lighting of the ignition batch was carried out by the top-down ignition technique as defined by the manufacturer in the QUG (**Figure 71**).

Figure 71: Lighting procedure of the ignition batch according to the top-down ignition technique

Therefore, two pieces of firewood, each 0.5 kg, were placed on the grate. On top of these two firewood pieces eight pieces of kindling material (0.5 kg) were stocked crosswise in three layers (1. Layer: 3 Pieces; 2. Layer: 2 pieces; 3. Layer: 3 pieces). The starting aids, (FLAMAX: ECOLOGICAL WOOD WOOL base of wood and wax, rolled and soaked to small balls [118]) were placed between the two kindling pieces used for the second layer.

Recharging criteria of *"beReal"* is defined as the measured CO_2 concentration of the flue gas reaching 25% of the maximum CO_2 concentration of the respective batch. In the case that the maximum CO_2 concentration of the respective fuel batch was lower than 12 vol.-%, the recharging was done at a measured CO_2 value of 3 vol.-% (absolutely). The air lever for adjustment of combustion air supply was set to fully open during the ignition batch and the nominal load batches (batches 2 – 5). When a change was made to a partial load operation, the setting for combustion air supply was changed to decrease the combustion air supply. Therefore, the lever was adjusted to 40% in the direction of "totally closed" settings. After the heating operation was finished, the lever remained at the settings of partial load operation. This behavior results in increased thermal heat losses after finishing heating operation, but this is a common practice of users of firewood stoves (see chapter 2.4.1: **Table 8**, Question 4.8). After cooling down, the remaining residues in the combustion chamber were collected. Subsequently, the share of unburnt carbon was determined according to standard EN 14775 [95].

<u>Standard-Type Test Protocol: EN 13240.</u>

Three combustion experiments according to the EN 13240 [13] test procedure were conducted. Each combustion experiment consisted of a heating cycle with seven consecutive batches (**Figure 72**).

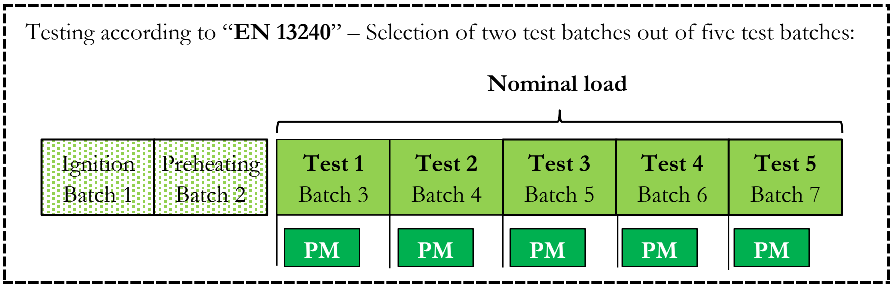


Figure 72: Test and PM sampling phases according to the EN 13240 test protocol

For each EN test five test batches were conducted. With a deviation from EN 13240 test protocol, the flue gas temperature was measured with the thermocouple (T_1) instead of a suction pyrometer. This was defined due to the impossibility of enabling the required flue gas velocity of 20 – 25 m/s in the suction pyrometer with the used gas sampling system. Previous tests showed that a sampling velocity below this range results in too low measured flue gas temperatures which affect efficiency determination significantly [131].

PM sampling was carried out in each test batch (**Figure 72**). According to CEN/TS 15883 [98] PM sampling started 3 min after recharging a new fuel batch and closing the door and lasted for 30 min. A fuel batch was terminated according to the signal of the balance when the mass of the test fuel batch was combusted (±0.05 kg according to EN 13240). The ignition of the first fuel batch was performed as described for the *"beReal"* test procedure. The second fuel batch, consisting of two firewood pieces with a total mass of 1.5 kg, was recharged when the flames of the ignition batch extinguished. Basic firebed was specified according to the balance signal when the flames of the preheating batch extinguished. The air valve setting during ignition and preheating was set at "fully open". For each test batch (batches 3 – 7) a mass of beech firewood of 1.5 kg equally proportioned over two firewood pieces without bark was used. The firewood pieces had a length of 0.33 m and were placed crosswise in the combustion chamber as defined in the manual. The air valve settings for the test batches were reduced as specified in the manual. The lever of combustion air supply was adjusted 20% in the direction of totally closed settings. At this setting the results of thermal heat output were close to the values as specified in the declaration of performance based on ott results.

7.2.5 Data evaluation and statistical analysis

Emissions

Emission concentrations were calculated for both test protocols for CO, OGC and PM emissions. They were calculated based on dry flue gas at standard temperature and pressure conditions (STP: 273.15 K = 0°C / 101,325 Pa) and subsequently transferred to 13 vol.-% O_2. For the standard EN 13240 the average CO, OGC and PM emission concentrations of each test batch were calculated (**Eq. 8** – **Eq. 11**). Out of the five test batches the average of the best two batches regarding CO emissions was used as EN 13240 test result.

$$CO_{mg/m^3,\,STP,\,dry} = CO_{ppm} \times 1.25 \qquad \textbf{(Eq. 8)}$$

$CO_{mg/m^3,\,STP,\,dry}$	Average CO concentration of the respective test phase or test batch in the dry flue gas at STP conditions; in mg/m³
CO_{ppm}	Average flue gas CO concentration of the respective test phase or test batch in the dry flue gas; in ppm

OGC emission test results of test batches were calculated based on methane (CH_4) equivalents according to technical specification CEN/TS 15883 (**Eq. 9**). Thereby, the measured average emission concentration of gaseous total hydrocarbons (THC) in ppm is transferred to mgC/m³ (STP) using the molar mass of carbon (12 g/mol) and the molar volume (ideal gas: 22.4 m³/mol at STP conditions). Since the THC concentrations are measured in wet flue gas they are transferred to dry flue gas conditions by **Eq. 9a** and **Eq. 9b** which base on fuel analysis and combustion calculations.

$$OGC_{mg/m^3,\,STP,\,dry} = \frac{THC_{ppm} \times 12}{22.4} \times \frac{G_W}{G_D} \tag{Eq. 9}$$

$$G_w = \frac{C - C_r}{0.536\left(CO_{2,\,mean} + CO_{mean}\right)} + 1.24\,\frac{9H + W}{100} \tag{9a}$$

$$G_D = \frac{C - C_r}{0.536\left(CO_{2,\,mean} + CO_{mean}\right)} \tag{9b}$$

$$C_r = 1.4925 \times 10^{-5} \times H_{u,\,f} \tag{9c}$$

$OGC_{mg/m^3,\,STP,\,dry}$	Average OGC concentration in the dry flue gas at STP conditions; in mg/m³
THC_{ppm}	Average total hydrocarbon (THC) concentration in the wet flue gas of the respective test batch; in ppm
G_w	Actual specific wet flue gas volume expressed; in m³/kg fuel (STP)
G_D	Actual specific dry flue gas volume expressed; in m³/kg fuel (STP)
$CO_{2,\,mean}$	Average CO_2 concentration of the respective test batch in the dry flue gas; in % of volume
CO_{mean}	Average CO concentration of the respective test batch in the dry flue gas; in % of volume
C	Carbon content of test fuel (as fired basis); in % of mass
C_r	Carbon content of residues referred to the quantity of the test fuel (as fired basis); in % of mass
H	Hydrogen content of test fuel (as fired basis); in % of mass
W	Moisture content of test fuel; in % of mass
$H_{u,\,f}$	Net calorific value of the test fuel (as fired basis); in kJ/kg

PM emissions of gravimetric measurements were calculated according to the technical specification CEN/TS 15883 based on the mass of loaded and unloaded filters as well as the sampled volume (**Eq. 10**) which is transferred to STP conditions according to **Eq. 10a**.

$$PM_{mg/m^3,\,STP,\,dry} = \frac{m_{filter,\,end} - m_{filter,\,start}}{V_{gasmeter,\,STP}} \tag{Eq. 10}$$

$$V_{gasmeter,\,STP} = \left(V_{gasmeter,\,end} - V_{gasmeter,\,start}\right) \times \frac{273.15}{(273.15 + T_{gasmeter})} \times \frac{p_{ambient}}{101{,}325} \times C_f \tag{10a}$$

$PM_{mg/m^3,\,STP,\,dry}$	Average PM concentration in the dry flue gas at STP conditions; in mg/m³
$V_{gasmeter,\,STP}$	Sampled flue gas volume of PM measurement in dry flue gas at STP conditions; in m³
$m_{filter,\,end}$	Mass of filter (plane filter and cartridge) at the end of sampling; in mg

$m_{filter,\, start}$	Mass of filter (plane filter and cartridge) at the start of sampling; in mg
$V_{gasmeter,\, end}$	Gas meter reading at the end of sampling; in m³
$V_{gasmeter,\, start}$	Gas meter reading at the start of sampling; in m³
$T_{gasmeter}$	Average temperature of the gas meter during sampling; in °C
$p_{ambient}$	Average ambient air pressure; in Pa
C_f	Dry gas meter calibration factor, standard value of 1 was used

Finally, emission concentrations of dry flue gas are transferred to 13 vol.-% O_2 according to the standard EN 13240 and the technical specification CEN/TS 15883 (**Eq. 11**).

$$E_{mg/m^3,\, STP, dry,\, 13\ vol.-\%\ O_2} = E_{mg/m^3,\, STP,\, dry} \times \frac{(21-13)}{(21-O_{2,\, mean})} \qquad (\textbf{Eq. 11})$$

$E_{\frac{mg}{m^3},\, STP, dry,\ 13\ vol.-\%\ O_2}$	Average emission concentration in the dry flue gas at STP conditions referred to 13 vol.-% O_2; in mg/m³
$E_{mg/m^3,\, STP,\, dry}$	Average emission concentration in the dry flue gas at STP conditions; in mg/m³
$O_{2,\, mean}$	Average O_2 concentration of the respective test batch in dry flue gases; in % of volume

For *"beReal"* the evaluation of emissions was widely following the standard EN 13240. In contrast to the evaluation of single batch results (EN 13240) the whole heating cycle consisting of nine test phases was considered. During EN 13240 testing the batch load and the air settings were not changed. Consequently, the batch durations and the volume flow conditions (STP) were similar for each test batch (**Figure 73**, right).

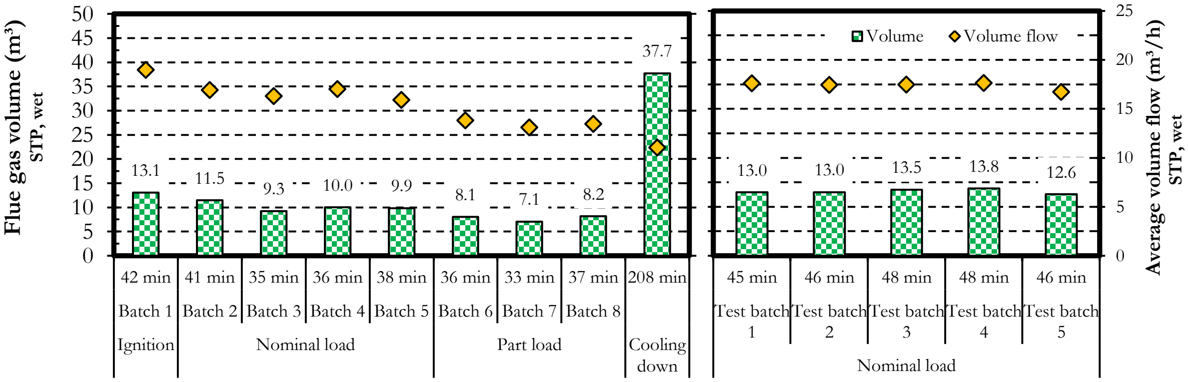


Figure 73: Average volume flow (STP) and total flue gas volume (STP) of batches or test phases for "*beReal*" (left) and EN tests (right) (example of experiment 3 of *"beReal"* and EN tests)

As it is illustrated in the right diagram of **Figure 73**, batch durations and the respective flue gas volume of single test batches according EN 13240 tests differed only marginal between 45 min and 48 min and between 12.6 m³ and 13.8 m³ (STP, wet). The volume flow was almost constant at 17.5 m³/h (STP, wet).

According to the standard EN 13240, a constant volume flow (STP conditions) and respectively mass flow is assumed and therefore average emission concentrations of single test batches are not weighed by volume or mass during the data evaluation. Furthermore, it is noted in the EN standard that "*calculation errors are deemed to be small*" [13]. The measurements of EN test batches of **Figure 73** (right diagram) confirmed this assumption. Consequently, a simple time-weighted data evaluation was applied for the EN 13240 test batches.

In contrast, for *"beReal"* (**Figure 73**, left diagram), the settings of combustion air supply and the durations of evaluated test phases changed. Consequently, the volume flow and the total amount of flue gas volume of different test phases were different. During the first fuel batch, where air settings were fully open, the highest volume flow of 19.2 m³/h (STP, wet) was observed. During the cooling down phase where air settings were closed the volume flow which resulted from leakiness of the stove was lowest at around 11.2 m³/h (STP, wet). The volume of single test phases of the "*beReal*" protocol ranged from 7.1 m³ to 13.1 m³ during heating operation and up to 37.7 m³ for the cooling down phase (all volumes at STP, wet). Consequently, a volume-weighted data evaluation was applied for *"beReal"* that considered the different operating conditions of the nine test phases (**Eq. 12**)(**Eq. 12**). Thereby, the total amount of wet flue gas volume for each test phase based on the average flue gas velocity (c_{mean}) was calculated and transferred to STP conditions (**Eq. 12a** and **Eq. 12b**).

$$E_{volume-weighted} = \frac{\sum_{i=Ignition\ (test\ phase\ 1)}^{cooling\ down\ (test\ phase\ 9)} \times E_{i,\ dry} \times V_{i,\ STP,\ wet}}{\sum_{i=Ignition}^{cooling\ down} \times V_{i,STP,wet}} \qquad \textbf{(Eq. 12)}$$

$$V_{i,STP,\ wet} = \dot{V}_{i,\ operation} \times \tau_i \times \frac{273.15}{(273.15\ +\ T_2)} \times \frac{p_{flue\ gas}}{101325} \qquad \textbf{(12a)}$$

$$\dot{V}_{i,operation} = \frac{d^2\pi}{4} \times c_{mean} \qquad \textbf{(12b)}$$

$E_{volume-weighted}$	Volume-weighted flue gas concentration of one parameter (i.e. O_2, CO_2, CO, OGC, PM) in the dry flue gas; in ppm, % of volume or in mg/m³,
$E_{i,\ dry}$	Average flue gas concentration of one parameter for the respective test phase (Ignition, batch 2, …, cooling down) in the dry flue gas; in ppm, % of volume or in mg/m³

$V_{i,\,STP,\,wet}$	Total wet flue gas volume (STP) of the respective test phase (Ignition, batch 2, …, cooling down); in m³
$\dot{V}_{i,operation}$	Wet volume flow of flue gas of the respective batch (1…8) or cooling down phase; in m³/s
τ_i	Duration of test phase; in seconds
T_2	Average flue gas temperature of test phase at measuring point T_2 (velocity measurement); in °C
$p_{flue\ gas}$	Absolute pressure of the flue gas; in Pa
d	Diameter of the velocity measurement section; in m
c_{mean}	Average flue gas velocity; in m/s

The average flue gas velocity (c_{mean}) of a test phase was calculated based on calibration pretests for assessing the flow factor. The calibration procedure is illustrated in the right diagram of **Figure 74**. Thereby, defined volume flows were generated by the Wöhler DP 600 in the expected range of measurements of the combustion experiments. The Wöhler DP 600 is a special instrument to evaluate the leakage of chimneys, exhaust systems and stoves. The calculated flow factor for the different calibration points was in the range of about 0.5 to 0.75 (**Figure 74**, right). A linear function was derived from all calibration points. With the linear function all velocity measurements (c) of the combustion experiments were transferred to the average flue gas velocity (c_{mean}). The average flue gas velocity (c_{mean}) of a "*beReal*" test cycle is illustrated in the left diagram of **Figure 74** and ranged between 0.5 m/s to 1.1 m/s.

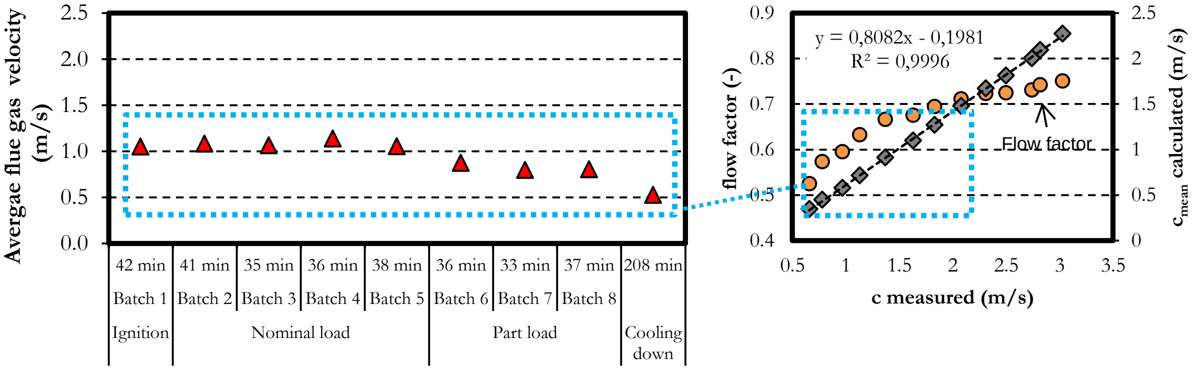


Figure 74: Average flue gas velocities for *"beReal"* testing (left) and the calibration function (right) used to calculate the average flue gas velocity (c_{mean}) for further evaluation (experiment 3 of *"beReal"* tests)

Eq. 8 – **Eq. 10** were used for determination of average emission concentrations for CO, OGC and PM. For PM emissions the total mass of collected particles (Batch 1, 3, 5 and 7) in relation to the total sampled volume of respective measurements was used. The average concentrations of gaseous components (O_2, CO_2, CO, OGC) for each batch (test phase 1 – 8) and the cooling down phase (test phase 9) were calculated separately. The volume-weighted emission concentrations were transferred to 13 vol.-% O_2 according to **Eq. 11** using the volume-weighted average O_2 concentration. For gaseous emissions (CO, OGC) the volume-weighted O_2 concentration of the whole test cycle was used. Due to the fact that no PM measurement during the cooling down phase was carried out for PM emissions the volume-weighted O_2 concentration without the cooling down phase (test phase 1 – 8) was used. Previous studies indicated a little effect of the char burn-out on total PM emissions [46] [56] [75].

Thermal efficiency

For both test protocols the indirect approach of thermal efficiency determination was used. Accordingly, the thermal (q_a) and chemical (q_b) flue gas losses as well as the losses due to unburnt constituents in the residues (q_r) were considered (see **Eq. 2**).

For EN 13240 experiments the evaluation of thermal ($q_{a,\,EN}$) and chemical ($q_{b,\,EN}$) flue gas losses was based on combustion calculations using the chemical composition of the fuel and balancing the C-content in the flue gas (CO, CO_2). For the proportion of heat losses through combustible constituents in the residues ($q_{r,\,EN}$) a constant value of 0.5% points of thermal efficiency was used as proposed by the EN test protocol. (see chapter 3: **Eq. 3**; **Eq. 4**)

The thermal efficiency was assessed for each test batch. The selected test batches with the lowest CO emissions were also used for evaluation of thermal efficiency performance.

