

# Particulate Emissions of a Modern Wood Stove

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## Introduction

In response to global warming, the use of biomass fuels is increasing as climate neutral fuels are sought. The use of wood for domestic heating has been heavily driven in the EU and also in Germany over the last years.

In 2008 ca. 14 Mio small stoves and boilers for solid fuel are used in Germany, and most of them combust wood. During the last years the number of wood stoves and also the consumption of wood for domestic heating in Germany increased. This causes continuous increase of the emission of fine particulate matter from domestic heating during the last couple of years, reaching an emission of 30.9 kt in 2012 for  $PM_{10}$  and 29.2 kt for  $PM_{2.5}$  (UBA, 2014). In 2012 the German total PAH emission, as sum of BaP, Benzo(b)fluoranthene, Benzo(k)fluoranthene, Indeno(1,2,3-cd)pyrene, was 191 t (UBA, 2014). More than 80% of the PAH emission was caused by domestic heating mostly with wood or wood products as fuel (UBA, 2012).

The aim of the present study was to investigate the particulate emissions of a modern wood stove to ascertain as such a stove can contribute to diminish the problem of air pollution mentioned above under realistic conditions of use. The main focus of the work was in the size segregated measurement of PM and PAH emissions.

## Experiment

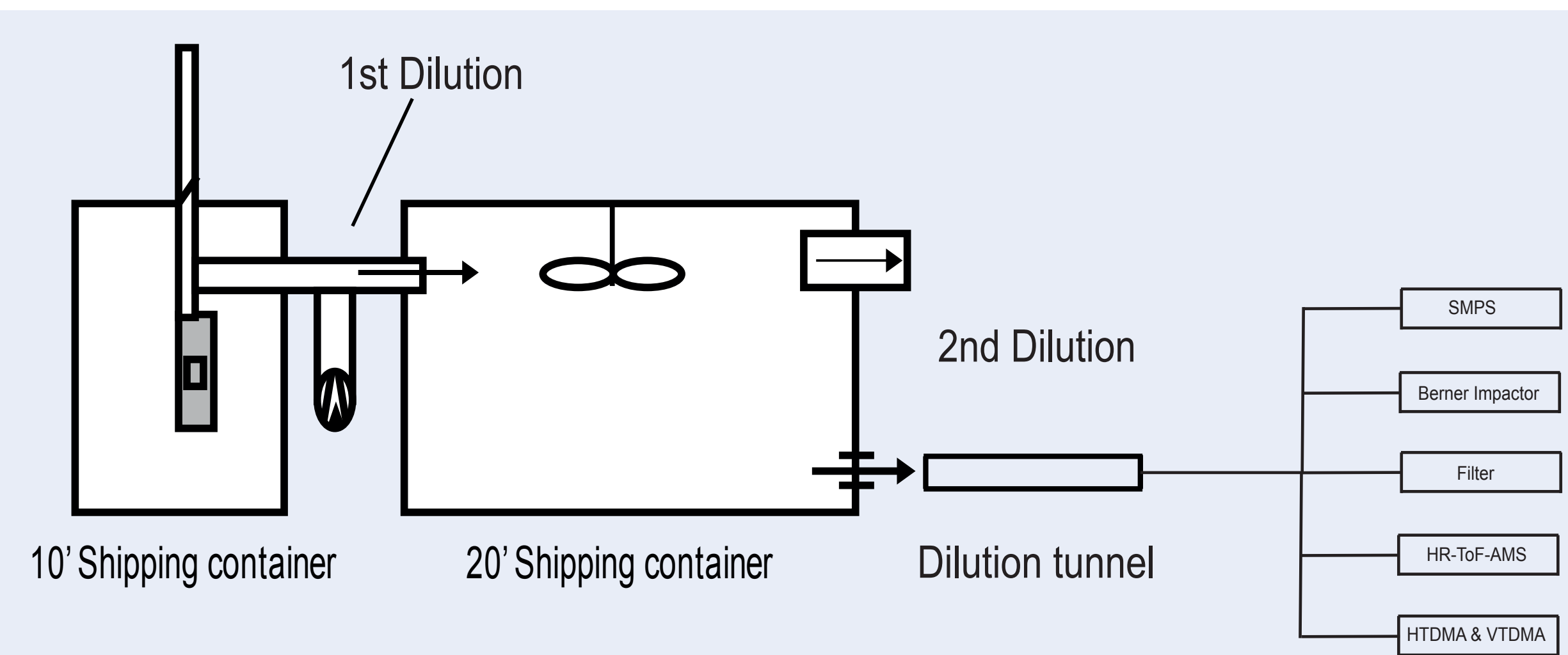


Fig. 1. Scheme of the Leipzig Biomass Burning Facility (LBFF).

Wood Combustion experiments were carried out in the Leipzig Biomass Burning Facility (LBFF) at the Leibniz Institute for Tropospheric Research (TROPOS) in Leipzig, Germany (Fig.1). A series of experiments were performed with a modern log wood oven based on down draft technology (Fig.2).

At the beginning of the chimney  $CO$ ,  $CO_2$  and the exhaust temperature were measured. In the 20' shipping container  $CO$  again was measured to calculate the first dilution ratio. The second dilution ratio was calculated by measuring the flow of the smoke into the dilution manifold before the experiment, whereas the dilution flow was set by a flow controller. Beech as well as spruce logs were used as fuel. The first two wood logs were ignited using