

HEALTH RELEVANCE OF PARTICLES FROM WOOD COMBUSTION IN COMPARISON TO DIESEL SOOT

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ABSTRACT: The aim of the present investigation is to assess the health relevance of different combustion particles, i.e.: Diesel soot, particulate emissions from a quasi complete combustion of wood in an automatic wood boiler, consisting mainly of inorganic matter such as salts, and particulate matter from an incomplete combustion of wood in a badly operated wood stove. In addition, the range of variability of particle emissions and size distributions from different types of residential wood combustion and of different operation modes of wood stoves was determined. Particles and condensates were sampled in the flue gas and the total particle mass and the particle size distributions were analysed. The samples were used for biological tests on cell toxicity and on chromosome defects with lung cells from the chinese hamster. In addition, polycyclic aromatic hydrocarbons (PAH) were analysed. The results show, that Diesel soot exhibits a medium level of toxicity and chromosome defects, while particles from the automatic wood combustion exhibits app. 5 times lower toxicity. However, soot from the badly operated wood stove exhibits app. 15 times higher toxicity and chromosome defects than Diesel soot and app. 20 times higher levels of PAH. The highest toxicities are found for condensable matter from the wood stove.

Keywords: Aerosols, health, combustion.

1 INTRODUCTION

Particulate matter smaller than 10 microns (PM10) is regarded as one of the most relevant parameters in air pollution [1, 2]. Limit values for both, particle emissions and immissions are indicated as mass concentrations and hence do not distinguish between toxic and non toxic substances. Earlier investigations on the formation mechanisms of particles from biomass combustion revealed that depending on the combustion conditions, mainly inorganic compounds are emitted as particulate matter from a quasi-complete combustion of wood, while soot and organic matter may be emitted under unfavourable combustion conditions [3]. The present paper summarizes the results of two investigations aiming at two targets:

First, to determine the influence of stove operation on mass concentrations, physical and chemical properties, and health relevance of particles from wood stoves [4], and second to compare the health relevance of different combustion particles [5].

1.1 Influence of operation on particles from wood stoves

Particle emissions from wood stoves strongly depend on furnace design, operation conditions, and type and properties of the wood fuel. The aim of this investigation is to determine the range of variability of particle emissions and size distributions. The ability to deal with wood of different quality, which is mainly determined by the water content, is compared for the following combustion devices (Fig. 1):

1. A metal stove with small combustion chamber and low mass of ceramics lining,
2. A modern wood stove with large combustion chamber and heavier ceramic lining which fulfils the requirements of the Swiss quality label,
3. A newly designed wood stove with two-stage combustion by gasification and consecutive gas oxidation in a separated combustion chamber with secondary air

4. A modern pellet stove operated with wood pellets and straw pellets.

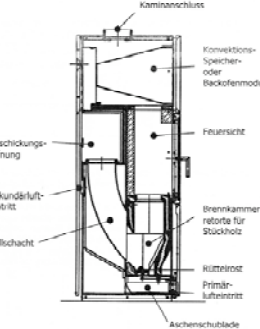
The experimental setup and the results are described in detail in [4].



Stove 1



Stove 2



Stove 3

Figure 1: Investigated wood stoves. Top: Conventional stoves. Bottom: Two-stage combustion stove (prototype).

1.2 Comparison of different combustion particles

Earlier investigations showed, that well designed and operated automatic wood combustion plants achieve a high combustion quality thus resulting in low emissions of unburned hydrocarbons and soot [3]. However, automatic wood boilers exhibit fairly high mass concentration of particle emissions in the raw gas, i.e., typically in the order of magnitude of 100 mg/m^3 at 13 Vol.-% O_2 for natural wood chips and up to several 100 mg/m^3 for other fuels such as bark. However, these particles consist mainly of inorganic matter such as salts, while particles from wood stoves operated under bad conditions consist mainly of soot and organic substances. Since Diesel engines and biomass combustion are the main sources of primary combustion particles in the environment, and since Diesel soot has been identified as being carcinogenic, the following three basic types of combustion particles were investigated to determine and compare their health relevance:

1. Diesel soot from a modern type Diesel engine (without Diesel particle filter),
2. Particulate emissions from a quasi complete combustion of wood in an automatic wood boiler, consisting mainly of inorganic matter such as salts (in the raw gas, hence without application of any flue gas after treatment),
3. Particulate matter from an incomplete combustion of wood in a badly operated wood stove, consisting mainly of soot and additionally containing condensable organic matter.

The experimental setup and the results are described in detail in [5], and first results from a Diesel engine and from an automatic wood boiler, yet without data from an incomplete wood combustion, have been presented in [6].