For *"beReal"* experiments the thermal ($q_{a,\,beReal}$) and chemical ($q_{b,\,beReal}$) flue gas losses were calculated based on the measurements of flue gas temperature, ambient temperature, flue gas composition (CO emissions) and average flue gas velocity respecting all measurements of the whole heating cycle. Additionally, the pressure conditions, mass of totally combusted firewood of the test cycle and chemical fuel analysis are used (**Eq. 13**; **Eq. 14**).

$$q_{a,\,beReal} = \frac{Q_{th.,\,flue\ gas}}{Q_{fuel}} \times 100\% \tag{Eq. 13}$$

$$Q_{th.,\,flue\ gas} = \int_{beReal\ start}^{beReal\ end} \dot{Q}_{th.,\,flue\ gas} \times d\tau \tag{13a}$$

$$Q_{fuel} = m_{fuel} \times H_{u,\,f} \tag{13b}$$

$$\dot{Q}_{th.,\,flue\ gas} = \dot{m}_{flue\ gas} \times c_p \times dT \tag{13c}$$

$$\dot{m}_{flue\ gas} = \frac{d^2\pi}{4} \times c_{mean} \times \rho_{flue\ gas} \tag{13d}$$

$$\rho_{flue\ gas} = \rho_{STP} \times \frac{273.15}{(273.15 + T_2)} \times \frac{p_{ambient} - 12}{101325} \tag{13e}$$

$$dT = T_{flue\ gas} - T_{ambient} \tag{13f}$$

$q_{a,\,beReal}$	Proportion of thermal heat losses in the flue gas ($Q_{th.,\,flue\ gas}$) referred to the energy of the fired test fuel (Q_{fuel}); in %
$Q_{th.,\,flue\ gas}$	Thermal energy flow of the flue gas; in kJ/s
$d\tau$	Time duration of measuring interval; in s
Q_{fuel}	Energy of the fired test fuel; in kJ
m_{fuel}	Total mass of fired test fuel; in kg
$\dot{m}_{flue\ gas}$	Mass flow of the flue gas; in kg/s
c_p	Specific heat capacity of dry air at the measured flue gas temperature (linear interpolation between the values given in **Table 33**; in kJ/(kg K)
d	Flue gas pipe diameter of the velocity measurement section; in m
$\rho_{flue\ gas}$	Density of the flue gas, assumed as dry air; in kg/m³
ρ_{STP}	Density of the flue gas (assumed as dry air) at STP conditions: 1.292 kg/m³
$p_{ambient}$	Mean ambient air pressure; in Pa
$T_{flue\ gas}$	Flue gas temperature at measuring point T_1; in °C
$T_{ambient}$	Ambient air temperature; in °C

Table 33: Specific heat capacity (c_p) of dry air which was used for calculation of thermal flue gas losses. Linear interpolation between the referred temperatures depending on the flue gas temperature T_1 was conducted.

Temperature (°C)	**Specific heat capacity (c_p) of dry air** (kJ/kg K)	**Temperature** (°C)	**Specific heat capacity (c_p) of dry air** (kJ/kg K)
0	1.005	300	1.045
100	1.011	400	1.070
200	1.025	500	1.093

$$q_{b,\,beReal} = \frac{Q_{ch.,\,flue\ gas}}{Q_{fuel}} \times 100 \tag{Eq. 14}$$

$$Q_{ch,\,flue\ gas} = H_{u,\,CO} \int_{beReal\ start}^{beReal\ end} \dot{m}_{CO} \times d\tau \tag{14a}$$

$$\dot{m}_{CO} = CO_{ppm} \times 1.25 \times \frac{d^2\pi}{4} \times c_{mean} \times \frac{273.15}{(273.15 + T_2)} \times \frac{p_{ambient} - 12}{101325} \tag{14b}$$

CO_{ppm}	Measured flue gas CO concentration; in ppm
$Q_{ch,\, flue\ gas}$	Chemical energy of the flue gas; in kJ/s
$H_{u,\, CO}$	Net calorific value of CO: 10103 kJ/kg
$\dot{m}_{CO}$	Mass flow of CO in the flue gas; in kg/s

The proportion of losses due to combustibles in the residues ($q_{r,\, beReal}$) was calculated according to **Eq. 15**. Additionally, the proportion of losses due to combustibles in the residues was experimentally determined ($q_{r,\, experiemntal}$) (**Eq. 16**).

$$q_{r,\, beReal} = \frac{\left(m_{residues} - m_{fuel} \times \frac{a}{100} \times 1.2\right) \times H_{u,\, C}}{m_{fuel} \times H_{u,\, f}} \times 100 \qquad \textbf{(Eq. 15)}$$

$q_{r,\, beReal}$	Proportion of losses due to combustibles in the residues referred to the net calorific value of the test fuel; in %
$m_{residues}$	Total mass of residues taken out of the stove after the *"beReal"* test cycle (kg)
a	Ash content of fired fuel; in % of mass
$H_{u,\, C}$	Net calorific value of carbon (C): 33480 kJ/kg

The factor of 1.2 of **Eq. 15** shall respect the carbonate content of the ash, which is formed during combustion. The calculated results for $q_{r,\, beReal}$ according to **Eq. 15** were compared with the experimental results of a sample treated according to EN 14775 ($q_{r,\, experiemntal}$). Thereby, the share of combustibles in the residues of the total consumed fuel mass was determined with the equations given in the EN13240 standard for non-woody solid fuels (**Eq. 16**). In contrast to the EN standard the total amount of residues was respected and not only the residues which passed through the grate.

$$q_{r,\, experiemntal} = \frac{Q_{r,\, experimental}}{H_{u,\, f}} \times 100 \qquad \textbf{(Eq. 16)}$$

$$Q_{r,\, experimental} = \frac{335 \times B \times A}{100} \qquad \textbf{(16a)}$$

$q_{r,\, experiemntal}$	Proportion of heat losses through combustible constituents in the residues $Q_{r,\, experimental}$ referred to the calorific value of the test fuel ($H_{u,\, f}$); in %

$Q_{r,\,experimental}$	Heat losses through combustible constituents in the residue referred to the unit of mass of the test fuel (as fired basis); in kJ/kg
A	Residues, referred to the mass of the fired test fuel; in % of mass
B	Combustible constituents in the residues, referred to the mass of residues; in % of mass

Based on the chemical composition of the fired fuel (**Table 31**) and the unburnt residues (**Table 34**), combustion calculations [119] and the combustibles content of the residues ($x_{combustibles}$) the total amount of wet flue gas volume ($V_{w,\,STP,\,calculated}$) was calculated and compared with wet flue gas volume ($V_{w,\,STP,\,measured}$) measured on the basis of the average flue gas velocity (c_{mean}) (**Eq. 17**; **Eq. 18**). The amount of flue gas volume which refers to the unburnt combustibles was also determined (**Eq. 17c**; **Eq. 17d**) and subtracted from the calculated wet flue gas volume (**Eq. 17**).

Table 34: Elemental composition (w.b. = wet based) of carbon (C), sulfur (S); hydrogen (H) and moisture content (W) of unburnt residues according to [137].

C (kg/kg$_{residues}$ w.b.)	**S** (kg/kg$_{residues}$ w.b.)	**H** (kg/kg$_{residues}$ w.b.)	**W** (kg/kg$_{residues}$ w.b.)
0.883	0.0002	0.0319	0.06

$$V_{w,\,STP,\,calculated} = V_{w,\,STP,\,fuel} \times m_{fuel} - V_{w,\,STP,\,combustibles} \times m_{combustibles} \qquad \textbf{(Eq. 17)}$$

$$V_{w,\,STP,\,fuel\ or\ combustibles} = 1.87\,C + 0.7\,S + 11.2\,H + 1.24\,W + L_{min} \times \left(\frac{21}{21 - O_{2,\,mean,\,volume\text{-}weighted}} - 0.21\right) \qquad \textbf{(17a)}$$

$$L_{min} = \frac{1.87\,C + 5.6\,H + 0.7\,S - 0.7\,O}{0.21} \qquad \textbf{(17b)}$$

$$m_{combustibles} = m_{residues} \times x_{combustibles} \qquad \textbf{(17c)}$$

$$x_{combustibles} = \frac{m_{residues} - m_{fuel} \times \frac{a}{100} \times 1.2}{m_{residues}} \qquad \textbf{(17d)}$$

$V_{w,\,STP,\,calculated}$	Calculated total wet flue gas volume at STP conditions; in m^3
$V_{w,\,STP,\,fuel}$	Calculated specific wet flue gas volume referred to the fired test fuel at STP conditions; in m^3/kg_{fuel}
$V_{w,\,STP,\,combustibles}$	Calculated specific wet flue gas volume referred to the combustibles in residues at STP conditions; in m^3/kg_{fuel}

C, H, S, W, O	Elemental composition of used fuel (**Table 31**) and unburnt residues (**Table 34**); in kg/kgfuel or kg/kg$_{residues}$
$m_{combustibles}$	Mass of combustibles in the residues; in kg
L_{min}	Stoichiometric minimum combustion air demand at STP conditions; in m³/kgfuel and m³/kg$_{combustibles}$
$O_{2,\,mean,\,volume-weighted}$	Volume-weighted, average O_2 concentration of the considered test duration (test phase 1-9); in % of volume
$m_{residues}$	Total mass of unburnt residues sampled out of the stove after the *"beReal"* test cycle; in kg
$x_{combustibles}$	Combustibles content of residues; in kg$_{combustibles}$/kg$_{residues}$

The total amount of measured flue gas volume ($V_{w,\,STP,\,measured}$) was assessed according to **Eq. 18** with the dimensions of the flue gas pipe and measured average flue gas velocity. Subsequently, data was transferred to STP conditions based on temperature and pressure measurements.

$$V_{w,\,STP,\,measured} = \int_{beReal\ start}^{beReal\ end} \frac{d^2\pi}{4} \times c_{mean} \times \frac{273.15}{(273.15 + T_2)} \times \frac{p_{ambient} - 12}{101325} \times d\tau \qquad \text{(Eq. 18)}$$

Based on the comparison of calculated and measured volumes the quality of measurements was validated. This validation calculation is obligatory required by the *"beReal"* test protocol as an element of quality assurance [136].

Statistical analysis

For evaluation of the repeatability the average ($\emptyset$) and standard deviation (σ) was calculated for the three experiments (n = 3) of each test protocol. The repeatability defines the variation of test results of EN 13240 and *"beReal"* measurements which were carried out with the same measurement devices by the same person under equal testing conditions in accordance to the respective test protocol respectively. The coefficient of variation (CV) was calculated according to **Eq. 19** in order to compare the repeatability of different parameters. The CV represents the relative standard deviation and enables the comparison of parameters with different orders of magnitudes.

$$CV = \frac{\sigma}{\emptyset} \times 100 \qquad \text{(Eq. 19)}$$

CV	Coefficient of variation, in %
σ	Standard deviation of the three combustion experiments for each test protocol
$\emptyset$	Average of the three combustion experiments for each test protocol

For statistical evaluation of the test results of *"beReal"* and EN 13240 a Student´s t-test was applied using "MS-Excel" software. Significant differences were defined by p-values ≤ 0.05 and highly significant differences by p-values ≤ 0.01

7.3 Results and discussion

7.3.1 Fundamental differences of test protocols

As illustrated in **Figure 75** real-life heating operation of firewood stoves consists of several batches starting with the ignition of the first fuel batch and ending with the cooling down phase. The appliance performance regarding emissions and thermal efficiency in real-life heating operation depends on three main aspects – technological aspects (e.g. features of the appliance like an automatic air control or an advanced air staging concept), the mode of heating operation by the user (e.g. correct heating operation or maloperation) and aspects referring to the installation conditions (e.g. the chimney design or potential retrofit applications, like an external draught control system).

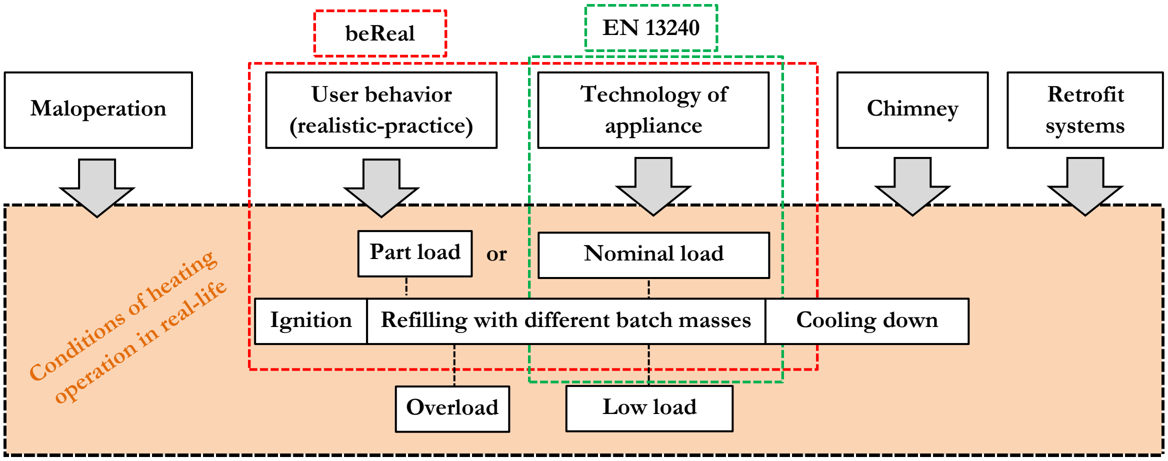


Figure 75: Aspects of real-life heating operation that are considered by the test protocols EN13240 and *"beReal"*

A testing protocol evaluating the technology of the appliance embedded in the whole heating system would reflect the best real-life relevance. This would require the evaluation of the appliance performance with respect to the installation conditions and all potential aspects of user behavior. However, since the situations in the field regarding those aspects are manifold such an evaluation, especially in the laboratory, seems impossible with regard to repeatability, reproducibility and testing costs. Furthermore, it might be difficult to guarantee the equivalent of opportunities for manufacturers at different testing bodies.

The *"beReal"* test protocol is based on the specifications given in the Quick-User-Guide (QUG) and reflects a more realistic heating operation than the actual standards. The test procedure covers typical elements of heating operation in real-life, that is, ignition, heating operation in different load settings, and the cooling-down process (**Figure 75**). The QUG defines relevant aspects of user behavior, like the ignition technique, number, and mass of used firewood pieces and kindling material for ignition, nominal load, and partial load as well as the lever settings of combustion air supply for the different test phases. Only firewood pieces of beech are permitted. The use of firewood with bark is not obligatory for testing since comparative combustion experiments showed no significant influence of bark on *"beReal"* test results regarding CO, OGC, and PM emissions [132]. As kindling material, beech or spruce may be used. The use of biobased fire starters is mandatory. At least two firewood pieces have to be used for each batch. The distribution of total batch mass over the used firewood pieces shall be similar (±10%). The mass of the ignition batch has to be at least 80% of the batch mass used for nominal load heating operation. Heating operation at partial load is obligatorily defined by using 50% of the mass of nominal load. Refilling of batch loads is carried out according to a CO_2 flue gas measurement which represents the instant of time when flames are extinguished. This was observed as a typical instant of time for refilling according to user surveys (see chapter 2.4.1: **Table 8**, Question 4.6) [74].

The EN test protocol considers predominantly appliances-specific design aspects, like the combustion chamber and air staging design, used materials, and the leakage rate of the appliance (**Figure 75**). As test fuel firewood of beech, birch, or hornbeam is obligatory. There are fuel requirements regarding chemical properties, for example, moisture content (16 ± 4 ma.-%) and ash content (<1 ma.-%), but the length of firewood pieces and the distribution of batch mass over the number of firewood pieces are not defined. EN testing is conducted only at nominal load which represents a performance evaluation of emissions and efficiency under optimal and quasi-stable heating operation. In contrast to the *"beReal"* test protocol, transient conditions, for example, ignition and preheating (batches 1 and 2) of the stove, load changes, or cooling-down phases are not considered in the EN test procedure (**Figure 75**).

But also the *"beReal"* test protocol covers not all situations or operational conditions which can occur during real-life operation. User behavior that deviates from the QUG is not considered. Therefore, increased emissions which can occur due to maloperation, for example, due to the use of inappropriate fuels like wet firewood [39] [40] a too high (overload) or too low (low load) batch size [21] [38] [42] inappropriate ignition technique (see chapter 3) [46] and air settings [41] or the use of litter [83] are not considered by the *"beReal"* test procedure (**Figure 75**). Furthermore, the chimney system which induces the flue gas draught according to dimensions, construction materials, weather, and temperature conditions is not considered in detail within the *"beReal"* test procedure. The constant flue gas draught of 12 ± 2 Pa used for "*beReal*" testing does not respect the effect of different draught conditions on emissions and thermal efficiency for different types of appliances (chapter 3). In long-term field measurements an average flue gas draught of 18 Pa (range: 3 Pa – 28 Pa) was observed [88]. As presented and discussed in chapter 3, experimental combustion tests at different firewood roomheaters revealed no statistically significant effect of draft conditions on gaseous and particulate emissions in the range of 12 − 48 Pa. However, for all evaluated stoves the thermal efficiency decreased significantly at higher draft conditions. On the basis of these results, the *"beReal"* flue gas draft was defined equally as the EN test protocol at 12 ± 2 Pa. A minimum flue gas draft of 12 Pa is required by manufacturer's manual that normal heating operation is guaranteed. Furthermore, potential technologies that can be used as retrofit applications to control the flue gas draft and combustion air supply or to reduce emissions, for example, filters or catalysts, are not part of *"beReal"* testing. Differences in *"beReal"* and EN test protocol regarding measuring methods and data evaluation are summarized in **Table 35**.