charcoal lighter. Every 20 minutes two wood logs were added on top of the glow bed, which results in an addition of ~ 650 g for spruce and ~850 g for beech, respectively. After 30 min from the start secondary combustion air was switched on and the wood smoke was transferred into the mixing container and when the  $CO$  concentration in the mixing container reached a constant value, the sampling at the end of the second dilution manifold was started. Sampling time was 90 - 120 min resulting in a sampling volume of 2.25 - 3.0 m<sup>3</sup>.



Fig. 2. Modern wood stove with two-stage combustion.

The particle number size distributions between 9 nm and 900 nm are measured by a scanning mobility particle sizer (SMPS) with a time resolution of 10 mins. An overview about the experiments performed and the basic experimental data is given in Table 1.

Table 1. Summary of basic experimental data

Date	Fuel	T		CO <sub>2</sub>	CO	Dilution Ratio
		°C	%			
2010-12-06	Spruce	260	7.56	248	1:216	
2010-12-07	Spruce	260	7.16	147	1:345	
2010-12-08	Spruce	267	5.11	199	1:451	
2012-03-20	Beech	398	12.3	809	1:2690	
2012-03-21	Beech	399	13.5	733	1:960	
2012-03-27	Beech	401	12.1	721	1:1190	

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PM samples were taken using a 10-stage Berner-type impactor with 50% cut offs at 0.037, 0.073, 0.14, 0.27, 0.48, 0.93, 2, 4, 8 and 16  $\mu m$  aerodynamic diameter. Mass determination was done by weighing the clean and particle loaded impactor foils on a microbalance UMT-2 (Mettler-Toledo, Switzerland). For ion analysis, about 25 % of the impactor foils were extracted in 1.5 ml MilliQ-water (>18 M  $\Omega$  cm, 15 min shaker, 15 min ultrasonic bath, 15 min shaker). Ion analysis was performed for cations  $Na^+$ ,  $NH_4^+$ ,  $K^+$ ,  $Mg^{2+}$ , and  $Ca^{2+}$  and anions  $Cl^-$ ,  $Br^-$ ,  $NO_3^-$ ,  $SO_4^{2-}$  and  $C_2O_4^{2-}$  using a standard ion-chromatography technique (ICS3000, DIONEX, USA). For the quantification and separation of carbonaceous PM into organic (OC) and elemental carbon (EC) a two-step thermographic method was applied using a commercial carbon analyzer (C-mat 5500, Ströhlein, Germany). A Curie Point Pyrolysis-GC/MS (CPP-GC/MS) technique was used to quantify PAHs and n-alkanes.