2 METHOD

Particle emissions were monitored according to the German norms (VDI) by sampling from the hot flue gas on a heated filter probe. Furthermore, the contribution of condensable organic matter in the flue gas sampled by quenching according to EPA was assessed under different combustion conditions and compared to the solid particulate matter as detected by VDI norms. In addition to these measurements, the time dependence and size distribution of the particle emissions was monitored in the size range from 20 nm to $10 \mu\text{m}$ by a combination of a Scanning Mobility Particle Sizer (SMPS) and an Optical Particle Counter (OPC). Furthermore, chemical analyses on organic carbon and polycyclic aromatic hydrocarbons (PAH) of solid particles and condensable matter were performed.

The composition of the particles is an important parameter for the interaction with the human body and resulting health effects. The toxicity of the particles was tested by a standard test method with lung cells of the Chinese hamster. This cell line allows also a standard detection of chromosome defects, which are an indicator for the carcinogenic potential of the applied particles. The health relevance was assessed by the cell toxicity and the chromosome aberration. The method for the biological cell tests have been presented earlier [6]. The present publication presents additional results on particles from incomplete wood combustion and on the influence of the

stove operation. Diesel soot was sampled during stationary operation of a modern Diesel passenger car on a test-bench as described in [5]. Similar sampling was applied for an automatic wood boiler and for wood stoves.

3 RESULTS ON STOVE OPERATION [4]

The resulting particle emissions from three different wood stoves under different operation conditions varied between 20 mg/m^3 at 13% O_2 and up to 5000 mg/m^3 for a stove operated under very bad conditions as shown in Fig. 2. The particle spectra for low emissions are comparable to modern oil burners, while bad operation creates an increased fraction of particles near 0.5 to $1 \mu\text{m}$. The comparison of different operation and stoves revealed:

1. Conventional updraft stoves (a small metal stove and a modern stove with ceramic lining) achieved particle emissions of $20 - 50 \text{ mg/m}^3$ (at 13 vol.-% O_2) including start-up emissions, when operated ideally. An ideal operation is achieved by ignition of very small streaks of dry wood which are put on top of small logs of dry wood. When a glow bed is available, two small logs of dry wood ($2 \times 750 \text{ g}$ at $w = 12\%$) are added. Even lower particle emissions were achieved in a prototype of an advanced two stage combustion stove operated with dry wood resulting in 10 mg/m^3 .
2. If the conventional stoves were operated under typical heating conditions with more than 50% filled fuel chamber using wood logs of 1.5 kg, the emissions increased by a factor of 5 to 10 due to incomplete combustion presumably as a consequence of insufficient mixing and too short residence time in the hot zone. On the other side, the two-stage combustion stove can be operated with fully filled fuel chamber. Here, the instantaneous heat output and the residence time in the combustion chamber are not affected by the level of logs used in the fuel chamber. Hence the two-stage combustion achieves a relevant advantage under typical heating conditions.
3. If the conventional stoves are operated under very bad conditions, i.e., by closing the air inlet after start-up to prolong the combustion time, the emissions increase by another factor of 10. Such an operation is not possible in the two-stage combustion stove.
4. The pellet stove achieved particle emissions below 30 mg/m^3 (at 13 vol.-% O_2) and NO_x emissions below 200 mg/m^3 when operated with wood pellets. The use of straw pellets resulted in particle emissions of more than 150 mg/m^3 and NO_x emissions of 800 mg/m^3 . In addition, slagging occurred which disabled a continuous operation. While technical measures to improve the combustion behaviour are possible, the increase of particle and NO_x is mainly caused by the increased content of ash and nitrogen in the fuel, as has been shown in earlier investigations with different herbaceous biofuels [7].

4 RESULTS ON HEALTH RELEVANCE [5]

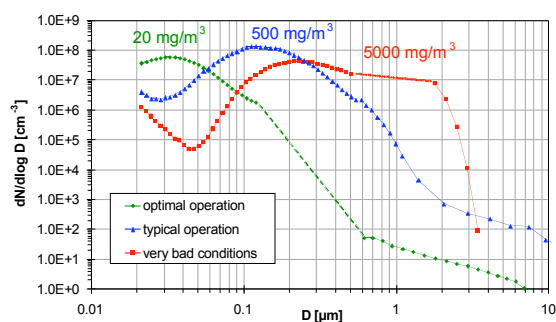


Figure 2: Number size distribution measured by SMPS and OPC of particles from wood stoves under different operation conditions, i.e.: 1. Ideal or optimal operation (20 mg/m^3 at 13 Vol.-% O_2), 2. typical operation (500 mg/m^3), and 3. very bad operation ($5'000 \text{ mg/m}^3$).