Table 35: Differences of the EN 13240 and *"beReal"* test protocol regarding measuring methods and data evaluation

Parameter	**EN 13240 test protocol & CEN/TS 15883 for PM**	***"beReal"* test protocol**
Measurements		
Gaseous components: (O_2, CO_2, CO, OGC)	Obligatory only CO and O_2 or CO_2; OGC typically tested according to CEN/TS 15883 due to national ELV	All components obligatory
Measuring interval	≤ 1 minute	≤ 10 seconds
Particulate emissions (PM)	Not part of the standard, but typically tested according to CEN/TS 15883 due to national ELV	Part of the test method, basically according to VDI-2066-1 [135], in future new EN-PME test method should be applied [138]·
Sampling	Fixed volume flow	Isokinetic; at least proportional of volume flow
Sampling start	3 minutes after refilling	Before opening the combustion chamber door for refilling
Sampling duration	30 minutes (first 3 minutes of start-up phase not respected)	Entire batch duration respected: start-up, intermediate and burn-out phase
Flue gas temperature	Suction pyrometer*	Thermocouple centrally placed in the flue gas pipe
Velocity of flue gas	Not required (constant volume flow assumed)	Obligatory measuring parameter, calibration for calculation of the average flue gas velocity (c_{mean}) required (see **Figure 74**).
Balance	Required for determination of batch duration and refilling	Not required, refilling according CO_2 flue gas measurement
Data evaluation		
Emission concentrations: CO, OGC, PM	Average of two batches arbitrarily chosen	Volume weighted average of total test duration; PM emissions measured in batch 1, 3, 5 and 7
Thermal efficiency	Indirect determination	Indirect determination
Thermal flue gas losses	Specific calculation based on fuel composition, CO, CO_2 and flue gas as well as ambient air temperature	Absolute calculation based on c_{mean}, c_p of dry air and the flue gas as well as ambient air temperature (see chapter 7.2.5)
Chemical flue gas losses	Specific calculation based on fuel composition, flue gas composition (CO, CO_2) as well as ambient air temperature	Absolute calculation based on CO mass flow
Losses through combustibles in the residues	Fixed value: 0.5% of thermal efficiency	Calculated based on the remaining mass in the combustion chamber and ash box (see **Eq. 15**)

*in this study: thermocouple centrally located in the flue gas pipe (see **Figure 69**)

The main differences of measuring methods between EN 13240 and *"beReal"* testing is the use of the suction pyrometer for flue gas temperature measurement, the balance signal for specification of batch duration, and the flue gas velocity measurement as essential basis for the *"beReal"* data evaluation (**Table 35**). The PM measurement procedure is basically not part of the standard EN 13240, but is typically performed according to CEN/TS 15883: "German and Austrian method" [98] within EN testing. For *"beReal"* testing PM measurement is basically performed according to VDI 2066-1 guideline [135] with an "out-stack" measuring approach. The main difference between both measuring protocols is the isokinetic sampling according to VDI 2066-1 compared to a fixed sampling volume flow according to CEN/TS 15883 (**Table 35**). Furthermore, rinsing of the sampling probe is obligatory according to the guideline VDI2066-1 and the heating temperature of filters is higher (at least 160 °C for VDI 2066-1 guideline compared to at least 70 °C for CEN/TS 15883). PM sampling for both test protocols were carried out according to VDI 2066-1 guideline to increase the comparability of test results.

7.3.2 Comparison of test results of emissions and thermal efficiency

For evaluation of the accuracy of flue gas velocity measurements, the calculated and measured total flue gas volume of each *"beReal"* test were compared (**Table 36**).

Table 36: Comparison of calculated and measured total flue gas volume of *"beReal"* tests as quality assurance for velocity measurement

"***beReal***" **Test**	**Unit**	***"*beReal*":** $V_{w, STP, calculated}$	***"*beReal*":** $V_{w, STP, measured}$	**Deviation** calculated vs. measured	**Average of deviation**
Test 1	m³, STP, wet	103.6	109.5	5.4%	8.7%
Test 2		106.1	118.4	10.4%	
Test 3		110.1	122.9	10.4%	

*… see **Eq. 17** and **Eq. 18**

The mean deviation was below 20% which is required according to the *"beReal"* test procedure [136]. Also t-test results showed no statistical significant difference (p = 0.10).

No significant differences between EN 13240 and *"beReal"* combustion experiments for gaseous and particulate emissions (CO: p = 0.517; OGC: p = 0.998; PM: p = 0.177) were observed. The absolute differences for average CO and OGC results were marginal (< 5%). PM emissions of *"beReal"* test results were around 17% higher compared to EN 13240 results (**Figure 76**). This could be explained by the immediate start of PM sampling at the beginning of the batch and the consideration of all characteristic combustion phases, defined as start-up, intermediate, and burn-out phases [75].

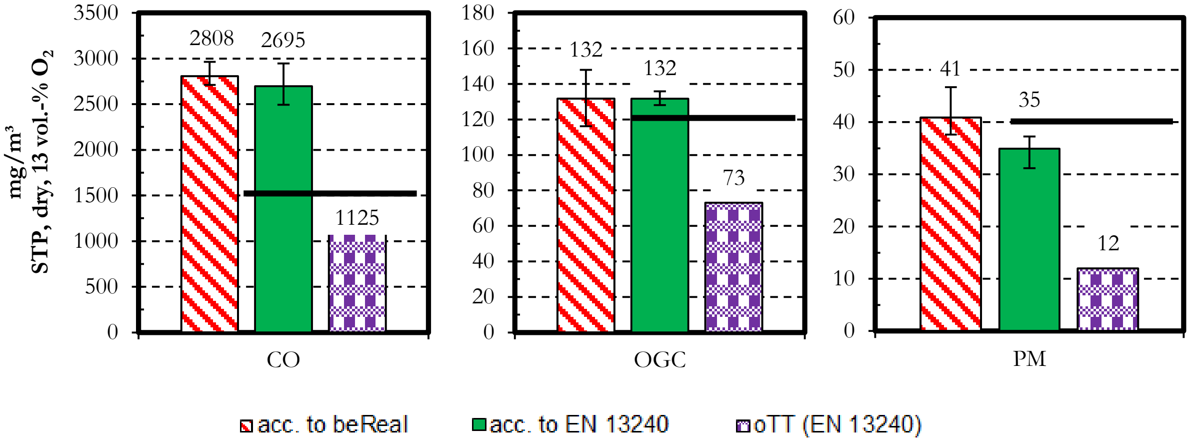


Figure 76: Emission results of combustion experiments according to the EN 13240 and *“beReal”* test protocol in comparison to the official-type-test results (ott) for the respective stove model. The black line represents the Ecodesign threshold value of the respective parameter

In contrast, in a comparison of the ott with *“beReal”* test results, big differences were observed for all measured emission concentrations. Compared to the ott results, the CO emissions of *“beReal”* testing were around 150% higher; OGC around 80%, and PM emissions around 241%. When the ott test results were compared with the EN 13240 test results of this study, also clear deviations for CO, OGC, and PM emissions can be seen. The highest deviation of about 192% was observed for PM emission test results (**Figure 76**).

The future Ecodesign-ELV for CO and OGC emissions were not met by the average results of “*“beReal”*” and EN 13240 test results. For OGC emissions only one and for PM emissions two of the three conducted *“beReal”* combustion tests met the Ecodesign-ELV. However, the average *“beReal”* test results were higher than Ecodesign-ELV. All three EN 13240 test results of PM emissions met the Ecodesign-ELV (**Figure 76**).

The comparison of thermal efficiency results showed similar results compared to the gaseous emissions (**Figure 77**).

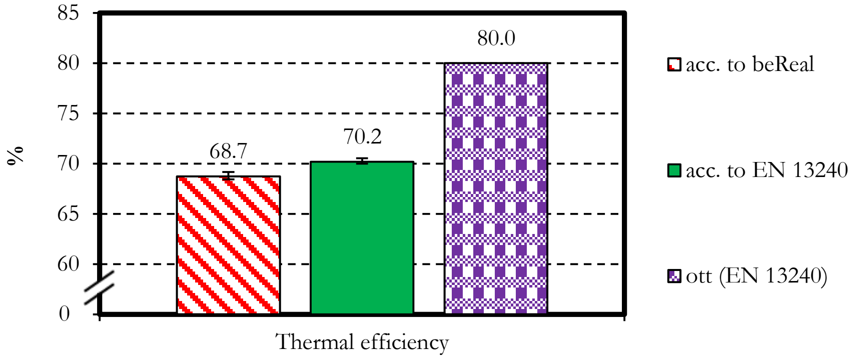


Figure 77: Thermal efficiency results of combustion experiments according to the EN 13240 and *"beReal"* test protocol in comparison to the official-type-test results (ott)

The absolute difference between the *"beReal"* and EN 13240 tests was 1.5% which was statistically highly significant ($p \leq 0.01$). The difference between EN 13240 test results and ott results were 9.2% absolutely. In a comparison of the share of losses which were calculated according to the indirect efficiency determination approach, it is obvious that the highest deviation is mainly caused by the losses due to unburnt material (**Table 37**).

Table 37: Share of thermal and chemical flue gas losses as well as losses due to combustibles in the residues of EN 13240 and *"beReal"* test protocol

Average losses of three tests	**Unit**	**EN 13240**	**"*beReal*"**
Thermal losses (q_a)	$kJ_{losses}/kJ_{fuel\ input}$	0.275	0.238
Chemical losses (q_b)	$kJ_{losses}/kJ_{fuel\ input}$	0.018	0.024
Losses due to unburnt material (q_r)	$kJ_{losses}/kJ_{fuel\ input}$	0.005*	0.051**/ 0.064***

*fixed value according to standard EN 13240 for wood fuel

** $q_{r,\,beReal}$…see **Eq. 15**

*** $q_{r,\,experiemntal}$…see **Eq. 16**

The proposed 0.5% is clearly lower compared to the calculated (5.1%) and measured (6.4%) values of this study. The EN 13240 respects only the residues passing through the grate due to the argumentation that the ash which remains on the grate is used at the next heating operation for ignition. However, the ignition batch is never respected by the EN 13240 testing procedure and an ash and charcoal layer on the grate could also negatively affect the combustion performance of the ignition batch. Consequently, the results indicated that the general use of 0.5% losses due to unburnt material is too low. This is also confirmed by MACK et al. [139] and SCHÜSSLER [140], which investigated q_r losses between 0.4% and 1.8% and between 3.6% and 4.7%, respectively, for two stoves. Furthermore, the results showed that **Eq. 15** could be used as an approximation, but an uncertainty of 25% can be estimated.

Comparing the EN 13240 test results with the ott results, it is obvious that it was not possible to reproduce the ott results. Potential reasons for the high deviations are manifold and are summarized in **Table 38**.

Table 38: Potential reasons for high deviations of EN 13240 test results of this study and ott results

Parameter	Reason	Explanation of potential effects on the test results
Test appliance	No serial-production appliance is used	In this study the tested appliance was a serial product. This might have resulted in a different operating performance
Fuel	Test fuel is supplied by the manufacturer	The single firewood pieces can be optimal designed and placed in the combustion chamber.
Testing procedure	Number of test batches not limited	In this study the number of test batches was limited. For each test 5 test batches were performed.
Flue gas temperature measurement	Suction pyrometer is used	Measurement method is error-prone. A deviation from the required suction velocity (20-25m/s) results in too low flue gas temperature [131]. Consequently, thermal flue gas losses are underestimated and thermal efficiency is overestimated in the ott.
Settings for combustion air supply	Room for interpretation: No constant air valve settings and "delayed" closing of combustion chamber door after refilling.	Combustion chamber door is not closed immediately after recharging or air settings are fully open during the first three minutes after refilling. Consequently, a faster ignition process and lower emissions during the start-up phase is achievable.
Data evaluation	Room for interpretation: Emission evaluation is often made until CO_2 reached 4 vol.-%	Emission data from the beginning of the batch load until CO_2 is 4 vol.-% is respected. High CO emissions during burn-out phase are neglected.
Measuring interval	Larger measuring interval	In this study measuring intervals of 1 second were used for both test protocols. A measuring interval of 1 minute, which is permissible according to EN 13240 standard, might disregard short emission peaks
Measuring error for PM	Sensitivity of measuring parameter	PM measurement is error-prone due to leakage rates in the sampling train which couldn´t be recognized during measurement. A leakage results in lower PM emission measurements.

The main reasons refer most probably to specifics of the tested appliance and the used fuel. Additionally, differences in operation due to room for interpretation of the standard EN 13240 and measuring errors could have been responsible for lower emissions and higher thermal efficiency of ott test results.

Unfortunately, the results of the measurements indicated that tested products during ott differ from the serial-production appliances. This is also underpinned by previous measurements performed during the "*beReal*" project and presented in the report of RÖNNBACK et al. [133] (**Figure 78**). In this study measurement results of 13 selected serial-production stoves classified according to EN 13240 are presented. The construction year of all tested stoves ranged between 2013 and 2015. All selected stoves were tested by different institutions in the laboratory according to the EN 13240 testing standard and the "*beReal*" test protocol. Subsequently, the appliances were installed in the field at the end-users. For all 13 stoves ott tests for CO and PM were available as well as ott results of OGC emissions for 9 stoves. After installation of the stoves in the field three field test days were carried out for each stove.

They were tested when the users operates their appliances according to their common habits, according to specifications of the QUG and according to the "*beReal*" test cycle [133].

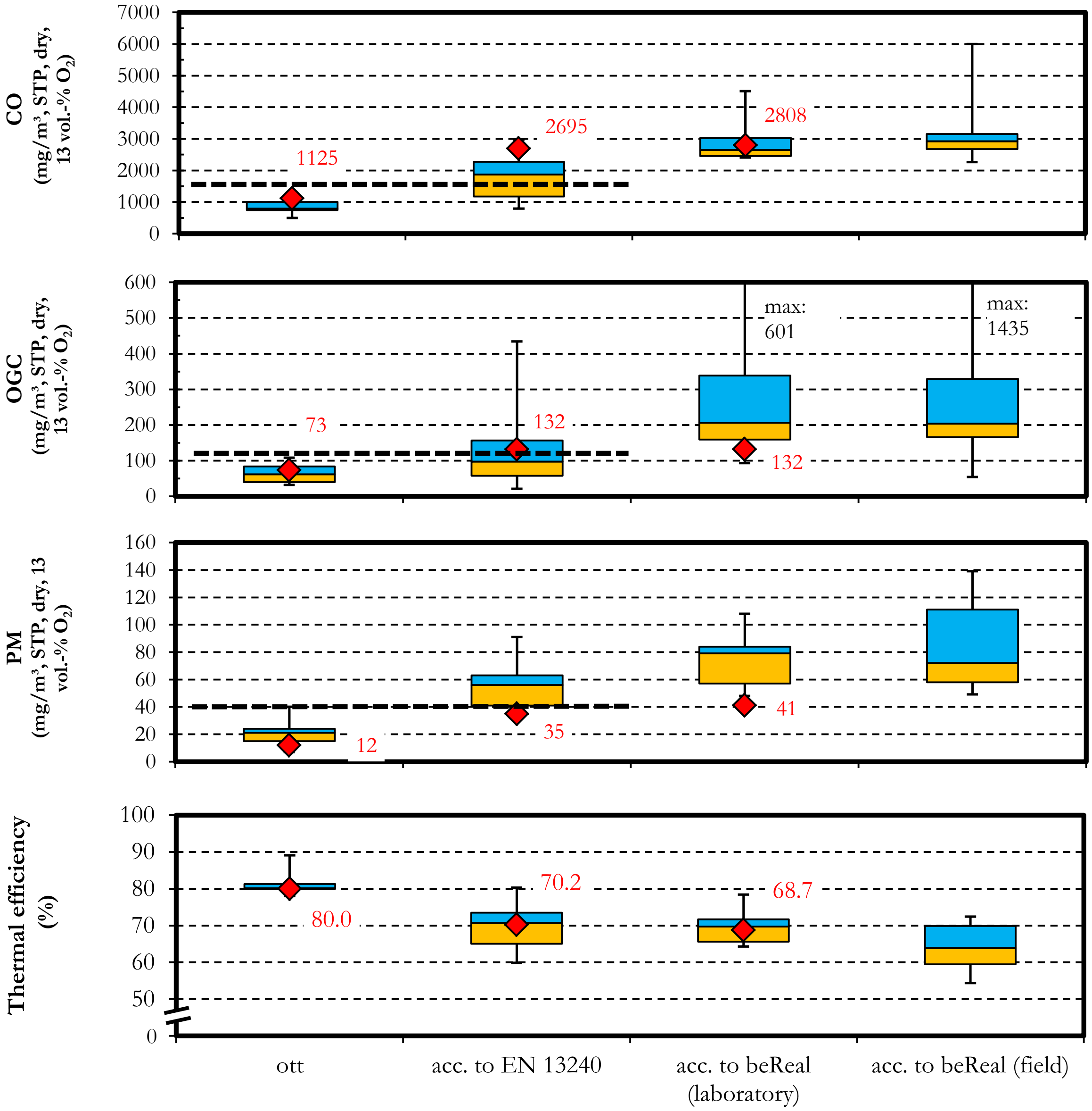


Figure 78: Comparison of test results at 13 serial-production stoves published in the report of RÖNNBACK et al. [133] (box-plots) with own test results (red bullets). The upper and lower whiskers of the box-plots represent the minimum and maximum measurements. The black dashed line represents the Ecodesign threshold value of the respective emission parameter.

Figure 78 illustrates the comparison of test results regarding emissions and thermal efficiency according ott, repeated EN 13240 tests with the serial-production stoves in the laboratory as well as the "*beReal*" test results in the laboratory and in the field. The black dashed line represents the Ecodesign

ELV of the respective emission parameters. The red bullets represent the results of the stove used for the measurements of this chapter (**Table 32**). The black line in the middle of the box-plots of **Figure 78** represents the median values of test results, the upper sides of the boxes indicate the 75% range of test results and the lower sides of the boxes represent the 25% range of test results. The upper and lower whiskers indicate the minimum and maximum measurements.

Comparing the results of the evaluated stove of this chapter (red bullets of **Figure 78**) with the results of the study of RÖNNBACK et al. [133] (box-plots of **Figure 78**) the same trends are obvious: The emissions of ott tests were lowest and increased for the repeated EN 13240 and "*beReal*" tests. For the efficiency ott results were highest and decreased for the repeated EN 13240 and "*beReal*" tests.

The ott results of the 13 stoves presented in RÖNNBACK et al. [133] were in the range between 500 mg/m^3 and 1250 mg/m^3 for CO, 32 mg/m^3 and 108 mg/m^3 for OGC and 7 mg/m^3 to 40 mg/m^3 for PM emissions. Therefore, all the selected stoves did already meet the future Ecodesign ELV for CO, OGC and PM emissions. The EN 13240 test results measured by the different R&D institutes ranged from 798 mg/m^3 to 2979 mg/m^3 for CO, 21 mg/m^3 to 434 mg/m^3 for OGC and 35 mg/m^3 to 91 mg/m^3 for PM emissions (**Figure 78**). The differences between ott results and repeated EN 13240 test results of R&D institutes were highly significant for CO and PM ($p \leq 0.01$). For OGC emissions the increase was not statistically significant ($p = 0.07$).

The ott results of the thermal efficiencies were in the range between 78.0% and 89.1%. The thermal efficiency test results of repeated EN 13240 tests were in the range of 59.9% and 80.3% (**Figure 78**). Also for test results of thermal efficiencies statistical analysis revealed highly significant differences ($p \leq 0.01$).

Concluding, also the measurements presented in RÖNNBACK et al. [133] showed that ott results of the 13 selected stoves were not reachable with the selected serial-production appliances. The CO and PM emissions were significantly higher and thermal efficiencies were significantly lower for the repeated EN 13240 tests. Consequently, it was confirmed that ott results are far away from the performance of serial-production stoves. That supports the hypothesis that appliances used for ott are different compared to serial-production appliances.

The "*beReal*" test results for the 13 stoves presented in RÖNNBACK et al. [133] and evaluated in the laboratory were in the range between 2410 mg/m^3 and 4506 mg/m^3 for CO, 93 mg/m^3 and 601 mg/m^3 for OGC, 48 mg/m^3 and 108 mg/m^3 for PM emissions and between 64.3% and 78.4% for thermal efficiencies (**Figure 78**). Comparing those results with the test results of repeated EN 13240 tests the differences were highly significant for CO emissions ($p \leq 0.01$) and significant for OGC ($p = 0.03$) as well as PM emissions ($p = 0.049$). No significant difference of thermal efficiencies was observed between EN 13240 test results and "*beReal*" test results at the lab ($p = 0.94$).