## Results

### Particulate Matter Emissions

The bulk PM emission factors (mg of mass emitted per 1 kg of dry wood) from this study together with results for wood stove emission factors available in the literature are summarized in Table 2. The  $PM_{10}$  emission factor of 0.12 for spruce observed in this study is at least a factor of 3 lower than the lowest emission factor for wood stove reported in literature (Kistler et al., 2012). The observed  $PM_{10}$  emission factor of ~ 1.3 for beech as fuel is comparable to the lower range of the values reported in literature. Nevertheless, the PM emission of the modern wood stove with beech as fuel is significantly higher than for spruce. Although the same experimental setup and conditions were used, strong differences in the burning conditions could be observed (Table 1). First of all the exhaust temperature was much higher than in the experiments with spruce used as fuel, which results in a higher emission of inorganic species (ash) cf. Figure 3 than at lower temperature.

Table 2. Wood stove emission factors for fine particulate matter

Date	Fuel	$PM_{10}$	$PM_{2.5}$	$PM_{10}$	References
		g/kg	g/kg	g/kg	
2010-12-06	Spruce	0.113 <sup>a</sup>	0.115 <sup>a</sup>	0.116 <sup>a</sup>	Concalves et al., 2010
2010-12-07	Spruce	0.102 <sup>a</sup>	0.105 <sup>a</sup>	0.112 <sup>a</sup>	
2010-12-08	Spruce	0.122 <sup>a</sup>	0.124 <sup>a</sup>	0.125 <sup>a</sup>	
2012-03-20	Beech	1.81 <sup>b</sup>	1.85 <sup>b</sup>	1.91 <sup>b</sup>	Concalves et al., 2011
2012-03-21	Beech	1.0 <sup>a</sup>	1.03 <sup>a</sup>	1.07 <sup>a</sup>	
2012-03-27	Beech	0.92 <sup>a</sup>	0.94 <sup>a</sup>	0.94 <sup>a</sup>	
Portuguese wood species		1.66-16.0 <sup>b,d</sup>			Fine et al., 2004
American tree species		0.88-3.4			
Oak				3.4 <sup>c</sup>	Kinsey et al., 2009
Douglas fir				2.4 <sup>c</sup>	
Various wood species				0.36-4 <sup>c</sup>	Kistler et al., 2012
Oak				2.8-41 <sup>c</sup>	
Oak		3.6-7.2 <sup>a</sup>			McDonnell et al., 2000
Mixed hardwoods		2.3-6.1 <sup>a</sup>			
Various wood species				2.6 <sup>d,e</sup>	Ozgen et al., 2014
Birch, Spruce, Pine				0.68-3.1 <sup>d,e</sup>	
Spruce				1.3-1.6 <sup>f</sup>	Pettersson et al., 2011
Beech				1.1-1.3 <sup>f</sup>	
Mainly birch		0.9			Tissari et al., 2007
Acacia nilotica		0.9-2.8 <sup>g</sup>			

<sup>a</sup> Emission factor calculated as g of PM/dry kg of fuel. <sup>b</sup> Initial temperature in the combustion chamber was 307±67°C (hot start). <sup>c</sup>  $PM_{10}$  calculated using a conversion factor of 18 MJ/kg. <sup>d</sup> Calculated using an average factor of 10 for the conversion of mg m<sup>-3</sup>STP, 13%CO<sub>2</sub> into mg/kg. <sup>e</sup> Data for advanced stove.