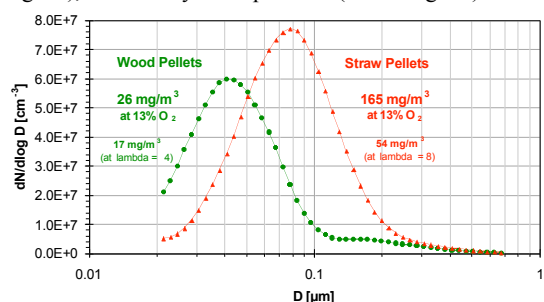


Figure 3: Number size distribution from a pellet stove operated with wood pellets (left curve) resulting in particle emissions of 26 mg/m^3 at 13 Vol.-% O_2 , and in operation with straw pellets, resulting in 165 mg/m^3 (right curve).

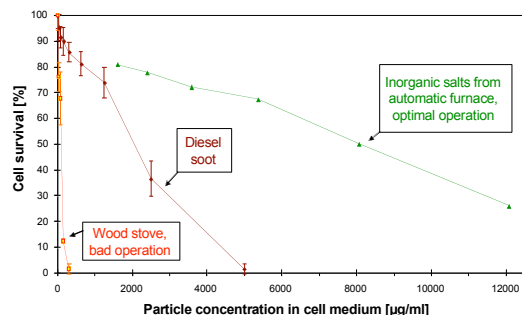


Figure 4: Cell survival as a function of particle concentration for Diesel soot, mainly inorganic particles from automatic wood combustion, and wood particles from incomplete combustion in a badly operated stove.

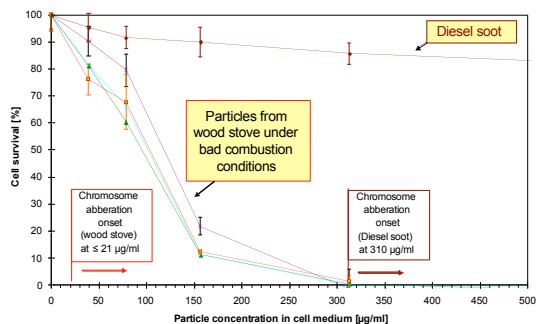


Figure 5: Cell survival as a function of particle concentration for Diesel soot and wood particles from incomplete wood combustion (data as in fig. 2 but in magnified scale). In addition, the level of chromosome aberration onset is indicated.

A comparison of the three different particle types, i.e., Diesel soot, mainly inorganic particles from a well operated automatic wood boiler, and particles from a badly operated wood stove, reveals the following ranking (Fig. 4 and 5):

The particles from automatic wood furnaces exhibit the lowest toxicity which corresponds to the effects found with pure potassium chloride. The chromosome defects of these particles were below the detection limit.

Diesel soot exhibits a medium toxicity (i.e. 5 times higher than inorganic particles in the selected tests) and detectable chromosome defects.

Particles from incomplete combustion of wood in a badly operated stove result in the highest toxicity and chromosome defects, i.e., app. 15 times higher than for Diesel soot. Furthermore, the chemical analyses reveal substantially higher concentrations of PAH, which are assumed to contribute to the increased toxicity (table 1). The high carcinogenic potential of wood particles from incomplete combustion is in line with results from investigations which reveal comparable lung cancer pathogenesis from smoking and cooking with wood [8].

Table 1: PAH concentrations in particles from badly operated wood stove and in Diesel soot.

mg/kg	Particles from wood stove	Diesel soot
Naphthalene	13	42
Acenaphthylene	129	7.1
Acenaphthene	17	< 3
Fluorene	173	< 3
Phenanthrene	231	3.7
Anthracene	65	< 3
Fluoranthene	154	< 3
Pyrene	170	< 3
Chrysene	54	< 3
Benzo(a)anthracen e	44	< 3
Benzo(b)fluoranthene	30	< 3
Benzo(k)fluoranthene	11	< 3
Benzo(a)pyrene	25	< 3
Indeno(1,2,3-cd)pyrene	9	< 3
Dibenzo(a,h)anthracene	< 8	< 3
Benzo(g,h,i)perylene	< 8	< 3
Total PAH	1 120	53

4 CONCLUSIONS

The results on wood stoves reveal a huge influence of the type of operation on the particle mass concentration and the particle toxicity for conventional stoves. If operated ideally, even simple wood stoves can achieve low level of particle emissions, i.e., as low as $20 - 50 \text{ mg/m}^3$ (at 13 vol.-% O_2). However, if operated under typical heating conditions, i.e., by utilisation of typical log sizes and with use of more than 50% of the available fuel storage volume, the emissions can increase by a factor of 5 to 10 thus resulting a relevant contribution to the ambient air pollution. If operated badly by closing the air inlet after the start-up, the emissions can increase by

another factor of 10 thus resulting in excessive air pollution with particle emissions of up to more than 5000 mg/m³. The application of a consistent two-stage combustion principle, which was demonstrated in the investigated prototype but not state-of-the art for wood stoves available on the market nowadays, enables particle emissions as low as 10 – 20 mg/m³ under good operation conditions and still less than 100 mg/m³ under bad operation conditions. Due to the design principle, a very bad operation during a long period of time is almost impossible as e.g. an operation with wet wood leads to an extinction of the combustion.