The "*beReal*" test results of field measurements at the respective end-users ranged between 2264 mg/m^3 and 5999 mg/m^3 for CO emissions, 54 mg/m^3 and 1435 mg/m^3 for OGC emission and between 49 mg/m^3 and 139 mg/m^3. Thermal efficiencies were in the range of 54.4% to 72.4% (**Figure 78**). The "*beReal*" test results measured at the field sites showed the highest range, but were in general close to the "*beReal*" test results evaluated in the laboratory for gaseous and particulate emissions. The differences between both "*beReal*" tests were statistically not significant regarding the emissions (CO: $p = 0.51$, OGC: $p = 0.50$, PM: $p = 0.33$). However, the thermal efficiencies of "beReal" tests at the field sites were significantly lower compared to "beReal" test results in the laboratory ($p = 0.01$).

Consequently, the analysis indicated that the "*beReal*" test protocol is in general more real-life relevant compared to the EN 13240 test protocol. However, "*beReal*" test results evaluated in the laboratory might still overestimate thermal efficiency. Most probably, this is due to generally higher average draught conditions in real-life heating operation which lead to the significant decrease of thermal efficiencies as already evaluated and described in chapter 3 (**Figure 24**).

7.3.3 Repeatability

For most parameters the EN 13240 test protocol revealed a lower CV and consequently a better repeatability compared to *"beReal"* test protocol (**Figure 79**).

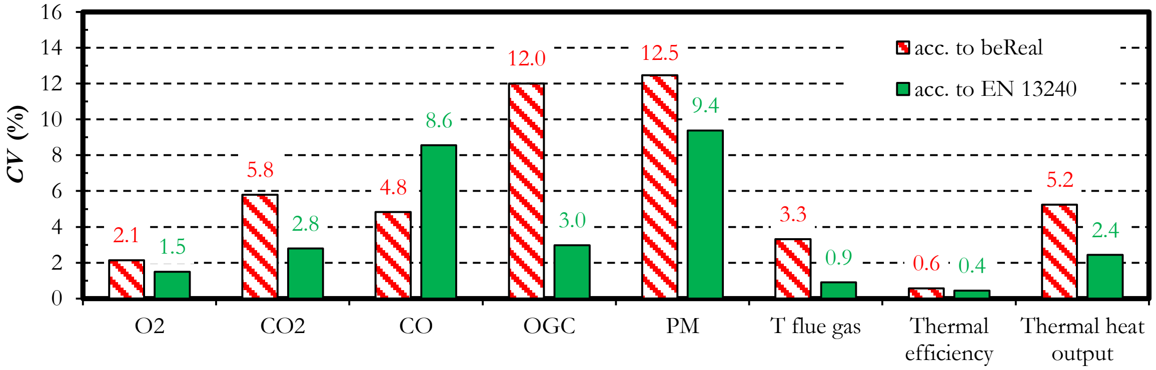


Figure 79: Coefficient of variation (CV) for selected test results according to the *"beReal"* and EN13240 test protocol

The CV of CO emissions was the only parameter which revealed a lower variation of results for *"beReal"* tests compared to EN 13240 test results (**Figure 79**). The CV of all selected parameters was below 12.5% which is similar or even lower comparing CV results from test protocols of advanced biomass cookers ranging from 10% to about 30% [141]. The higher variability of *"beReal"* test results is most probably due to considering the different combustion phases of the heating cycle, especially phases with transient conditions (**Figure 70**). In these phases, that is, ignition, load changes, and cooling down, the variations for single-test cycles might be higher than the variability of selecting two batches at nominal load from a test cycle (**Figure 72**; **Figure 75**).

In a comparison of the results of the CV, it is evident that the EN 13240 test protocol has a higher reliability compared to the *"beReal"* test protocol and is therefore appropriate for setting a benchmark between different products. However, considering the higher variability of operating phases respected by the *"beReal"* test protocol, the reliability of the new test protocol is quite good and suitable for setting a benchmark between different products under more realistic conditions.

7.4 Summary – Official-type-test EN 13240, repeated type test EN 13240 and test protocol "*beReal*"

Two testing concepts for firewood roomheaters were compared in order to assess their differences regarding testing procedure, data evaluation, repeatability and real-life relevance:

- The official-type-test (ott) method according to the European standard (EN 13240)
- A novel real-life oriented test method, called *"beReal"*.

The main important difference comparing both testing concept is, that the ott method according to EN 13240 evaluates the emissions and thermal efficiency of firewood roomheaters only under optimal conditions, which is heated up and at nominal load. In contrast, the *"beReal"* test concept consists of a heating cycle with eight consecutive batches and covers all typical phases of real-life operation, i.e. ignition, heating-up, nominal load, part load and cooling-down. Considering the variable conditions a specific procedure based on volume flow measurements was defined for evaluating the emission and the efficiency of the whole test cycle.

The comparative assessment of the *"beReal"* and EN test protocol was conducted with a serial-production stove. Combustion tests performed according to EN 13240 standard and "*beReal*" were compared with ott results of the respective stove model. The repeatability of EN and *"beReal"* test results regarding emissions and thermal efficiency was evaluated.

The results showed that the ott results for CO, OGC, and PM emissions were not reachable with the used serial-production stove. The repeated EN test results were significantly higher compared to ott results of the used stove model. For example, the deviation was up to 192% for PM emissions. Hence, it seems that the tested product during ott differs from the serial-production products. This is also underpinned by comparing the test results with previous measurements published in the report of RÖNNBACK et al. [133]. Concluding, it seems that future Ecodesign requirements for CO and OGC emissions are hardly reachable with serial-production products, although they have low official-type-test results.

Regarding the repeatability, the comparative evaluation of both test protocols showed a better repeatability for the EN 13240 standard ($CV <$ 9.4% for all parameters). However, the repeatability of the new "*beReal*" test protocol was quite good ($CV <$ 12.4% for all parameters), especially against the background that transient conditions, like ignition, load changes, and cooling down phase are respected.

Compared to the ott results, the *"beReal"* test method resulted in 150% higher CO, 80% higher OGC, and 241% higher PM emissions. The thermal efficiency of *"beReal"* tests was significantly lower compared to repeated EN type test results. However, regarding emissions, no significant differences were observed by the comparative tests according to both test protocols with the serial-production stove.

Although the *"beReal"* test protocol is not capable of evaluating maloperation and the total heating system consisting of the appliance, the chimney and the user behavior, the test protocol evaluates the appliance performance more realistically.

8 CONCLUSIONS[10]

Within this thesis potential measures to improve the real-life performance regarding low emissions and high thermal efficiency of firewood room heating appliances were identified and evaluated.

A user survey shed light on how different types of appliances are typically operated by the end-users. The common user behavior of firewood roomheaters, tiled stoves and biomass cookers was quite similar. Typically the users operate their appliances with hardwood. As expected before, a general fuel overloading of appliances was not indicated by the respondents. In most cases, air valve settings are either set to constant settings or adjusted once per batch. Unfortunately, air valve settings are typically not closed after finishing heating operation. This implicates high thermal losses during cooling down and stand-by phases. Draught conditions were generally estimated as "optimal". For lighting the first fuel batch the bottom-up ignition technique is predominantly used. An interesting insight was that only one third of respondent users operate their appliance knowingly according to the manual whereas another third declared that they knowingly do not operate the stoves according to the manual.

Following the information of the survey, the questions about the effects of draught conditions on combustion performance regarding emissions and thermal efficiency as well as "*if the top-down ignition is generally preferable for all technologies?*" were answered by experimental combustion tests. This was the first time that both parameters, i.e. ignition technique and draught conditions, were evaluated systematically at different serial-production appliances. Draught conditions significantly correlated with thermal efficiency performance and showed an appliance specific increase or decrease of gaseous emissions. For PM emissions no effect of draught conditions were observed. A general advantage of the top-down ignition technique for all appliances was not confirmed by the experiments. This contradicts the content of many leaflets for correct heating which suggest generally the top-down ignition technique as most advantageous regrading low emissions.

Consequently, non-technological optimization measures, like external training arrangements or Quick-User- Guides, containing the most relevant specifications for environmental friendly operation on one page, are suggested. They should clearly specify correct heating operation. This implements the right

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[10] Segments of this section have already been published in [1]-[6].

choice of fuel and fuel properties, the fuel amount per batch, correct air valve settings during and after heating operation and the procedure of emission minimized ignition. However, the ignition technique should be defined appliance specifically. Moreover, non-technological optimization measures should be supported and enhanced by the development and implementation of technological measures. For instance, the development and implementation of automatically air control systems for correct adapted combustion air supply would not only reduce gaseous and particulate emissions but also increase thermal efficiency during heating operation. The automatically adapted air supply system should also respect different draught conditions. For example, this could be achieved by the implementation of an O_2 sensor. This system would significantly limit incorrect user behavior. Furthermore, such add-on devices will increase the user comfort during heating operation. Additionally, it could be used for an automatically close of air valves after finishing heating operation for avoidance of thermal heat losses during cooling down and stand-by conditions. Consequently, this will enhance not only the efficiency factor but might also decrease emissions since the heat demand of the room will be lower resulting on lower operation times.

Possibly different kinds of secondary abatement technologies, like catalysts or filters should be combined with the mentioned non-technological and technological measures in order to minimize emissions also in transient conditions or during maloperation.

In this thesis two commercial oxidizing honeycomb catalysts integrated in firewood room heating appliances were analyzed and evaluated regarding their potential for emission reduction and their applicability under critical operating conditions and long term durability. The catalytic efficiency was evaluated using a novel methodology and a specific test-setup (DemoCat). Thereby, it was possible to evaluate conversion characteristics under real firewood combustion conditions.

The combustion experiments showed reductive effects on gaseous (CO/ OGC) and particulate (PM) emissions. The highest net emission reduction potential was observed for CO (~90%), followed by OGC (~30%) and PM emissions (~20%). All tested catalyst integrated solutions showed the potential to meet future Ecodesign emission limit values, even under a close-to-real-life test cycle. Furthermore, the combustion experiments confirmed that the elevated temperature level at the catalyst due to the integration design promotes better catalytic conversion rates, especially for catalytically convertible OGC emissions and deposited soot or organic compounds on the catalyst surface. Additionally, the most effective catalytic emission reduction in terms of absolute emission reduction is achievable during the start-up and burn-out phases of a firewood batch. Consequently, the use of integrated honeycomb converters and implemented primary measures revealed highly synergetic effects. However, the impact of the catalyst´s carrier material on primary combustion conditions overlay the catalytic conversion performance. This so called primary effect, which was quantified for the first time within this study, has to be considered during the development process since it could negatively influence primary combustion conditions. Therefore, primary optimization of catalyst integrated solutions should be conducted with integrated uncoated catalyst carriers (dummies).

The experiments of the applicability of the integrated catalysts during critical operating conditions and long term heating operation confirmed the risk of blocking the catalyst by carbonaceous and mineral agglomerated deposits. Therefore, integrated honeycomb catalysts need a regular cleaning of about three to five times per year to enable safe and efficient operating conditions. Technological add-ons monitoring the increase of pressure drop in combination with chimney draught conditions appear as a possible solution to indicate a need for cleaning.

The comparative analysis and evaluation of the official-type-test method and the advanced test protocol, called "*beReal*", showed that the EN test protocol has indeed a better repeatability, but evaluates the appliance performance not realistically. The analysis confirmed that for realization of a further emission reduction in real-life operation and for pushing technological development further, new real-life oriented test protocols, like "*beReal*" are needed. In principal, the testing procedures have to reflect real-life heating operation as closely as possible, but also ensure repeatability and reproducibility of the tested parameters. Transient phases, like cooling down, load changes, and ignition processes, shall be included in the testing cycle since they typically occur in each heating cycle in real-life. Suitable measuring methods for evaluating the different heating phases have to be applied and a quality assurance concept must guarantee the correct application and evaluation of measurements. If necessary, emission limit values and efficiency requirements have to be adapted to new test methods. In order to guarantee the quality of serial-production appliances legal initiatives should establish an efficient market surveillance to guarantee that testing results are valid for serial-production products that are sold on the market.

9 OUTLOOK[11]

This thesis evaluated the typical user behavior of firewood room heating appliances and identified specific aspects of the user behavior which can be improved by non-technological measures. For example, the application of a technology specific ignition technique, correct air valve settings and appropriate chimney draught conditions can significantly improve the real-life operation performance of firewood stoves.

Future work should focus on the demonstration of the effectiveness of the proposed optimization measures of user behavior, like external training arrangements or appliance specific Quick-User-Guides. This should be done by comparative combustion tests and by long term field measurements at the end-user before and after implementing the respective optimization measures. In parallel, the effectiveness of optimization measures should be validated and demonstrated by ambient air quality monitoring. This methodology would also enable a better quantification of the optimization potential of real-life emission and efficiency performance of optimized user behavior.

Furthermore, future research work should focus on the effectiveness of the combined optimization approach of technological and non-technological primary measures and on suitable secondary abatement technologies under real-life conditions.

As presented in this thesis oxidative honeycomb catalysts integrated in firewood stoves reveal a promising solution to decrease emissions significantly during real-life operation, even under off-specification operating conditions. This approach appears as a close-to-market solution. However, from a technical point of view, there are still open research topics which need clarification. For example, the effect of the catalytic process on PM emission composition and on different species of gaseous organic compounds, e.g. PAH like Benzo(a)pyrene is still not totally clear. For example, recent combustion experiments with PM sampling in diluted flue gases indicated the risk that PAH and other organic compounds might be nitrated to highly genotoxic nitro-PAH due to catalytic reactions. The nitrification of PAH due to catalytic reactions was already observed during combustion experiments of diesel-soot [142] and therefore clarification is needed.

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[11] Segments of this section have already been published in [1]-[6].

Additionally, investigations on irreversible catalyst deactivation mechanisms, e.g. poisoning due to the use of sulfur rich material, like litter etc. and quantification of lifetime as integrated solution are necessary. Therefore, SEM/EDX analysis should be applied with systematically aged catalysts and varied fuel qualities.

A market penetration of catalytic systems would require different legal framework conditions. For instance advanced real-life oriented test methods in combination with challenging emission limits or obligatory field testing could require (secondary) emission reduction measures for most products. In such a scenario, catalysts as simple and relatively cheap solutions would have economic advantages over alternative emission abatement technologies. Further research on the economic viability and suitable instruments to penetrate the market with appropriate secondary emission abatement technologies is needed.

The implementation of a real-life oriented test method, e.g. the *"beReal"* test protocol, in a label or a standard was proposed as an effective non-technological measure to realize an emission reduction in real-life operation and to push technological development further.

Currently, different associations and stove manufactures think about the option to implement the *"beReal"* test protocol as a voluntary labelling scheme. Therefore, the whole label framework conditions, e.g. legal form, management structure, an appropriate market surveillance concept, etc., has to be defined. Additionally, an adequate benchmark system has to be established which specifies the minimum requirements of emissions and efficiency which labeled products have to achieve. Therefore, further measurements to establish such a benchmark system are needed in order to set challenging requirements which are only achievable by best-performing products. Subsequently, the superiority of labeled products compared to not-labeled products regarding real-life performance has to be demonstrated by field monitoring.

A further possibility to apply the *"beReal"* test protocol appears in the standardized assessment of emission factors. The benefit of measuring emission factors according to a suitable test concept is a standardized monitoring of technological development and a regular update of emission inventories.
This application of the *"beReal"* test protocol requires further investigations, especially comparative lab and field tests with different types of appliances. Based on those results it might be necessary to adapt the *"beReal"* test procedure.

REFERENCES

[1] G. REICHERT, C. SCHMIDL, W. HASLINGER, M. SCHWABL, S. AIGENBAUER, M. WÖHLER, C. HOCHENAUER, Investigation of user behavior and assessment of typical operation mode for different types of firewood room heating appliances in Austria, Renew. Energy 93 (2016) 245-254. https://doi.org/10.1016/j.renene.2016.01.092.

[2] G. REICHERT, H. HARTMANN, W. HASLINGER, H. OEHLER, R. MACK., C. SCHMIDL, C. SCHÖN, M. SCHWABL, H. STRESSLER, R. STURMLECHNER, C. HOCHENAUER, Effect of draught conditions and ignition technique on combustion performance of firewood roomheaters, Renew. Energy 105 (2017) 547-560. https://doi.org/10.1016/j.renene.2016.12.017.

[3] G. REICHERT, C. SCHMIDL, W. HASLINGER, H. STRESSLER, R. STURMLECHNER, M. SCHWABL, N. KIENZL, M. WÖHLER, C. HOCHENAUER, Long term durability and safety aspects of oxidizing honeycomb catalysts integrated in firewood stoves, Biomass Bioenergy 105 (2017) 428-442. http://dx.doi.org/10.1016/j.biombioe.2017.07.018.

[4] G. REICHERT, C. SCHMIDL, W. HASLINGER, H. STRESSLER, R. STURMLECHNER, M. SCHWABL, M. WÖHLER, C. HOCHENAUER, Catalytic efficiency of oxidizing honeycomb catalysts integrated in firewood stoves evaluated by a novel measuring methodology under real-life operating conditions, Renew. Energy 117 (2018) 300-313. https://doi.org/10.1016/j.renene.2017.10.065.

[5] G. REICHERT, C. SCHMIDL, W. HASLINGER, H. STRESSLER, R. STURMLECHNER, M. SCHWABL, C. HOCHENAUER, Novel method evaluating real-life performance of firewood roomheaters in Europe, Energy Fuels 3 (2), (2018) 1874-1883. https://doi.org/10.1021/acs.energyfuels.7b03673.

[6] G. REICHERT, C. SCHMIDL, W. HASLINGER, H. STRESSLER, R. STURMLECHNER, M. SCHWABL, M. WÖHLER, C. HOCHENAUER, Impact of oxidizing honeycomb catalysts integrated in firewood stoves on emissions under real-life operating conditions, Fuel Process. Technol. 177 (2018) 109-118. https://doi.org/10.1016/j.fuproc.2018.04.016.

[7] A. DEMIRBAS, Potential applications of renewable energy sources, biomass combustion problems in boiler power systems and combustion related environmental issues, Prog. Energy Combust. Sci. 31 (2) (2005) 171-192. http://dx.doi.org/10.1016/j.pecs.2005.02.002.

[8] J. MIGUREZ, J. MORAN, E. GRANADA, J. PORTEIRO, Review of technology in small scale biomass combustion systems in the European market, Renew. Sustain. Energy Rev. 16 (6) (2012) 3867-3875, http://dx.doi.org/10.1016/j.rser.2012.03.044.

[9] IEA, World Energy Outlook, Special Report, Energy and Air Pollution, 2016. Online available: http://www.iea.org/publications/freepublications/publication/WorldEnergyOutlookSpecialReport2016EnergyandAirPollution.pdf (accessed 28/05/2018).

[10] M.P. KSHIRSAGAR, V.R. KALAMKAR, A comprehensive review on biomass cookstoves and a systematic approach for modern cookstove design, Renew. Sust. Energ. Rev. 30 (2014) 580-603. https://doi.org/10.1016/j.rser.2013.10.039.

[11] EC DG TREN, Preparatory Studies for Eco-design Requirements of EuPs (II) – LOT 15 – Solid Fuel Small Combustion Appliances, Task 2: Economic and Market Analysis, France, 2009, pp. 1-47. Online available: https://www.eceee.org/static/media/uploads/site-2/ecodesign/products/solid-fuel-small-combustion-installations/bio-eup-lot-15-task2-final.pdf (accessed 28/05/2018).