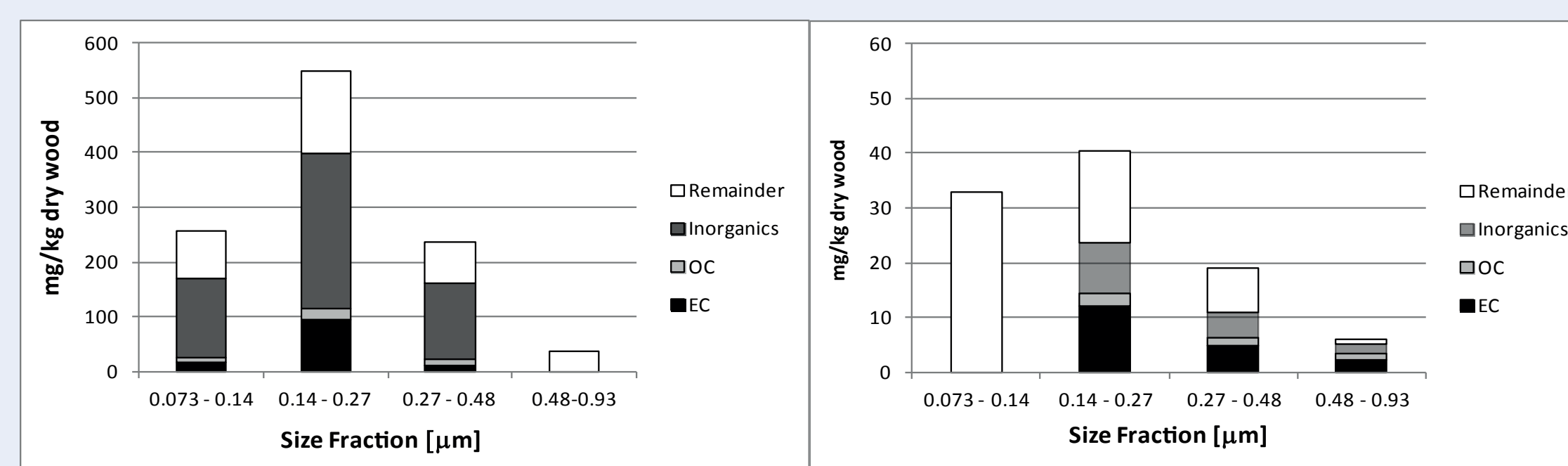


Fig. 3. Size-resolved EC, OC and soluble ion emission factors for spruce (left part) and beech (right part)