The results on the health relevance of different combustion particles reveals, that particles from incomplete combustion of wood exhibits a far higher toxicity and carcinogenic potential than Diesel soot, i.e., by app. an order of magnitude. On the other hand, particles from quasi-complete combustion of wood in an automatic wood boiler exhibit app. 5 times lower toxicity than Diesel soot which corresponds to the effects found for potassium chloride.

Hence the following conclusions from the two reported investigations are proposed:

- Low emissions from manually operated wood stoves are possible, in conventional stoves if operated ideally, in two stage stoves (not available on the market today) also under conditions as comparable to real heating applications.
- However, real life emissions of conventional wood stoves are presumably several times higher than estimated from emission factors as usually measured and reported from type tests.
- Since nowadays type tests do not exclude an operation with ideal conditions, advantages of improved stove design is not rewarded in nowadays certifications. Hence improved type tests should be developed where emissions under practical conditions are determined.
- Bad operation of wood stove does not only lead to far increased particle emissions, but also to considerably increased toxicity and carcinogenic potential of the emitted particles. Since nowadays emission limits and immission limits do not consider the toxicity of the particles, but only the mass, the health relevance of particle emissions of badly operated wood combustion devices and presumably of open fires might be highly underestimated.
- Well designed and operated pellet stoves achieve a high combustion quality at low particle emissions, i.e., below 30 mg/m³ (at 13 vol.-% O₂) for the investigated pellet stove and often below 15 mg/m³ in modern pellet boilers. In addition, pellet combustion can be automatically controlled thus avoiding very bad operation in practice, which is regarded as the main advantage of pellets in comparison to log wood. However, the use of other raw materials than natural wood with low bark content should be restricted to large combustion plants equipped with flue gas cleaning, since the combustion of straw pellets, bark pellets and similar pellets lead to significantly increased emissions of particles and NO_x caused by the increased content of ash and nitrogen in the fuel.
- Automatic wood combustion also significantly contributes to the total particulate matter in the ambient air and hence measures for particle removal are

needed as well. However, these particles are less toxic than Diesel soot. Therefore, measures to reduce particle emissions from incomplete wood combustion are of higher priority.

6 LITERATURE

- [1] Dockery D., C. Pope, X. Xu, J. Spengler, J. Ware, M. Fay, B. Ferris, F. Speizer. (1993): An association between air pollution and mortality in six U.S. Cities. *The New England J. of Medicine*, Vol. 329, pp. 1753-1759.
- [2] Donaldson K., Brown D., Clouter A., Duffin R., MacNee W., Renwick L., Tran L., Stone V. (2002): The pulmonary toxicology of ultrafine particles. *Journal of Aerosol Medicine*, 15(2), pp. 213-20
- [3] Oser, M.; Nussbaumer, Th.; Müller, P.; Mohr, M.; Figi, R.: Mechanisms of particle formation in biomass combustion. *Second World Biomass Conference*, 10-14 May 2004, Rome, ETA Florence and WIP Munich, ISBN 88-89407-04-2, 1246–1249
- [4] Klippel, N.; Nussbaumer, T.: Einfluss der Betriebsweise auf die Partikelemissionen von Holzöfen, Bundesamt für Energie und Bundesamt für Umwelt, Schlussbericht, Zürich 2007, ISBN 3-908705-12-6, Download from www.verenum.ch
- [5] Klippel, N.; Nussbaumer, T.: Wirkung von Verbrennungspartikeln – Vergleich der Umweltrelevanz von Holzfeuerungen und Dieselmotoren, Bundesamt für Energie und Bundesamt für Umwelt, Schlussbericht, Zürich 2007, ISBN 3-908705-16-9, Download from www.verenum.ch
- [6] Klippel, N.; Nussbaumer, T.: Health relevance of combustion particles from automatic wood furnaces and Diesel engines indicated by cytotoxicity, 14th European Biomass Conference, Paris, 17-21 October 2005
- [7] Kaufmann, H.; Nussbaumer, Th.; Baxter, L.; Yang, N.: Deposit formation on a single cylinder during combustion of herbaceous biomass, *Fuel* 79 (2000), 141 – 151
- [8] Delgado, J.; Martinez, L.; Sanchez, T.; Ramirez, A.; Iturria, C.; Gonzalez-Avila, G.: Lung Cancer Pathogenesis Associated With Wood Smoke Exposure, *Chest* (2005) 128, 124–131

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