[12] G. BENKE, K. LEUTGÖB: Einzelöfen in Österreich. Marktbeschreibung, Endbericht: Studie der e7 Energie Markt Analyse GmbH im Auftrag von Umweltbundesamt Wien, Februar 2011, 37 p.

[13] ÖNORM EN 13240:2001 + AC:2003, Roomheaters Fired by Solid Fuel - Requirements and Test Methods, Austrian Institute for Standardization, Vienna, 2003.

[14] ÖNORM EN 13229:2006, Inset Appliances Including Open Fires Fired by Solid Fuels – Requirements and Test Methods, Vienna, 2006.

[15] ÖNORM EN 12815:2007, Residential Cookers Fired by Solid Fuel e Requirements and Test Methods, Austrian Institute for Standardization, Vienna, 2007.

[16] ÖNORM EN 15250:2007, Slow Heat Release Appliances Fired by Solid Fuel – Requirements and Test Methods, Vienna, 2007.

[17] ÖNORM EN 15544:2009, One off Kachelgrundöfen/Putzgrundöfen (Tiled/ mortared Stoves) – Dimensioning, Austrian Institute for Standardization, Vienna, 2009.

[18] S. BRAM, J. DE RUYCK, D. LAYRIC, Using biomass: a system perturbation analysis, Appl. Energy 86 (2009) 194-201. http://dx.doi.org/10.1016/j.apenergy.2012.03.034.

[19] R. MADLENER, M. KOLLER, Economic and CO_2 mitigation impacts of promoting biomass heating systems: an input-output study for Vorarlberg, Austrian Energy Policy 35 (2007) 602-635. http://dx.doi.org/10.1016/j.enpol.2007.08.023.

[20] COMMISSION OF THE EUROPEAN COMMUNITIES, Limiting Global Climate Change to 2 °C: the Way Ahead for 2020 and beyond. Brussels, 2007. Online available: http://eur-lex.europa.eu/legal-content/EN/TXT/?qid=1525425993562&uri=CELEX:52007SC0008 (accessed 28/05/2018).

[21] C. SCHMIDL, M. LUISSER, E. PADOUVAS, L. LASSELSBERGER, M. RZACA, C. RAMIREZ- SANTA CRUZ, M. HANDLER, H. BAUER, H. PUXBAUM, Particulate and gaseous emissions from manually and automatically fired small scale combustion systems, Atmos. Environ. 45 (2011) 7443-7454. http://dx.doi.org/10.1016/j.atmosenv.2011.05.006.

[22] L.S. BÄFVER, B. LECKNER, C. TULLIN, M. BERNTSEN, Particle emissions from pellets stoves and modern and old-type wood stoves, Biomass Bioenergy 35 (2011) 3648-3655. https://doi.org/10.1016/j.biombioe.2011.05.027.

[23] R.S. CHAPMAN, W.P. WATKINSON, K.L. DREHER, D.L. COSTA, Ambient particulate matter and respiratory and cardiovascular illness in adults: particle-borne transition metals and the heart-lung axis, Environ. Toxicol. Pharmacol. 4 (1997) 331-338. http://dx.doi.org/10.1016/S1382-6689(97)10031-X.

[24] N. ENGLERT, Fine particles and human health e a review of epidemiological studies, Toxicol. Lett. 149 (2004) 235-242. https://doi.org/10.1016/j.toxlet.2003.12.035.

[25] D.B. PEDEN, The epidemiology and genetics of asthma risk associated with air pollution, J. Allergy Clin. Immunol. 115 (2005) 213-219. https://doi.org/10.1016/j.jaci.2004.12.003.

[26] M.A. BARI, G. BAUMBACH, B. KUCH, G. SCHIFFKNECHT, Particle-phase concentrations of polycyclic aromatic hydrocarbons in ambient air of rural residential areas in southern Germany, Air Qual. Atmos. Health 3 (2010) 103-116. https://doi.org/10.1007/s11869-009-0057-8.

[27] J. KELZ, T. BRUNNER, I. OBERNBERGER, P. JAVALA, M.-R. HIRVONEN, PM emissions from old and modern biomass combustion systems and their health effects, in: Proceedings of the 18th European Biomass Conference & Exhibition, ETA-Florence Renewable Energies, 2010, pp. 1231-1243. Lyon, France. Online available: https://www.bios-bioenergy.at/uploads/media/Paper-Brunner-PM-Healtheffects-2010-06.pdf (accessed 25/05/2018).

[28] M.-R. HIRVONEN, P. JAVALA, M. TAPANAINEN, M. HAPPO, O. USKI, Health related toxicological effects of particulate emission from small-scale biomass combustion systems, in: Central European Biomass Conference 2011, Graz, Austria, 2011.

[29] M. GLASIUS, M. KETZEL, B. JENSEN, J. MØNSTER, R. BERKOWICZ, F. PALMGREN, Impact of wood combustion on particle levels in a residential area in Denmark, Atmos. Environ. 40 (2006) 7115-7124. https://doi.org/10.1016/j.atmosenv.2006.06.047.

[30] M.A. BARI, G. BAUMBACH, J. BRODBECK, M. STRUSCHKA, B. KUCH, W. DREHER, G. SCHIFFKNECHT, Characterization of particulates and carcinogenic polycyclic aromatic hydrocarbons in wintertime wood-fired heating in residential areas, Atmos. Environ. 45 (2011) 7627-7634. https://doi.org/10.1016/j.atmosenv.2010.11.053.

[31] K.E. YTTRI, C. DYE, O.-A. BRAATHEN, D. SIMPSON, E. STEINNES, Carbonaceous aerosols in Norwegian urban areas, Atmos. Chem. Phys. 9 (2009) 2007-2020. https://doi.org/10.5194/acp-9-2007-2009.

[32] A. CASIERO, B. HEIDI, C. SCHMIDL, A. CASIMIRO, H. PUXBAUM, Wood burning impact on PM10 in three Austrian regions, Atmos. Environ. 43 (2009) 2186-2195. https://doi.org/10.1016/j.atmosenv.2009.01.012.

[33] M. ANDERL, S. HAIDER, C. LAMPERT, L. MOOSMANN, K. PAZDERNIK, K. PERL, M. PINTERITS, S. POUPA, M. PURZNER, B. SCHODL, G. STRANNER, M. WIESNER, A. ZECHTMEISTER, Austria's National Inventory Report (IIR) 2015, Umweltbundesamt GmbH, 2015, ISBN 978-3-99004-316-5, p. 372. Vienna, Austria.

[34] COUNCIL DIRECTIVE 12008/50/EC: Directive 2008/50/EC of the European Parliament and of the Council of 21 May 2008 on ambient air quality and cleaner air for Europe, http://eur-lex.europa.eu/legal-content/EN/TXT/PDF/?uri=CELEX:32008L0050&from=DE (accessed 28/05/2018).

[35] X. QUEROL, A. ALASTUEY, C.R. RUIZ, B. ARTINANO, H.C. HANSSON, R.M. HARRISON, E. BURINGH, H.M. TEN BRINK, M. LUTZ, P. BRUCKMANN, P. STRAEHL, J. SCHNEIDER, Speciation and origin of PM_{10} and $PM_{2.5}$ in selected European cities, Atmos. Environ. 38 (2004) 6547-6555. https://doi.org/10.1016/j.atmosenv.2004.08.037.

[36] R. Van DINGENEN, F. RAES, J.-P. PUTAUD, U. BALTENSBERGER, A. CHARRON, M.- C. FACCHINI, S. DECESARI, S. FUZZI, R. GEHRIG, H.-C. HANSSON, R.M. HARRISON, C. HÜGLIN, A.M. JONES, P. LAJ, G. LORBEER, W. MAENHAUT, F. PALMGREN, X. QUEROL, S. RODRIGUEZ, J. SCHNEIDER, H. TEN BRINK, P. TUNVED, K. TORSETH, B. WEHNER, E. WEINGARTNER, A. WIEDENSOHLER, P. WAHLIN, A European aerosol phenomenology – 1: physical characteristics of particulate matter at curbside, urban, rural and background sites in Europe, Atmos. Environ. 38 (2004) 2561-2577. https://doi.org/10.1016/j.atmosenv.2004.01.040.

[37] COMMISSION REGULATION (EU) 2015/1185 of 24, Implementing Directive 2009/125/EC of the European Parliament and of the Council with regard to ecodesign requirements for solid fuel local space heaters, Official J. Eur. Union L193 (April 2015) 1-19, 21.7.2015. Online available: http://www.eceee.org/static/media/uploads/site-2/ecodesign/products/lot-20-local-room-heating-products/celex-32015r1185-en-txt.pdf (accessed 28/05/2018).

[38] C. SCHÖN, H. HARTMANN, Log wood combustion in stoves – influence on emissions and efficiency, ETA Renewable Energies, in: Proceedings 20th European Biomass Conference & Exhibition e from Research to Industry and Markets. Milano, Italy, 18-22 June 2012, Florence, Italy, 2012, pp. 1293-1298. https://doi.org/10.5071/20thEUBCE2012-2CV.4.14.

[39] G. SHEN, M. XUE, S. WEI, Y. CHEN, Q. ZHAO, B. LI, H. WU, S. TAO, Influence of fuel moisture, charge size, feeding rate and air ventilation conditions on the emissions of PM, OC, EC, parent PAHs, and their derivatives from residential wood combustion, J. Environ. Sci. 25 (2013) 1808-1816. http://dx.doi.org/10.1016/S1001-0742(12)60258-7.

[40] C. SCHÖN, H. HARTMANN, P. TUROWSKI, User and fuel impacts on flue gas emissions of a chimney stove, ETA Renewable Energies, in: Proceedings 19th European Biomass Conference & Exhibition - from Research to Industry and Markets. Berlin, Germany, 06-10 June 2012, 2011, pp. 960-966. Florence, Italy. https://doi.org/10.5071/19thEUBCE2011-OA7.4.

[41] T.B. JORDAN, A.J. SEEN, Effect of airflow setting on the organic composition of wood heater emissions, Environ. Sci. Technol. 39 (2005) 3601-3610. https://doi.org/10.1021/es0487628.

[42] J. ORASCHE, S. TORBEN, C. SCHÖN, H. HARTMANN, H. RUPPERT, J.M. ARTEAGA-SALAS, R. ZIMMERMANN, Comparison of emissions from wood combustion. Part 2: impact of combustion conditions on emission factors and characteristics of particle-bound organic species and polycyclic aromatic hydrocarbon (PAH)-related toxicological potential, Energy fuels. 27 (2013) 1482-1491. https://doi.org/10.1021/ef301506h.

[43] J. TISSARI, K. HYTÖNEN, O. SIPPULA, J. JOKINIEMI, The effects of operating conditions on emissions from masonry heaters and sauna stoves, Biomass Bioenergy 33 (2009) 513-520. https://doi.org/10.1016/j.biombioe.2008.08.009.

[44] C. SCHMIDL, I.L. MARR, A. CASEIRO, P. KOTIANOVA, A. BERNER, H. BAUER, A. KASPAR- GIEBL, H. PUXBAUM, Chemical characterization of fine particle emissions from wood stove combustion of common woods growing in mid-European Alpine regions, Atmos. Environ. 42 (2008) 126-141. https://doi.org/10.1016/j.atmosenv.2007.09.028.

[45] P.M. FINE, G.R. CASS, B.R.T. SIMONEIT, Chemical characterization of fine particle emissions from the wood stove combustion of prevalent United States tree species, Environ. Eng. Sci. 21 (2004) 705-721. https://doi.org/10.1089/ees.2004.21.705.

[46] T. NUSSBAUMER, A. DOBERER, N. KLIPPEL, R. BÜHLER, W. VOCK, Influence of ignition and operation type on particle emissions from residential wood combustion, in: Proceedings of the 16th European Biomass Conference and Exhibition, Valencia, Spain, 2008, p. 5. Online available: http://www.verenum.ch/Publikationen/Biomass-Conf9.5.pdf (accessed 28/05/2018).

[47] T. NUSSBAUMER, C. CZASCH, N. KLIPPEL, L. JOHANSSON, C. TULLIN, Particulate Emissions from Biomass Combustion in IEA Countries – Survey on Measurements and Emission Factors, January 2008, ISBN 3-908705-18-5, p. 40. Zürich. Online available: http://www.ieabcc.nl/publications/Nussbaumer_et_al_IEA_Report_PM10_Jan_2008.pdf (accessed 28/05/2018).

[48] H. HARTMANN, C. SCHÖN, P. TUROWSKI, I. OBERNBERGER, F. BIEDERMANN, T. BRUNNER, L. BÄFVER, J. FINNAN, J. CAROLL, Low Emission Operation Manual for

Chimney Stoves, Report within the scope of the ERA-NET Bioenergy Project, FutureBio Tec, Graz, Austria, October 2012, p. 23. Online available: http://futurebiotec.bioenergy2020.eu/files/FutureBioTec-Low%20emission%20operation%20manual%20for%20chimney%20stove%20users.pdf (accessed 28/05/2018).

[49] G. REICHERT, C. SCHMIDL, W. HASLINGER, W. MOSER, S. AIGENBAUER, F. FIGL, M. WÖHLER, Investigation of user behavior and operating conditions of residential wood combustion (RWC) appliances and their impact on emissions and efficiency, 4th Central European Biomass Conference, Graz, Austria, 15-18. January 2014, 373-380. Online available: http://www.biomasseverband.at/veranstaltungen/tagungen-und-vortraege/archiv/4-mitteleuropaeische-biomassekonferenz/biowaerme-simulation-und-effizienzoptimierung/ (accessed 28/05/2018).

[50] F. ELLNER-SCHUBERTH, H. HARTMANN, P. TUROWSKI, P. ROSSMANN, Partikelemissionen aus Kleinfeuerungsanlagen für Holz und Ansätze für Minderungsmaßnahmen, Berichte aus dem TFZ, Straubing, März 2010, p. 134. Online available: http://www.tfz.bayern.de/mam/cms08/festbrennstoffe/dateien/22_bericht.pdf (accessed 28/05/2018).

[51] V. LENZ, D. THRÄN, H. HARTMANN, P. TUROWSKI, F. ELLNER-SCHUBERTH, J. Gerth, Bewertung und Minderung von Feinstaubemissionen aus häuslichen Holzfeuerungsanlagen, 2010, p. 274. DBFZ Report Nr. 1, Leipzig, Germany.

[52] T. BRUNNER, I. OBERNBERGER, R. SCHARLER, Primary measures for low-emission residential wood combustion – comparison of old with optimized modern systems, in: Proceedings of the 17th European Biomass Conference & Exhibition, ETA-renewable Energies, Italy, Hamburg, Germany, June 2009. Online available: http://bios-bioenergy.at/uploads/media/Paper-Brunner-primary-measures-for-low-emission-wood-combustion-Hamburg-2009.pdf (accessed 25/05/2018).

[53] H. KHODAEI, F. GUZZOMI, D. PATIÑO, B. RASHIDIAN, G. H. YEOH, Air Staging Strategies in Biomass Combustion-Gaseous and Particulate Emission Reduction Potentials. Fuel Process. Technol. 157 (2017) 29-41. https://doi.org/10.1016/j.fuproc.2016.11.007.

[54] A. VIREN, L. HEIKKI, J. TISSARI, O. SIPPULA, J. JOKINIEMI, I. OBERNBERGER, F. BIEDERMANN, T. BRUNNER, H. HARTMANN, C. SCHÖN, Guidelines for Low

Emission Chimney Stoves Design, Report within the scope of the ERA-NET Project "FutureBio Tec", Graz, Austria, September 2012. Online available: http://futurebiotec.bioenergy2020.eu/files/FutureBioTec-Guidelines%20for%20Low%20Emission%20Chimney%20Stove%20Design.pdf (accessed 28/05/2018).

[55] M. OBAIDULLAH, V. DYAKOV, J.D. THOMASSIN, T. DUQUESNE, S. BRAM, F. CONTINO, J. DE RUYCK, CO emission measurement and performance analysis of 10 kW and 20 kW wood stoves, Energy Procedia 61 (2014) 2301-2306. https://doi.org/10.1016/j.egypro.2014.12.443.

[56] M. ELSASSER, C. BUSCH, J. ORASCHE, C. SCHÖN, H. HARTMANN, J. SCHNELLE-KREIS, R. ZIMMERMANN, Dynamic changes of the aerosol composition and concentration during different burning phases of wood combustion, Energy fuels. 27 (2013) 4959-4968. https://doi.org/10.1021/ef400684f.

[57] T. KAIVOSOJA, A. VIREN , J. TISSARI, J. RUUSKANEN, J. TARHANEN, O. SIPPULA, J. JOKINIEMI, Effects of a catalytic converter on PCDD/F, chlorophenol and PAH emissions in residential wood combustion, Chemosphere 88 (2012) 278-285. https://doi.org/10.1016/j.chemosphere.2012.02.027.

[58] F. OZIL, V. TSCHAMBER, F. HAAS, L. TROUVE, Efficiency of catalytic process for the reduction of CO and VOC emissions from wood combustion in domestic fireplaces, Atmos. Environ. 90 (2009) 1053-1061. https://doi.org/10.1016/j.fuproc.2009.03.019.

[59] A. HUKKANEN, T. KAIVOSOJA, O. SIPPULA, K. NUUTINEN, J. JOKINIEMI, J. TISSARI, Reduction of gaseous and particulate emissions from small-scale wood combustion with a catalytic combustor, Atmos. Environ. 50 (2012) 16-23. https://doi.org/10.1016/j.atmosenv.2012.01.016.

[60] R. BINDIG, S. BUTT, I. HARTMANN, M. MATTHES, C. THIEL, Application of heterogeneous catalysis in small-scale biomass combustion systems, Catalysts 2 (2012) 223-243. https://doi.org/10.3390/catal2020223.

[61] S. BENSAID, F.A. DEORSOLA, D. FINO, N. RUSSO, After-treatment of household wood-fired stove emissions: from catalyst formulation to full-scale system, Catal. Today 197 (2012) 76-89. https://doi.org/10.1016/j.cattod.2012.06.026.

[62] J. CARNÖ, M. BERG, S. JÄRAS, Catalytic abatement of emissions from small-scale combustion of wood, Fuel 75 (8) (1996) 959-965. https://doi.org/10.1016/0016-2361(96)00047-6.

[63] M. WÖHLER, G. REICHERT, J.-M. KIRCHHOF, M. SCHWABL, C. SCHMIDL, W. HASLINGER, Operation characteristics of a bulk catalyst in a test stand under similar conditions to a firewood stove, in: WSEDnext Joung Researcher's Conference - Biomass, 2013, p. 1-7. Online available: http://biocat.bioenergy2020.eu/img/MWoehler_WSED13_paper_final.pdf (accessed 28/05/2018)

[64] M. FERRANDON, M. BERG, E. BJÖRNBOM, Thermal stability of metal-supported catalysts for reduction of cold-start emissions in a wood-fired domestic boiler, Catal. Today 53 (1999) 647-659, https://doi.org/10.1016/S0920-5861(99)00152-2.