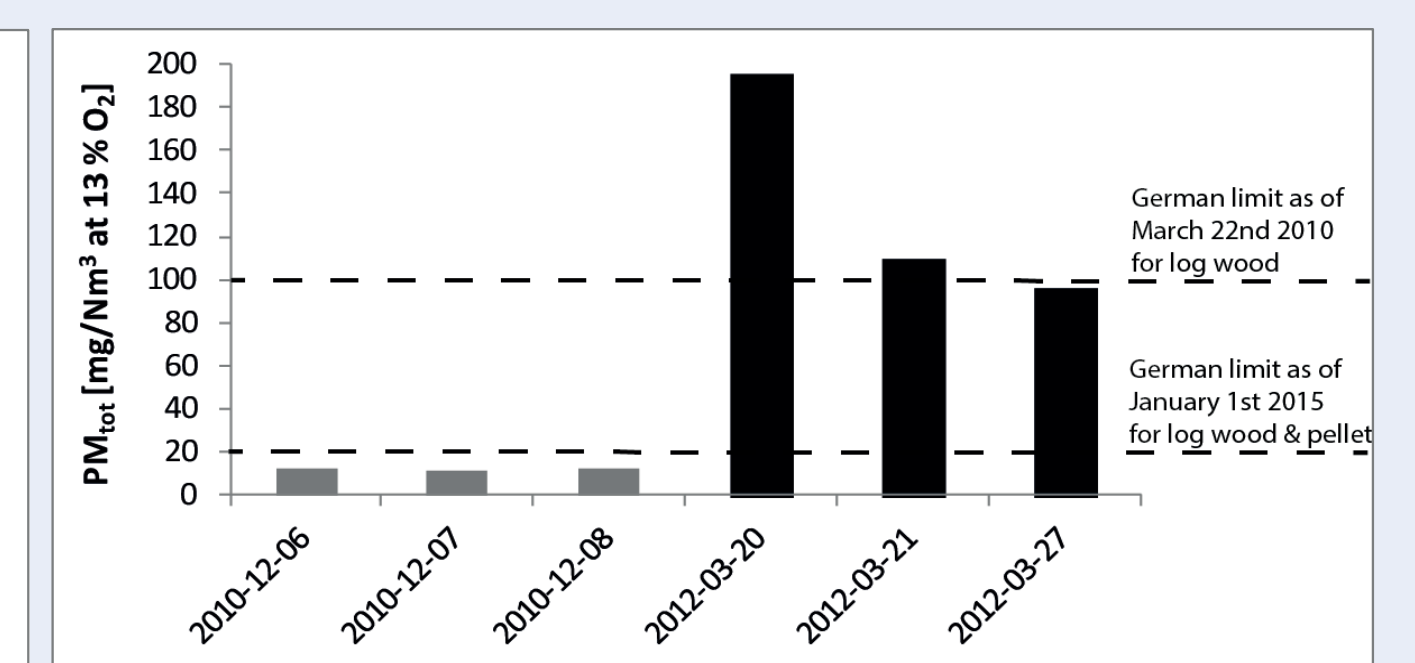


Fig. 4. Total PM emission factors from spruce combustion (left-hand side) and beech (right-hand side) together with German emission limit values.

Additionally, the higher  $CO_2$  concentration in the exhaust of the experiments with beech used as fuel indicates an insufficient amount of combustion air supply. Together with the high temperature this leads to too fast fuel pyrolysis and a high emission of incomplete combustion products. All these factors result in a higher PM emission for beech combustion observed in our experiments. Figure 4 present the PM emission factors together with the German emission limit values (1.BImSchV, 2010). The emission values were calculated in mg per dry standard cubic meter with 13%  $CO_2$ . From the experiments with spruce used as fuel can be seen that the emission factors are clearly below limit values. But it is difficult to compare results which were obtained to verify the emission factors on a test station with values obtained under real conditions using a natural draft chimney.

### Size-Segregated PM and Size Distribution

Figure 5 shows the size-segregated PM emission factors in mg/kg of dry wood for spruce and beech. The PM emission factors for spruce and beech have a similar mass size distribution, peaking at stage 4 (0.14-0.27  $\mu m$ ) of the impactor. These results are comparable to measurements published in the literature. Figure 5 shows clearly that there very low emissions of particles larger than  $PM_{10}$ . The average particle size distributions for spruce and wood combustion experiments are shown in Figure 6. For both wood types a unimodal size distribution can be observed, which is in accordance with observations in the literature for advanced burning period. The particle size was especially small in the experiments with spruce used as fuel. The particle mean (PMD) diameter varied from 54 to 64 nm for spruce combustion and 99 to 108 nm for beech combustion.