[65] H. BACHMAIER, H. HARTMANN, C. SCHMIDL, Flue gas catalysts for wood combustion: test stand results, in: Proceedings of 22th European Biomass Conference and Exhibition, Hamburg, June 2014, pp. 399-401. https://doi.org/10.5071/22ndEUBCE2014-2AO.4.4.

[66] J.E. HOUCK, P.E. TIEGS, R.C. MC CRILLIS, C. KEITHLEY, J. CROUCH, Air emissions from residential heating: the wood heating option put into environmental perspective, in: Proceedings of a U.S. Environmental Protection Agency and Air and Waste Management Association Conference, Emission Inventory: Living in a Global Environment, 1998, pp. 373-384.

[67] US EPA Method 28 - Certification and Auditing of wood heaters, 1401-1446. Online available: http://www2.epa.gov/sites/production/files/2013-12/documents/m-28.pdf (accessed 28/05/2018).

[68] CSA B415.1-10:2010, Performance Testing of Solid-fuel-burning Heating Appliances, Canadian Standards Association, March 2010, p. 76.

[69] I. OBERNBERGER, C. MANDL, Survey on the Present State of Particle Precipitation Devices for Residential Biomass Combustion with a Nominal Capacity up to 50 kW in IEA Bioenergy Task 32 Member Countries, Final Version, Graz, December 2011. Online available: http://www.bios-bioenergy.at/uploads/media/Filter-study-IEA-Dez-2011.pdf (accessed 28/05/2018).

[70] H. HARTMANN, Status on emissions, regulation and technical improvements and future developments for residential wood burning appliances in Germany, Seminar on wood combustion and air quality in Aarhus/Denmark, March 15th, 2012. Online available: http://www.skorstensfejerfredensborg.dk/upload/5.%20hans%20hartmann,%20tfz.pdf (accessed 28/05/2018).

[71] E. CARLON, M. SCHWARZ; L. GOLICZA, V. K. VERMA, A. PRADA, M. BARATIERI, W. HASLINGER, C. SCHMIDL, Efficiency and operational behavior of small-scale pellet boilers installed in residential buildings. Appl. Energy 155 (2015) 854-865. https://doi.org/10.1016/j.apenergy.2015.06.025.

[72] M. SCHWARZ, M. HECKMANN, L. LASSELSBERGER, W. HASLINGER, Determination of annual efficiency and emission factors of small-scale biomass boiler. Central European Biomass Conference 2011, 26−29, January 2011. Online available: https://www.bioenergy2020.eu/files/publications/pdf/Schwarz_etal_Annual_Efficiency.pdf (accessed 28/05/2018).

[73] EC, in: Draft Minutes-meeting on Emission Measurement Methods for Solid Fuel Local Space Heaters, Brussels, 17 December 2012, 2012.

[74] M. WÖHLER, J.S. ANDERSEN, G. BECKER, H. PERSSON, G. REICHERT, C. SCHÖN, C. SCHMIDL, D. JAEGER, S. K. PELZ, Investigation of real life operation of biomass room heating appliances - results of a European survey, Appl. Energy 169 (2016) 240-249, http://dx.doi.org/10.1016/j.apenergy.2016.01.119.

[75] E. PETTERSON, C. BOMAN, R. WESTERHOLM, D. BOSTRÖM, A. NORDIN, Stove performance and emission characteristics in residential wood log and pellet combustion, part 2: wood stove, Energy Fuels 25 (2011) 315-323. http://dx.doi.org/10.1021/ef1007787.

[76] F. KIENLE, Market development and perspectives, IEA Workshop on "Highly Efficient and Clean Wood Log Stoves", IEA Bioenergy Conference, Berlin, 29 October 2015, p. 47. Online available: http://task32.ieabioenergy.com/wp-content/uploads/2017/03/01-Frank-Kienle.pdf (accessed 22/11/2018).

[77] M. SCHLICHTER, Aktuelle Empfehlungen aus der Überwachung von Kleinfeuerungsanalgen, 22. Sitzung Arbeitskreis Holzfeuerungen, Straubing, 5. June 2018.

[78] H. HANEDER, Biomasse – Heizungserhebung 2017, Landwirtschaftskammer Niederösterreich, St. Pölten, March 2018, p. 20. Online available: https://noe.lko.at/biomasse-heizungserhebung-2017+2500+2552656 (accessed 23/11/2018).

[79] H. HARTMANN, C. SCHÖN, P. TUROWSKI, Richtig heizen. Der Betrieb von Kaminöfen. TFZ-Wissen, Nr.1, Straubing, 2015, p. 31. Online available: http://www.tfz.bayern.de/mam/cms08/festbrennstoffe/dateien/tfz_wissen_b_web_s.pdf (accessed 23/11/2018).

[80] H. HARTMANN, K. REISINGER, K. THUNEKE, A. HÖLDRICH, P. ROSSMANN, Handbuch Bioenergie –Kleinanlagen. Fachagentur Nachwachsende Rohstoffe, Gülzow, March 2007, p. 230. Online available: http://mediathek.fnr.de/media/downloadable/files/samples/b/i/bioenergie_kleinanlagen_2007_erggaenst.pdf (accessed 23/11/2018).

[81] Vereinbarung gemäß Art. 15a B-VG über das Inverkehrbringen von Kleinfeuerungen und die Überprüfung von Feuerungsanlagen und Blockheizkraftwerken, Fassung vom 23.11.2018, Vienna. Online available: https://www.ris.bka.gv.at/GeltendeFassung.wxe?Abfrage=LrW&Gesetzesnummer=20000005 (accessed 23/11/2018)

[82] Erste Verordnung zur Durchführung des Bundes-Immissionsschutzgesetzes (Verordnung über kleine und mittlere Feuerungsanlagen - 1. BImSchV), Bundesministerium der Justiz und Verbraucherschutz, 26.01.2010. Online available: https://www.gesetze-im-internet.de/bimschv_1_2010/BJNR003800010.html (accessed 23/11/2018).

[83] W. SCHIEDER, A. STORCH, D. FISCHER, P. THIELEN, A. ZECHMEISTER, S. POUPA, S. WAMPL, Luftschadstoffausstoß von Festbrennstoff-Einzelöfen – Untersuchung des Einflusses von Festbrennstoff-Einzelöfen auf den Ausstoß von Luftschadstoffen, Umweltbundesamt GmbH, 2013, ISBN 978-3-99004-253-3, Wien, 2013, p. 368. Online available: http://www.umweltbundesamt.at/fileadmin/site/publikationen/REP0448.pdf (accessed 28/05/2018).

[84] ÖNORM B 803:1999, Dimensioning of Tiled Stoves - Testing, Austrian Institute for Standardization, Vienna, 1999.

[85] P. SPITZER, G. ENZINGER, G. FANKHAUSER, W. FRITZ, F. GOLJA, R. STIGLBRUNNER, Emissionsfaktoren für feste Brennstoffe, Endbericht, Johanneum Research, 1998, p. 36. Online available: https://www.wien.gv.at/kontakte/ma22/studien/pdf/brennstoffe-zus.pdf (accessed 09/06/2018).

[86] I. LEHNER (CEO Austroflamm GmbH), Use of waste material in firewood roomheaters Final project Meeting of project "Ofenprüfung 2020", Wieselburg, Austria, 25.03.2014 (Personal communication).

[87] C. SCHÖN, H. HARTMANN, Nutzer- und Brennstoffeinflüsse auf Feinstaubemissionen aus Kleinfeuerungsanlagen, Berichte aus dem TFZ, Nr. 36, Technologie- und Förderzentrum (TFZ), Straubing, 2014, p. 180. Online available: http://www.tfz.bayern.de/mam/cms08/festbrennstoffe/dateien/36_bericht.pdf (accessed 28/05/2018).

[88] G. REICHERT, H. HARTMANN, W. HASLINGER, H. ÖHLER, S. PELZ, C. SCHMIDL, M. SCHWABL, H. STRESSLER, R. STURMLECHNER, M. WÖHLER, C. HOCHENAUER, "beReal" - Development of a new test method for firewood roomheaters reflecting real life operation, Proceedings of 24th European Biomass Conference and Exhibition, 6-9 June, 2016, Amsterdam, Netherlands, 382-387. http://dx.doi.org/10.5071/24thEUBCE2016-2AO.2.1.

[89] Schornsteinwerk K. -H. Schreyer GmbH, https://commons.wikimedia.org/wiki/File:LAS-Schnitt.jpg (accessed 29/11/2018)

[90] E.D. VICENTE, M.A. DUARTE, A.I. CALVO, T.F. NUNES, L. TARELHO, C.A. Alves, Emission of carbon monoxide, total hydrocarbons and particulate matter during wood combustion in a stove operating under distinct conditions, Fuel Process. Technol. 131 (2015) 182-192. http://dx.doi.org/10.1016/j.fuproc.2014.11.021.

[91] G. REICHERT, C. SCHMIDL, S. AIGENBAUER, F. FIGL, H. STRESSLER, W. HASLINGER, in: Development of a New Test Method for Residential Wood Combustion (RWC) Appliances Focusing on Real Life Operation, 22nd European Biomass Conference and Exhibition, Hamburg, Germany, 2014, pp. 373-380. http://dx.doi.org/10.5071/22ndEUBCE2014-2AO.1.3.

[92] ÖNORM EN 14961-5:2011, Solid Biofuels – Fuel Specifications and Classes – Part 5: Firewood for Non-industrial Use, Austrian institute for standardization, Vienna, 2011.

[93] ÖNORM EN 14774-1:2009, Solid Biofuels - Determination of Moisture Content – Oven Dry Method – Part 2: Total Moisture – Simplified Method, Austrian institute for standardization, Vienna, 2009.

[94] ÖNORM EN 14918:2010, Solid Biofuels – Determination of Calorific Value, Austrian institute for standardization, Vienna, 2010.

[95] ÖNORM EN 14775:2009, Solid Biofuels – Determination of Ash Content, Austrian institute for standardization, Vienna, 2009.

[96] ÖNORM EN 15104:2011, Solid Biofuels – Determination of Total Carbon, Hydrogen and Nitrogen e Instrumental Methods, Austrian institute for standardization, Vienna, 2011.

[97] ÖNORM EN 15289:2011, Solid Biofuels – Determination of Total Content of Sulfur and Chlorine, Austrian institute for standardization, Vienna, 2011.

[98] CEN/TS 15883:2009, Residential Solid Fuel Burning Appliances e Emissions Test Methods, German Version, Beuth Verlag, 2009, pp. 1-28.

[99] J.D. MCDONALD, B. ZIELINSKA, E.M. FUJITA, J.C. SAGEBIEL, J.C. CHOW, J.G. WATSON, Fine particle and gaseous emission rates from residential wood combustion, Environ. Sci. Technol. 34 (2000) 2080-2091. http://dx.doi.org/10.1021/es9909632.

[100] R. ESSER, R. HEINEN, D. KALKOWSKI, in: Heizen mit Holz – So geht's richtig, Fachagentur Nachwachsende Rohstoffe (FNR), Gülzow e Prüzen, Germany, 2014, p. 20. Online available: https://www.schornsteinfeger-mv.de/verbraucher/pdf-kunden/richtig_heizen_fnr_web.pdf (accessed 28/05/2018).

[101] J. PRAPAS, M.E. BAUMGARDNER, A.J. MARCHESE, B. WILLSON, M., Influence of chimneys on combustion characteristics of buoyantly driven biomass stoves, Energy Sustain. Dev. 23 (2014) 286-293. http://dx.doi.org/10.1016/j.esd.2014.08.007.

[102] R. MACK, D. KUPTZ, C. SCHÖN, H. HARTMANN, Optimierungspotenziale bei Kaminöfen, Berichte aus dem TFZ, Nr. 57, Technologie- und Förderzentrum im Kompetenzzentrum für Nachwachsende Rohstoffe (TFZ), Straubing, 2018, p. 116. Online

available: http://www.tfz.bayern.de/festbrennstoffe/publikationen/index.php (accessed 29/11/2018)

[103] TATAREK RT-08OS-G, https://www.klimaworld.com/tatarek-abbrandsteuerung-trockene-kamine-rt-08-os-grafik-ii-td-titanium.html (accessed 26/11/2018)

[104] J. B. ILLERUP, B. B. HANSEN, W. LIN, J. NICKELSEN, K. DAM JOHANSEN, Intelligent heat system – High-Energy efficient wood stoves with low emissions, Seminar on real-world emissions from residential wood combustion, Copenhagen, 3 December, 2015, p. 24. Online available: https://envs2.au.dk/Projekter/Seminar_RealWorldEmissions/Illerup_RealWorldEmissions.pdf (accessed 29/11/2018)

[105] Principle of external draught control devices, https://www.kutzner-weber.de/deu/produkte/zugbegrenzer.html (accessed 29/11/2018)

[106] Zugbegrenzer ZUK 130 DW, https://www.kutzner-weber.de/deu/produkte/zugbegrenzer/zugbegrenzer-zuk-130-dw.html (accessed 29/11/2018)

[107] FLORIAN, fumis by ATech Electronics, http://www.fumis.si/en/florian (accessed 29/11/2018)

[108] M. WÖHLER, D. JAEGER, S. K. PELZ, H. THORWARTH, Potential of integrated emission reduction systems in a firewood stove under real life operation conditions, Energy Fuel 31 (7) (2017) 7562–7571. https://doi.org/10.1021/acs.energyfuels.7b00803.

[109] A. BEHR., D. AGAR., J. JORISON, Andreas J. Vorholt, Einführung in die technische Chemie, 2. Auflage, Springer-Verlag GmbH Deutschland, Berlin, 2016, p. 316. https://doi.org/10.1007/978-3-662-52856-3.

[110] File:CatalysisScheme.png, https://commons.wikimedia.org/wiki/File:CatalysisScheme.png (accessed 29/11/2018)

[111] N. M. MLADENOV, Modellierung von Autoabgaskatalysatoren, genehmigte Dissertation, Universität Karlsruhe (TH), Karlsruhe, 2009, p. 180. Online available: https://www.itcp.kit.edu/deutschmann/download/09_Mladenov_DrIng_UKA.pdf (accessed 29/11/2018)

[112] Buch-Kat-Technik G. P. MERKER, C. SCHWARZ (Hrsg.), Grundlagen Verbrennungsmotoren, Simulation der Gemischbildung, Verbrennung, Schadstoffbildung und Aufladung, 4. Auflage, Vieweg+Teubner, Wiesbaden, 2009, p. 619. Online available: https://link.springer.com/book/10.1007%2F978-3-8348-9344-4 (accessed 29/11/2018)

[113] VDI 3476-1, Guideline: Waste gas cleaning-Methods of catalytic waste gas cleaning, Fundamentals, Kommission Reinhaltung der Luft im VDI und DIN - Normenausschuss KRdL, Beuth Verlag GmbH, Berlin, 2005, p. 2-25.

[114] R. MATTERN, Heterogene Katalyse, Prozessschritte, 15 August 2010, https://commons.wikimedia.org/wiki/File:Heterogene_Katalyse_2.svg (accessed 29/11/2018)

[115] J. A. MOULIJN, A. E. VAN DIEPEN, F. KAPTEIJN, Catalyst deactivation: is it predictable? What to do? Applied Catalysis A: General 212 (2001) 3–16. https://doi.org/10.1016/S0926-860X(00)00842-5.

[116] C.H. BARTHOLOMEW, Mechanisms of catalyst deactivation, Appl. Catal. General 2012 (2001) 17-60. http://dx.doi.org/10.1016/S0926-860X(00)00843-7.

[117] http://www.chemgapedia.de/vsengine/vlu/vsc/de/ch/10/heterogene_katalyse/katalysator_desaktivierung/katalysator_desaktivierung.vlu.html (accessed 29/11/2018)

[118] Carl Warrlich GmbH, Falkner Landstraße 9; 99830 Treffurt, Germany, http://www.warrlich.de/, carl@warrlich.de.

[119] M. KALTSCHMITT, H. HARTMANN, H. HOFBAUER, Energie aus Biomasse. Grundlagen, Techniken und Verfahren. 2. Auflage, Springer, Berlin Heidelberg, 2009.

[120] L. THEODORE, Nanotechnology: Basic Calculations for Engineers and Scientists, Wiley-Interscience, United States of America, 2006, p. 458.

[121] P. WHITMORE J., Thermo- and diffusiophoresis for small aerosol particles, J. Aerosol Sci. 12 (1) (January 1981) 1-9. https://doi.org/10.1016/0021-8502(81)90003-3.

[122] K. LEONG H., Thermophoresis and diffusiophoresis of large aerosol particles of different shapes, J. Aerosol Sci. 15 (4) (January 1984) 511-517. https://doi.org/10.1016/0021-8502(84)90047-8.

[123] R. BINDIG, I. HARTMANN, C. KOCH, M. MATTHES, M. SCHENKER, C. THIEL, M. KRAUS, U. ROLAND, W.-D. EINICKE, Exhaust gas purification at small-scale biomass combustion and experimental investigation of a combination of catalytic and electrostatic exhaust aftertreatment, Chem. Ing. Tech. 83 (12) (2011) 2105-2120. https://doi.org/10.1002/cite.201100062.

[124] R.B. ANDERSON, K.C. STEIN, J.J. FEENAN, L.J.E. Hofer, Catalytic oxidation of methane, Ind. Eng. Chem. 53 (1961) 809-812. https://doi.org/10.1021/ie50622a024.

[125] CLARIANT PRODUKTE (Deutschland) GMBH, Product Fact Sheet – Catalysts for Wood Stoves: ENVICAT® LONGLIFE PLUS, Material Numbers: 24602828008 (Ceramic Catalyst), 24662528008 (Metallic Catalyst), March 2015. www.clariant.com.

[126] T. NUSSBAUMER, Overview on Technologies for Biomass Combustion and Emission Levels of Particulate Matter, Verenum Press, Zürich, Switzerland, 7 May 2010, p. 79. Online available: http://cmapspublic.ihmc.us/rid=1PV8Z4D92-24JH8TC-2JS7/Nussbaumer_EGTEI-Report_final.pdf (accessed 13/06/2018).

[127] N. JANKOWSKI, C. SCHMIDL, I.L. MARR, H. BAUER, H. PUXBAUM, Comparison of methods for the quantification of carbonate carbon in atmospheric PM10 aerosol samples, Atmos. Environ. 42 (2008) 8055-8064. http://dx.doi.org/10.1016/j.atmosenv.2008.06.012.

[128] I. OBERNBERGER, Aerosols from Small-scale Biomass Combustion Plants - Introduction. IEA Bioenergy Task 32 Workshop: Aerosols from Small-scale Biomass Combustion Plants 27th January 2011, Graz, Austria, 2011. Online available: http://www.ieabcc.nl/workshops/task32_2011_graz_aerosols/01_Obernberger_Introduction.pdf. (accessed 28/05/2018).

[129] http://www.bereal-project.eu/ (accessed 28/05/2018).

[130] R. STURMLECHNER, G. REICHERT, H. STRESSLER, C. SCHMIDL, M. SCHWABL, W. HASLINGER, H. ÖHLER, J. BACHMAIER, R. MACK, H. HARTMANN, M. WÖHLER, beReal – Development of a new testing method close to real-life for domestic biomass room heaters. ProScience 3 (2016) 123-128. https://doi.org/10.14644/dust.2016.020.