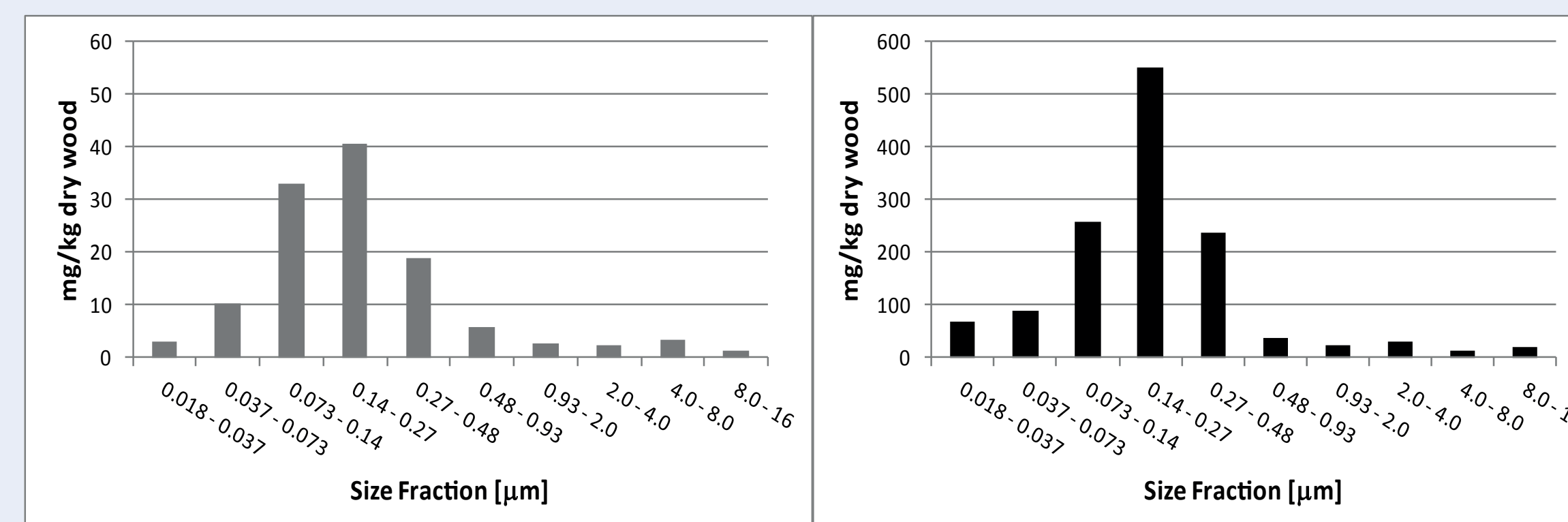


Fig. 5. Size-resolved PM emission factors for spruce (left part) and beech (right part) used as fuel.