[131] G. REICHERT, C. SCHMIDL, H. STRESSLER, R. STURMLECHNER, C. HOCHENAUER, H. HARTMANN, C. SCHÖN, R. MACK, H. ÖHLER, The “beReal” Project - Scientific Highlights. IEA Bioenergy Task 32 workshop: Practical test methods for

small-scale furnaces, 5. Central European Biomass Conference, 18–20 January, 2017, Graz, Austria. Online available: http://task32.ieabioenergy.com/wp-content/uploads/2017/03/14.20_The__beReal__Project_-_Scientific_Highlights.pdf (accessed 28/05/2018)

[132] G. M. JESPERSEN, J. H. JENSEN, M. RÖNNBACK, H. PERSSON, M. WÖHLER, Deliverable D8.1 - Report on Round Robin tests. 30 September 2016, p. 48. Online available: http://www.bereal-project.eu/uploads/1/3/4/9/13495461/d8._1_round_robin_report_final_1.pdf (accessed 28/05/2018).

[133] M. RÖNNBACK, H. PERSSON, G. M JESPERSEN, J. H. JENSEN, Deliverable D7.1 - Documentation and evaluation of field data demonstration. 30 September 2016, p. 36. Online available: http://www.bereal-project.eu/uploads/1/3/4/9/13495461/d7.1_documentation_and_evaluation_of_field_data_demonstration_final_1.pdf (accessed 28/05/2018).

[134] CONTURA 586 of the company NIBE AB: For detailed information, see http://www.contura.eu/de/deutsch/kaminofen/kaminofen/kaminofen-contura-586-style/ (accessed 28/05/2018).

[135] VDI 2066-1 Guideline: Particulate matter measurement: Dust measurement in flowing gases - Gravimetric determination of dust load. Beuth Verlag GmbH, 10772 Berlin, November 2006, p. 111.

[136] G. REICHERT, R. STURMLECHNER, H. STRESSLER, M. SCHWABL, C. SCHMIDL, H. ÖHLER, R. MACK, H. HARTMANN, Deliverable D3.3-Final Report: Definition of Suitable Measurement Methods and Advanced Type Testing Procedure for Real Life Conditions. 30 September 2016, p. 35. Online available: http://www.bereal-project.eu/uploads/1/3/4/9/13495461/d3.3_definition_of_suitable_measurement_methods_final_1.pdf (accessed 28/05/2018).

[137] http://www.halbmikrotechnik.de/service/chemie/energie/fossile_energie/holz-verkokung/holz.htm (accessed 13/06/2018).

[138] M. SATLLER, EN-PME-TEST: Determination of particulate matter emissions from solid biomass fuel burning appliances and boilers - Proposal for a common European test method.

IEA Bioenergy Task 32 workshop: Practical test methods for small-scale furnaces, 5. Central European Biomass Conference, 18−20 January, 2017, Graz, Austria. Online available: http://task32.ieabioenergy.com/wp-content/uploads/2017/03/16.00_EN-PME-TEST.pdf (accessed 28/05/2018)

[139] R. MACK, H. HARTMANN, F. VOLZ, Retrofit controlling units and modern draught stabilizers for stoves. Workshop of the Project ERA-NET Bioenergy "WoodStoves 2020", Stockholm, 13 June, 2017. Online available: http://www.tfz.bayern.de/mam/cms08/en/dateien/vortrag_mack_retrofit_controlling_units_and_modern_draught_stabilizers_for_stoves_2017.pdf (accessed 28/05/2018).

[140] I. SCHÜSSLER, Evaluation of metal based mesh catalysts for stoves. Workshop of the Project ERA-NET Bioenergy "WoodStoves 2020", Stockholm, 13 June, 2017. Online available: http://www.tfz.bayern.de/mam/cms08/en/dateien/vortrag_schuessler_selection_and_testing_of_medium_temperature_metal_based_mesh_catalysts_for_stoves_2017.pdf (accessed 28/05/2018).

[141] C. L'ORANGE, M. DEFOORT, B. WILLSON, Influence of testing parameters on biomass stove performance and development of an improved testing protocol. Energy Sustainable Dev. 16 (2012) 3-12. https://doi.org/10.1016/j.esd.2011.10.008.

[142] N.V. HEEB, P. SCHMID, M. KOHLER , E. GUJER, M. ZENNEGG, D. WENGER , A. WICHSER, A. ULRICH, U. GFELLER, P. HONEGGER, K. ZEYER, L. EMMENEGGER, J.-L. PETERMANN, J. CZERWINSKI, T. MOSIMANN, M. KASPER, A. MAYER, Secondary Effects of Catalytic Diesel Particulate Filters: Conversion of PAHs versus Formation of Nitro-PAHs. Environ. Sci. Technol. 42 (2008), 3773-3779. https://doi.org/10.1021/es7026949.

APPENDIX

Measurement data

Table A1: Results of ignition test runs of respective ignition techniques (chapter 3)

	Ignition technique	**Test**	**CO**	**OGC**	**PM**	**η (indirect)**	$\boldsymbol{T_{flue\ gas}}$	**Lambda**	**Duration**	**Batch mass**	**Burn rate**
			mg/m³, STP, dry, 13 vol.-% O_2			%	°C	-	min	kg	Kg/h
Roomheater A	Variation1: Top-Down – spruce kindling	1	2090	189	80	78.9	196	3.2	65	2.93	1.9
		2	2554	321	70	78.5	192	3.3	68	2.91	2.2
		3	2231	226	169	81.8	189	2.8	58	2.92	2.2
	Variation 2: Top-Down – beech kindling	1	2856	410	84	78.7	186	3.4	66	2.89	2.1
		2	2622	345	69	79.5	186	3.3	66	2.89	2.1
		3	2719	331	147	79.2	185	3.3	69	2.92	2.1
	Variation 3: Bottom-up – spruce kindling	1	2126	325	80	82.5	211	2.4	45	2.92	3.0
		2	3257	573	111	81.4	188	2.8	56	2.91	3.0
		3	1587	291	64	86.0	205	2.0	39	2.92	3.2
	Variation 4: Bottom-up – beech kindling	1	1890	370	80	84.3	218	2.0	39	2.90	3.5
		2	2349	489	92	83.3	221	2.1	44	2.95	3.1
		3	2038	283	61	84.3	205	2.2	43	2.91	3.1
Roomheater B	Variation1: Top-Down – spruce kindling	1	541	62	50	86.0	121	7.9	46	2.90	2.0
		2	760	71	39	86.3	119	8.6	49	2.87	1.7
		3	788	107	60	87.9	126	6.5	42	2.93	2.0
	Variation 2: Top-Down – beech kindling	1	609	43	47	86.2	116	7.8	47	2.88	1.5
		2	818	36	57	86.6	121	6.9	45	2.91	1.7
		3	642	33	61	85.8	122	7.5	46	2.90	1.6
	Variation 3: Bottom-up – spruce kindling	1	756	37	38	86.4	119	10.5	49	2.89	1.7
		2	1357	216	81	87.7	120	8.8	40	2.86	2.0
		3	1319	243	91	87.1	118	5.1	43	2.87	1.8
	Variation 4: Bottom-up – beech kindling	1	2092	256	87	88.7	114	8,0	35	2.88	2.1
		2	1185	117	70	88.3	119	6.5	36	2.89	2.4
		3	1576	139	41	85.9	121	8.0	50	2.90	1.8

Table A2: Average results of combustion test cycles at different draught conditions (batch 1 – 5), PM emissions (1-5) represent time-weighted average values of batch 1, 3 and 5 (chapter 3)

Roomheater	Batch	CO	OGC	PM	η (indirect)	$T_{flue\ gas}$	Lambda	Duration	Batch mass	Burn rate
		mg/m³, STP, dry, 13 vol.-% O_2			%	°C	-	min	kg	kg/h
Roomheater A – 12 Pa	1	3560	607	111	75.5	229	2.9	53	2.89	2.7
	2	2288	329		67.1	297	3.1	62	2.41	2.3
	3	2905	284	26	70.9	281	2.9	73	2.39	1.9
	4	3410	222		70.1	265	3.1	90	2.40	1.6
	5	2720	243	32	72.6	275	2.7	71	2.44	2.0
	1 – 5	2983	317	50	71.2	271	2.9	348	12.53	2.0
Roomheater A – 24 Pa	1	3945	779	122	68.5	232	4.0	59	2.92	2.6
	2	2177	282		65.1	315	3.2	49	2.38	2.5
	3	2216	122	27	65.3	295	3.4	73	2.42	1.9
	4	2300	219		66.0	290	3.4	66	2.41	2.1
	5	2631	146	22	66.6	296	3.2	68	2.41	2.1
	1 – 5	2602	280	52	66.2	285	3.4	315	12.54	2.2
Roomheater A – 48Pa	1	1733	180	65	67.3	284	3.3	37	2.87	3.6
	2	2708	267		54.7	325	4.0	49	2.41	2.6
	3	3211	338	34	57.6	305	4.0	58	2.40	2.4
	4	1917	194		60.0	320	3.6	57	2.44	2.4
	5	1584	71	19	61.8	338	3.2	48	2.33	2.7
	1 – 5	2227	208	37	60.2	316	3.7	249	12.45	2.7
Roomheater C – 12 Pa	1	1226	114	145	78.1	228	17.2	30	2.07	2.5
	2	2018	65		76.6	297	2.8	36	1.61	2.6
	3	3516	167	118	76.4	285	2.9	42	1.58	2.2
	4	4511	280		76.7	289	2.8	37	1.60	2.5
	5	3590	178	103	77.2	287	2.8	36	1.57	2.5
	1 – 5	3060	163	124	76.9	240	5.2	182	8.43	2.5
Roomheater C – 24 Pa	1	1371	145	116	71.5	256	8.4	31	2.07	2.5
	2	941	39		74.4	315	2.9	28	1.62	3.4
	3	2356	30	187	74.3	331	2.7	33	1.57	2.8
	4	2792	102		74.5	334	2.6	28	1.60	3.3
	5	3684	77	67	74.1	329	2.6	33	1.56	2.7
	1 – 5	2211	79	118	73.7	273	3.9	152	8.42	2.9
Roomheater C – 48Pa	1	1252	146	73	65.1	272	8.1	29	2.07	2.6
	2	1210	28		72.7	352	2.9	23	1.61	4.1
	3	1802	72	118	71.9	393	2.6	24	1.58	3.8
	4	2922	151		71.7	382	2.7	25	1.60	3.7
	5	1370	18	103	74.3	384	2.5	21	1.57	4.3

Roomheater	Batch	CO	OGC	PM	η (indirect)	$T_{flue\ gas}$	Lambda	Duration	Batch mass	Burn rate
		mg/m³, STP, dry, 13 vol.-% O_2			%	°C	-	min	kg	kg/h
	1 – 5	1710	88	108	70.8	311	3.9	121	8.43	3.7
Roomheater D – 12 Pa	1	2178	317	85	73.6	201	4.2	45	2.59	3.2
	2	1500	57		64.6	289	3.8	44	2.00	2.7
	3	1367	57	63	62.0	295	4.0	46	2.02	2.6
	4	1081	49		66.4	304	3.3	38	2.06	3.2
	5	1887	72	96	63.8	316	3.4	41	2.00	2.9
	1 – 5	1597	107	81	66.0	280	3.7	214	10.67	2.9
Roomheater D – 24 Pa	1	3441	431	92	68.8	202	5.1	45	2.60	3.1
	2	1809	90		59.9	291	4.3	40	2.00	2.9
	3	1831	108	74	56.8	297	4.5	42	1.96	2.7
	4	1975	149		57.5	297	4.4	43	2.03	2.8
	5	1399	60	61	60.1	310	3.9	38	2.01	3.1
	1 – 5	2070	164	77	60.6	278	4.4	208	10.6	2.9
Roomheater D – 48Pa	1	2776	387	78	63.5	201	6.1	45	2.62	2.9
	2	2084	118		54.6	291	4.9	39	2.04	3.1
	3	1818	112	70	51.4	307	5.0	38	1.96	3.0
	4	1871	106		52.0	310	4.9	37	1.98	3.1
	5	1986	108	111	52.3	319	4.7	36	1.98	3.2
	1 – 5	2103	164	86	54.8	282	5.1	194	10.58	3.1

Table A3: DemoCat test results for different variations measured with the ceramic honeycomb catalyst (chapter 4)

Parameter	Batch	Variation I		Variation II		Variation III		Variation IV		Variation V	
		Catalyst	Dummy	Catalyst	Dummy	Catalyst	Dummy	Catalyst	Dummy	Catalyst	Dummy
CO (mg/m^3, STP, dry,13 vol.-% O_2)	1	2392	4171	952	1869	960	2116	1024	2490	966	3865
	2	1915	5075	1065	2075	752	1919	494	1573	424	2128
	3	1340	2845	918	2130	643	1721	339	1392	199	1317
	4	1182	3140	701	1477	384	1208	356	1426	261	1726
	5	457	919	485	1162	547	1736	587	2390	132	1019
OGC (mg/m^3, STP, dry,13 vol.-% O_2)	1	387	586	70	99	83	136	169	237	306	599
	2	323	601	151	222	103	173	88	144	101	149
	3	175	247	67	105	68	97	58	97	44	78
	4	163	283	26	32	43	85	57	91	63	109
	5	52	58	36	79	90	171	92	163	30	69
PM (mg/m^3, STP, dry,13 vol.-% O_2)	1	57	99	48	65	58	87	69	90	73	121
	2	50	52	60	63	37	45	22	39	61	69
	3	37	39	31	36	34	43	18	32	29	33
	4	27	35	22	28	37	42	12	25	46	56
	5	32	44	29	36	43	55	30	40	82	111

Table A4: DemoCat test results for different variations measured with the metallic honeycomb catalyst (chapter 4)

Parameter	Batch	Variation I		Variation II		Variation III		Variation IV	
		Catalyst	Dummy	Catalyst	Dummy	Catalyst	Dummy	Catalyst	Dummy
CO (mg/m^3, STP, dry,13 vol.-% O_2)	1	804	3741	213	2934	73	2177	92	3736
	2	245	2507	61	1966	19	2181	8	2313
	3	239	2530	55	1797	13	1561	7	2455
	4	370	3957	63	2084	10	1598	8	2237
	5	327	3495	44	1404	13	1976	7	1951
OGC (mg/m^3, STP, dry,13 vol.-% O_2)	1	206	394	147	316	82	170	122	338
	2	119	203	89	201	108	285	133	471
	3	115	254	66	111	55	132	64	151
	4	189	258	49	95	65	91	73	268
	5	202	392	68	128	80	255	65	155
PM (mg/m^3, STP, dry,13 vol.-% O_2)	1	78	82	59	73	48	155	80	99
	2	38	62	34	43	25	34	18	47
	3	12	19	37	48	13	15	12	16
	4	36	43	24	57	21	25	30	34
	5	16	33	35	55	19	35	17	21

Table A5: Batch results (test cycle 1, 2 and 3) of each firewood stove with integrated ceramic honeycomb catalyst (chapter 5)

		Integrated ceramic honeycomb catalyst											
		Stove A			Stove B			Stove D			Stove E		
Test cycle		3	2	1	3	2	1	3	2	1	3	2	1
Parameter	Batch	Catalyst	Dummy	Without	Catalyst	Dummy	Without	Catalyst	Dummy	Without	Catalyst	Dummy	Without
CO (mg/m^3,STP, dry,13 vol.-% O_2)	1	454	2593	2244	1018	3717	3828	577	2477	1940	660	3171	3201
	2	147	2228	1868	457	3804	2451	268	1980	2289	449	2942	2722
	3	183	1144	1374	161	3296	990	318	2753	1819	373	2426	2500
	4	248	1227	1037	214	2929	2615	383	2264	2274	403	4229	1716
	5	169	2070	916	202	1412	3662	326	2128	2334	320	1950	2351
OGC (mg/m^3, STP, dry,13 vol.-% O_2)	1	66	124	117	147	265	194	256	219	109	208	461	536
	2	28	119	99	80	255	123	117	215	261	283	374	295
	3	40	32	65	32	206	51	71	97	83	151	223	301
	4	48	50	38	59	101	69	91	143	76	150	658	144
	5	24	73	25	58	34	210	67	88	64	95	237	206
PM (mg/m^3,STP, dry,13 vol.-% O_2)	1	30	60	42	80	91	60	80	120	95	63	83	94
	2	15	40	36	43	43	34	10	24	21	36	56	57
	3	9	16	37	6	36	10	42	22	18	55	46	89
	4	10	29	18	10	16	13	25	37	48	43	57	43
	5	23	50	38	15	11	29	57	32	24	35	26	63

Table A6: Batch results (test cycle 1, 2 and 3) of each firewood stove with integrated metallic honeycomb catalyst (chapter 5)

		Integrated metallic honeycomb catalyst								
		Stove C			Stove D			Stove E		
Test cycle		3	2	1	3	2	1	3	2	1
Parameter	Batch	Catalyst	Dummy	Without	Catalyst	Dummy	Without	Catalyst	Dummy	Without
CO (mg/m^3, STP, dry,13 vol.-% O_2)	1	87	3348	1983	380	3858	1940	407	2584	3201
	2	40	1118	1046	503	2038	2289	179	2822	2722
	3	29	850	952	83	2280	1819	43	3102	2500
	4	38	1055	910	161	2840	2274	50	4174	1716
	5	80	943	1024	58	2011	2334	50	3103	2351
OGC (mg/m^3, STP, dry,13 vol.-% O_2)	1	72	532	280	156	535	109	244	296	536
	2	60	79	91	367	269	261	168	306	295
	3	41	84	74	77	93	83	79	352	301
	4	35	74	65	94	240	76	135	508	144
	5	73	53	49	32	67	64	91	331	206
PM (mg/m^3, STP, dry,13 vol.-% O_2)	1	51	158	46	130	170	95	70	54	94
	2	15	63	40	45	24	21	33	23	57
	3	5	22	20	22	23	18	41	59	89
	4	10	8	40	38	41	48	49	51	43
	5	9	15	20	31	24	24	44	60	63

Table A7: Test results of safety tests with integrated ceramic honeycomb catalyst (stove A)/ (chapter 6)

Test batch	T_{max} (upstream cat)	$\emptyset T$ (upstream cat)	T_{max} (downstream cat)	$\emptyset T$ (downstream cat)	$\Delta p_{cat.\,max}$	$\emptyset \Delta p_{cat}$	$\Delta p_{max.\,flue\,gas}$	$\emptyset \Delta p_{flue\,gas}$
	°C				Pa			
1	428	262	431	269	12	6	26	16
2	403	263	409	268	11	6	27	15
3	482	257	470	261	14	6	28	15
4	398	267	384	273	12	7	26	16
5	400	267	385	269	11	6	25	14
6	443	273	421	273	11	6	24	14
7	391	233	378	245	13	7	43	14
8	407	253	396	254	17	8	38	15
9	400	259	378	258	12	7	24	14
10	393	262	372	260	14	8	25	14
11	377	244	356	247	21	12	60	13
12	429	261	415	258	26	10	55	16
13	297	221	298	233	14	7	20	13
14	433	259	402	255	24	9	37	16
15	378	250	352	248	13	7	22	14
16	358	255	348	253	12	8	19	13
17	360	242	334	241	18	10	26	13
18	390	264	362	257	21	10	24	13
19	376	263	350	255	16	10	26	12
20	377	265	343	258	20	11	25	13

Table A8: Test results of safety tests with integrated metallic honeycomb catalyst (stove A) / (chapter 6)

Test batch	T_{max} (upstream cat)	$\emptyset T$ (upstream cat)	T_{max} (downstream cat)	$\emptyset T$ (downstream cat)	$\Delta p_{cat.\,max}$	$\emptyset \Delta p_{cat}$	$\Delta p_{max.\,flue\,gas}$	$\emptyset \Delta p_{flue\,gas}$
	°C				Pa			
1	453	270	451	278	15	7	29	17
2	464	259	473	265	17	7	27	16
3	414	247	407	253	18	8	37	17
4	480	260	536	266	23	9	31	18
5	391	243	387	254	14	8	22	15
6	454	260	465	264	16	8	26	16
7	397	263	380	268	14	8	23	14
8	367	257	365	265	14	9	21	13
9	405	269	385	269	15	9	23	14
10	382	254	370	259	18	10	21	14
11	464	287	464	288	19	10	22	14
12	371	245	389	248	16	9	21	13
13	375	271	357	272	14	10	21	12
14	347	263	353	272	25	15	21	17
15	383	272	359	267	27	14	30	20
16	364	262	332	249	30	15	26	18
17	391	250	376	250	21	10	26	14
18	387	249	366	250	18	9	29	14
19	445	257	421	253	23	11	30	14
20	453	271	417	267	20	9	26	15

Table A9: Test results of safety tests with integrated ceramic honeycomb catalyst (stove B) / (chapter 6)

Test batch	T_{max} (upstream cat)	$\emptyset T$ (upstream cat)	T_{max} (downstream cat)	$\emptyset T$ (downstream cat)	$\Delta p_{cat.\,max}$	$\emptyset \Delta p_{cat}$	$\Delta p_{max.\,flue\,gas}$	$\emptyset \Delta p_{flue\,gas}$
	°C				Pa			
1	369	222	376	230	19	5	27	17
2	387	222	385	226	10	5	27	17
3	374	227	369	230	10	5	34	16
4	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
5	441	225	412	224	20	7	59	22
6	390	228	379	227	13	6	31	17
7	424	241	398	238	12	7	38	17
8	407	228	386	224	12	7	25	17
9	400	215	373	212	27	11	66	33
10	484	234	439	227	16	7	31	18
11	399	227	371	219	14	7	31	16
12	415	232	374	225	16	8	42	19
13	441	251	401	241	17	8	51	19
14	395	227	362	218	18	7	47	16
15	416	212	382	205	22	8	69	27
16	398	226	359	214	11	6	23	16
17	430	227	383	218	12	7	25	18
18	442	229	396	218	11	6	23	16
19	424	234	386	224	12	7	27	18
20	421	218	376	208	17	8	46	23

n.d. – no data available (measurement error)

Table A10: Test results of safety tests with integrated metallic honeycomb catalyst (stove B) / (chapter 6)

Test batch	T_{max} (upstream cat)	$\emptyset T$ (upstream cat)	T_{max} (downstream cat)	$\emptyset T$ (downstream cat)	$\Delta p_{cat.\,max}$	$\emptyset \Delta p_{cat}$	$\Delta p_{max.\,flue\,gas}$	$\emptyset \Delta p_{flue\,gas}$
	°C				Pa			
1	386	221	394	227	12	5	40	16
2	422	187	414	188	45	11	≥ 100*	53
3	388	197	380	199	21	9	85	42
4	461	213	437	212	25	9	85	38
5	404	213	384	211	28	8	≥ 100*	31
6	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
7	410	226	383	223	17	7	46	19
8	367	224	351	218	11	7	23	17
9	340	189	342	186	10	6	26	14
10	412	232	386	228	14	7	23	15
11	407	232	377	225	13	7	23	15
12	367	218	349	219	12	6	21	15
13	367	225	348	219	14	7	35	16
14	384	225	365	220	12	7	28	16
15	369	230	348	224	18	8	42	19
16	367	232	346	227	13	7	28	13
17	476	215	444	208	37	10	≥ 100*	33
18	461	225	421	216	32	10	91	35
19	395	227	375	218	12	6	25	15
20	410	226	383	223	17	7	46	19

n.d. – no data available (measurement error)/ * 100 = detection limit of pressure drop

Table A11: Test results of long term durability tests of initial and final assessment tests regarding gaseous (CO, OGC) and particulate (PM) emissions with integrated ceramic and metallic honeycomb catalyst (stove A) / (chapter 6)

Combustion test		**Initial assessment**			**Final assessment**			**Final assessment – after cleaning**		
		CO	OGC	PM	CO	OGC	PM	CO	OGC	PM
Type of catalyst	Batch	(mg/m^3, dry, STP, 13 vol.-% O_2)			(mg/m^3, dry, STP, 13 vol.-% O_2)			(mg/m^3, dry, STP, 13 vol.-% O_2)		
Ceramic honeycomb catalyst	1	763	252	206	626	152	72	-	-	-
	2	403	106	57	578	220	34	-	-	-
	3	407	84	19	438	103	31	-	-	-
	4	318	53	39	339	54	26	-	-	-
	5	233	61	37	350	88	18	-	-	-
	1 – 5	412	106	71	457	120	35	-	-	-
Metallic honeycomb catalyst	1	105	130	63	623	187	54	325	173	76
	2	145	108	70	567	175	54	271	107	12
	3	176	73	41	345	78	31	173	51	43
	4	267	53	59	451	83	18	217	71	34
	5	120	68	38	548	69	15	174	33	46
	1 – 5	167	84	54	513	122	34	227	82	42

Table A12: Test results of long term durability tests of initial and final assessment tests regarding relevant parameters indicating primary combustion conditions with integrated ceramic and metallic honeycomb catalyst (stove A) / (chapter 6)

Combustion test		**Initial assessment**			**Final assessment**			**Final assessment – after cleaning**		
		O_2	Duration	T_{cat}	O_2	Duration	T_{cat}	O_2	Duration	T_{cat}
Type of catalyst	Batch	vol.-%	minutes	°C	vol.-%	minutes	°C	vol.-%	minutes	°C
Ceramic honeycomb catalyst	1	13.7	54	308	12.8	52	290	-	-	-
	2	11.1	46	401	12.7	60	370	-	-	-
	3	13.3	63	370	11.6	60	389	-	-	-
	4	12.0	55	400	11.9	60	395	-	-	-
	5	11.7	50	405	12.2	60	391	-	-	-
	1 – 5	12.4	268	376	12.2	292	369	-	-	-
Metallic honeycomb catalyst	1	12.3	43	331	8.5	53	334	12.7	48	320
	2	11.1	54	417	9.4	60	411	12.2	52	388
	3	11.7	56	404	10.6	54	396	11.8	55	406
	4	11.7	58	410	11.0	59	385	12.5	60	405
	5	11.5	50	415	11.7	64	379	11.5	55	414
	1 – 5	11.6	261	398	10.3	290	382	12.1	270	389

Table A13: Standard deviation and average of three combustion experiments according to the EN13240 and *"beReal"* test protocol (chapter 7)

Parameter	**Unit**	**acc. to EN 13240**			**acc. to *"beReal"***		
		Standard deviation	Average	CV	Standard deviation	Average	CV
O_2	vol.-%	0.2	12.4	2.1	0.3	14.3	1.5
CO_2		0.2	7.9	5.8	0.4	6.1	2.8
CO	mg/m³, STP, dry, 13 vol.-% O_2	231	2695	4.8	135	2808	8.6
OGC		4	132	12.0	16	132	3.0
PM		3	35	12.5	5	41	9.4
$T_{flue\ gas}$	°C	3	322	3.3	8	245	0.9
Thermal efficiency	%	0.3	70.2	0.6	0.4	68.7	0.4
Thermal heat output	kW	0.1	5.8	5.2	0.2	3.8	2.4

Table A14: Measurement results of selected batches for EN 13240 combustion experiments according best CO emission concentrations (chapter 7)

Parameter	**Unit**	**EN 1**		**EN 2**		**EN 3**	
Test batch	#	4	6	3	5	6	7
Start time	hh:mm:ss	10:33:45	12:05:30	08:56:51	10:28:24	12:15:34	13:03:34
End time	hh:mm:ss	11:20:21	12:50:59	09:42:15	11:15:08	13:03:34	13:49:48
Duration	s	2796	2729	2724	2804	2880	2774
CO	ppm, dry	2213	2463	2522	2527	2156	2067
THC (based on CH_4)	ppm, wet	247	269	283	203	256	212
O_2	vol.-%	12.1	12.3	12.3	12.5	12.7	12.3
CO_2	vol.-%	8,23	8.1	7.9	7.7	7.6	7.9
CO	mg/m³, STP, dry, 13 vol.-% O_2	2478	2818	2912	2980	2598	2387
OGC	mg/m³, STP, dry, 13 vol.-% O_2	129	143	152	111	143	114
Flue gas temperature (T_1)	°C	327	323	324	316	321	319
Ambient temperature ($T_{ambient}$)	°C	24	27	27	28	28	27
PM	mg/m³, STP, dry, 13 vol.-% O_2	22	51	38	37	35	27
C_{pmd}	kJ/(Km³)	1.36	1.36	1.36	1.36	1.36	1.36
C_{pmH2O}	kJ/(Km³)	1.55	1.55	1.55	1.55	1.55	1.55
$Q_{a,\,EN}$	kJ/kg	4055	4024	4101	4057	4219	4055
$q_{a,\,EN}$	%	27,2	27,0	27,6	27,3	28,4	27,3
$Q_{b,\,EN}$	kJ/kg	250	284	296	303	265	244
$q_{b,\,EN}$	%	1.68	1.91	1.99	2.04	1.78	1.64
$q_{r,\,EN}$	%	0.5	0.5	0.5	0.5	0.5	0.5
η_{EN}	%	70.6	70.5	70.0	70.2	69.4	70.6
Thermal heat output	kW	6.0	5.9	5.8	5.5	5.7	5.7

Table A15: Measurement results of *“beReal”* combustion experiments (chapter 7)

Parameter	**Unit**	**“*beReal*” 1**	**“*beReal*” 2**	**“*beReal*” 3**
Start time	hh:mm:ss	07:17:37	09:25:30	07:29:48
End time	hh:mm:ss	14:58:56	17:21:12	15:55:43
Duration	s	27679	28542	30355
CO	ppm, dry	1878	1986	1737
THC (based on CH_4)	ppm, wet	180	217	184
O_2	vol.-%, dry	14.1	14.0	15.0
CO_2	vol.-%, dry	6.0	6.2	6.0
CO	mg/m³, STP, dry, 13 vol.-% O_2	2712	2962	2749
EF_{CO}	mg/MJ	1766	1930	1790
OGC	mg/m³, STP, dry, 13 vol.-% O_2	116	148	131
EF_{OGC}	mg/MJ	76	96	85
Flue gas temperature (T_1)	°C	252	246	236
Ambient temperature ($T_{ambient}$)	°C	26	27	29
Thermal heat output	kW	3.9	3.8	3.5
PM	mg/m³, STP, dry, 13 vol.-% O_2	38	47	38
EF_{PM}	mg/MJ	24	30	25
$Q_{th,\,flue\ gas}$	kJ	34726	36023	35880
$q_{a,\,beReal}$	%	23.2	24.2	24.0
$Q_{ch,\,flue\ gas}$	kJ	3383	3815	3499
$q_{b,\,beReal}$	%	2.3	2.6	2.3
$q_{r,\,beReal}$	%	5.4	4.8	5.0
η_{beReal}	%	69.2	68.4	68.6
$q_{r,\,experimental}$	%	6.6	6.1	6.4
$V_{w,\,STP,\,calculated}$	m³, STP, wet	103.6	106.1	110.1
$V_{w,\,STP,\,measured}$	m³, STP, wet	109.4	118.4	122.9
Deviation calculated vs. measured	%	5.4	10.4	10.4

List of publications

The following list summarizes publications linked to this PhD thesis.

Journal papers

G. REICHERT, C. SCHMIDL, W. HASLINGER, M. SCHWABL, S. AIGENBAUER, M. WÖHLER, C. HOCHENAUER, Investigation of user behavior and assessment of typical operation mode for different types of firewood room heating appliances in Austria, Renew. Energy 93 (2016) 245-254. https://doi.org/10.1016/j.renene.2016.01.092.

G. REICHERT, H. HARTMANN, W. HASLINGER, H. OEHLER, R. MACK, C. SCHMIDL, C. SCHÖN, M. SCHWABL, H. STRESSLER, R. STURMLECHNER, C. HOCHENAUER, Effect of draught conditions and ignition technique on combustion performance of firewood roomheaters, Renew. Energy 105 (2017) 547-560. https://doi.org/10.1016/j.renene.2016.12.017.

G. REICHERT, C. SCHMIDL, W. HASLINGER, H. STRESSLER, R. STURMLECHNER, M. SCHWABL, N. KIENZL, M. WÖHLER, C. HOCHENAUER, Long term durability and safety aspects of oxidizing honeycomb catalysts integrated in firewood stoves, Biomass Bioenergy 105 (2017) 428-442. http://dx.doi.org/10.1016/j.biombioe.2017.07.018.

G. REICHERT, C. SCHMIDL, W. HASLINGER, H. STRESSLER, R. STURMLECHNER, M. SCHWABL, M. WÖHLER, C. HOCHENAUER, Catalytic efficiency of oxidizing honeycomb catalysts integrated in firewood stoves evaluated by a novel measuring methodology under real-life operating conditions, Renew. Energy 117 (2018) 300-313. https://doi.org/10.1016/j.renene.2017.10.065.

G. REICHERT, C. SCHMIDL, W. HASLINGER, H. STRESSLER, R. STURMLECHNER, M. SCHWABL, C. HOCHENAUER, Novel method evaluating real-life performance of firewood roomheaters in Europe, Energy Fuels 32, (2) (2018) 1874-1883. https://doi.org/10.1021/acs.energyfuels.7b03673.

G. REICHERT, C. SCHMIDL, W. HASLINGER, H. STRESSLER, R. STURMLECHNER, M. SCHWABL, M. WÖHLER, C. HOCHENAUER, Impact of oxidizing honeycomb catalysts integrated in firewood stoves on emissions under real-life operating conditions, Fuel Process. Technol. 177 (2018) 109-118. https://doi.org/10.1016/j.fuproc.2018.04.016.

Conference papers and presentations

G. REICHERT, C. SCHMIDL, M. SCHWABL, H. STRESSLER, R. STURMLECHNER, W. HASLINGER, C. HOCHENAUER, Integration of oxidative honeycomb catalysts in firewood stoves – Performance analysis, safety aspects and long term durability, 23rd European Biomass Conference and Exhibition, 1. – 4. June, 2015, Vienna, Austria, 452-456. https://doi.org/10.5071/23rdEUBCE2015-2AO.2.3.

G. REICHERT, H. HARTMANN, W. HASLINGER, H. ÖHLER, S. PELZ, C. SCHMIDL, M. SCHWABL, H. STRESSLER, R. STURMLECHNER, M. WÖHLER, C. HOCHENAUER, "*beReal*" – Development of a new test method for firewood roomheaters reflecting real life operation, 24th European Biomass Conference and Exhibition, 6. – 9 June, 2016, Amsterdam, The Netherlands 382-387. https://doi.org/10.5071/24thEUBCE2016-2AO.2.1.

G. REICHERT, H. STRESSLER H., SCHMIDL C., SCHWABL M., R. STURMLECHNER, W. HASLINGER, C. HOCHENAUER, Emission reduction of firewood roomheaters by optimization of operating conditions and catalyst integration, 24th European Biomass Conference and Exhibition, 6. – 9 June, 2016, Amsterdam, The Netherlands 654-659. https://doi.org/10.5071/24thEUBCE2016-2BV.1.14.

G. REICHERT, H. HARTMANN, W. HASLINGER, H. ÖHLER, R. MACK, C. SCHMIDL, C. SCHÖN, M. SCHWABL, H. STRESSLER, R. STURMLECHNER, C. HOCHENAUER, Real life gap of emissions and efficiency for firewood roomheaters, WSEDnext Young Researcher´s Conference, 1. March, 2018, Wels, Austria.

Presentations

G. REICHERT, H. HARTMANN, W. HASLINGER, H. ÖHLER, R. MACK, C. SCHMIDL, C. SCHÖN, M. SCHWABL, H. STRESSLER, R. STURMLECHNER R., HOCHENAUER C.: Batch-wise wood combustion in firewood roomheaters – impact of ignition mode and draught conditions, WSEDnext Young Researcher´s Conference, 25. February, 2016, Wels, Austria.

G. REICHERT, C. SCHMIDL, H. STRESSLER, R. STURMLECHNER, C. HOCHENAUER, H. HARTMANN, C. SCHÖN, R. MACK, H. OEHLER, The „beReal" Project – Scientific Highlights, IEA Bioenergy Task 32 workshop: Practical test methods for small-scale furnaces, 5th Central European Biomass Conference, 18-20 January, 2017, Graz, Austria. Online available, http://task32.ieabioenergy.com/wp-content/uploads/2017/03/14.20_The__beReal__Project_-_Scientific_Highlights.pdf.

G. REICHERT, W. HASLINGER, J.-M. KIRCHHOF, C. SCHMIDL, I. SEDLMAYER, M. SCHWABL, H. STRESSLER, R. STURMLECHNER, M. WÖHLER, C. HOCHENAUER, Honeycomb catalysts integrated in firewood stoves – potentials and limitations, 5th Central European Biomass Conference, 18-20 January, 2017, Graz, Austria. Online available, http://www.cebc.at/service/publikationen/5-mitteleuropaeische-biomassekonferenz/ps-5/.

Curriculum vitae

PERSONAL INFORMATION

Name:	Gabriel Reichert
Address:	Arbeitergasse 14, 3250 Wieselburg, Austria
Mail:	Gabriel.Reichert@bioenergy2020.eu
Sex:	male
Date of birth:	06.02.1985
Nationality:	Germany

WORK EXPERIENCE

Since 04/2015	RESEARCHER BIOENERGY 2020+ GmbH, Location Wieselburg, Gewerbepark Haag 3, 3250 Wieselburg-Land, Austria, www.bioenergy2020.eu Activities: Research and development in the field of small-scale biomass combustion systems
04/2012 – 03/2015	JUNIOR RESEARCHER BIOENERGY 2020+ GmbH, Location Wieselburg, Gewerbepark Haag 3, 3250 Wieselburg-Land, Austria, www.bioenergy2020.eu Activities: Research and development in the field of small-scale biomass combustion systems
07/2009 – 03/2012	RESEARCH ASSOCIATE University of Applied Forest Sciences Rottenburg, Schadenweilerhof, 72108 Rottenburg, Germany, www.hs-rottenburg.net Activities: Research and development in the field of biomass resources and thermal biomass utilization (combustion & gasification)

EDUCATION

Since 04/2015	Graz University of Technology – Institute of Thermal Engineering Degree (exp.): Dr. techn.
10/2012 – 10/2014	University of Applied Sciences Wiener Neustadt for Business and Engineering Degree: M.Sc. in Engineering
10/2004 – 03/2009	University of Applied Forest Sciences Rottenburg Degree: Dipl.-Ing. (FH) in Forestry
09/1995 – 06/2004	Robert-Bosch-Gymnasium Langenau Degree: University-entrance diploma