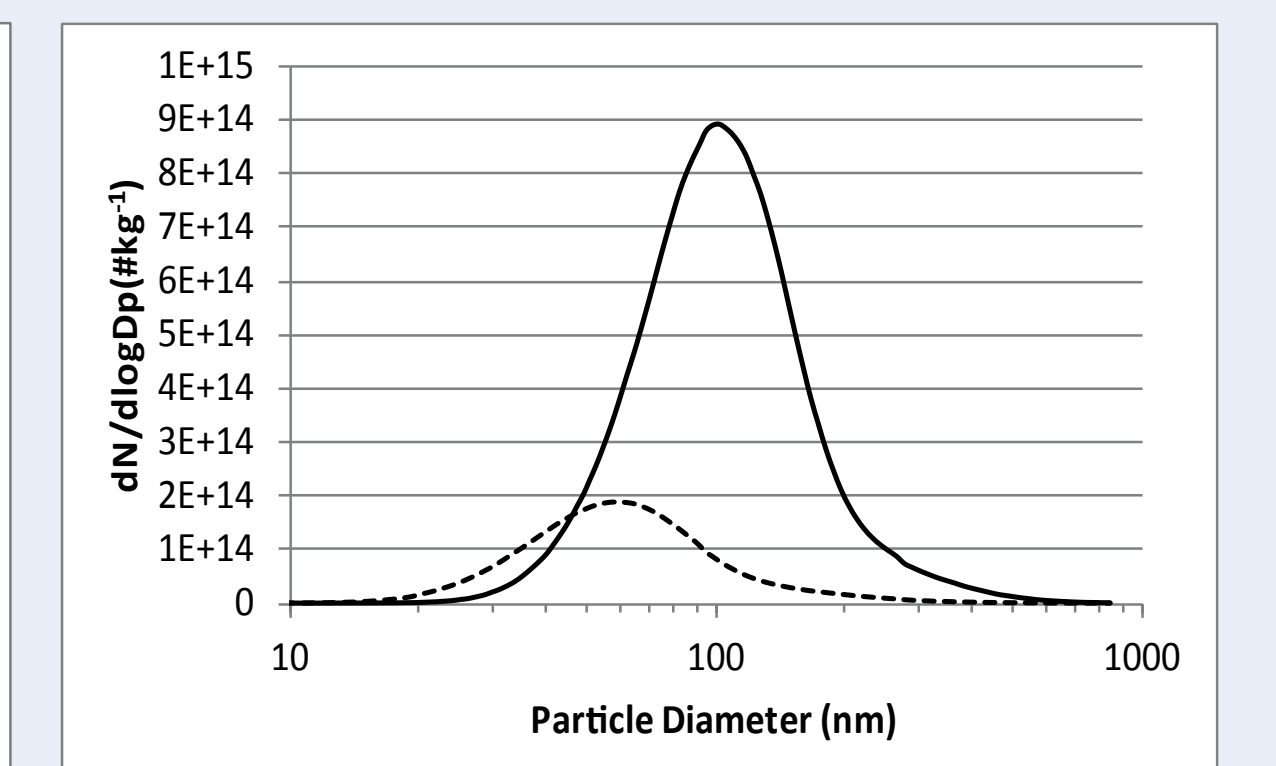


Fig. 6. Averaged particle number size distributions for spruce (dashed line) and beech (solid line).

### PAH Emissions

PAH emissions reported in Table 3 include only particulate PAH. They were determined from the sampled impactor foils. The PAH emission factors for the experiments with spruce used as fuel are significantly lower than the values reported in the literature. The higher PAH emission in the experiments with beech used as fuel could be explained by the higher combustion temperature in these experiments (Table 1), because it is known that PAH emission increase with increasing temperature. Incomplete combustion due to insufficient amount of combustion air supply could also contribute to higher PAH emission in the beech combustion experiments.

Table 3. Wood stove emission factors for particulate PAHs

Date	Fuel	$PM_{10}$	$PM_{total}$	References
		mg/kg	mg/kg	
2010-12-06	Spruce	0.24 <sup>a</sup>		McDonnell et al., 2000
2010-12-07	Spruce	0.34 <sup>a</sup>		
2010-12-08	Spruce	0.21 <sup>a</sup>		
2012-03-20	Beech	15.4 <sup>b</sup>		Fine et al., 2004
2012-03-21	Beech	10.1 <sup>b</sup>		
2012-03-27	Beech	6.7 <sup>b</sup>		
Mixed hardwoods		22 <sup>c</sup>		Pettersson et al., 2011
American tree species		2.72-7.66		
Birch, Spruce, Pine		23.4-176 <sup>d</sup>		Hedberg et al., 2002
Birch		217		
Mostly Birch		3.19 <sup>e</sup>		Tissari et al., 2007
Birch		15.5 <sup>e</sup> ; 208.5 <sup>e</sup>		
				Hytönen et al., 2009

<sup>a</sup> Emission factor calculated as mg of PAH/dry kg of fuel. <sup>b</sup> Calculated using a conversion factor of 18 MJ/kg. <sup>c</sup> Volatile PAHs (Acenaphthylene, Acenaphthene) are subtracted for comparability. <sup>d</sup> Emission factors for normal combustion. <sup>e</sup> Emission factors for smoldering combustion.

## Conclusions

The  $PM_{10}$  emission factor of 0.12 g/kg dry wood for spruce observed in this study is to our best knowledge about a factor of 3 lower than the lowest emission factor for wood stove reported in the literature. The PAH emission factor for spruce combustion is at least a factor of ten lower than the lowest factor reported. Although the same experimental setup and conditions were used, strong differences in the burning conditions, especially higher exhaust temperatures and an insufficient amount of combustion air supply caused the higher emissions in the experiments with beech logs used as fuel. From the experiments with spruce used as fuel can be seen that the emission factor for PM is clearly below the actual German limit value. A substantial reduction in the emission of PM and PAH can be realized in domestic heating using a modern wood stove, with good operational practices, and with sufficient combustion air supply.

## References and Funding

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