P1.5 WATER UPTAKE OF SOOT PARTICLES EMITTED FROM A JING-CAST SOOT GENERATOR

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1 INTRODUCTION

Hygroscopic growth of soot particles is of importance in atmospheric science since hydration may result in an increase in the specific absorption of the particles. The hygroscopicity of the soot particles in the atmosphere also affects their lifetime. Generally, it is believed that black carbon is hydrophobic and does not take up water at saturations found in the atmosphere. However, it seems that even a thin organic film on black carbon surface can render it hygroscopic (Andrews and Larson (1993), Rissler et al. (2005), Saathoff et al. (2003), Novakov and Penner (1993)). It has been reported by Noone et al. (1992) that the interstitial aerosol inside urban fogs is enriched in elemental carbon, which would tend to increase its lifetime in the atmosphere with respect to other species like organic carbon (Nunes and Pio, 1993).

Soot particles with different organic carbon content are generated with a propane burner. Aerosol number concentrations are obtained with two condensation particle counters (CPC), one working with butanol and one with water. When comparing the number counts of the two different CPCs the following behavior of the soot particles may be observed: When the organic carbon content rises above about 40%, the number counts as measured with the water CPC plunge dramatically. The butanol CPC still counts more than 10^7 particles. Therefore, soot aerosols with a predominant fraction of EC can be activated under laboratory conditions with water vapor whereas soot aerosols with a high OC content cannot be activated anymore.

2 INSTRUMENTATION

Soot particles are generated with a CAST (Combustion Aerosol STandard) soot generator of Jing-CAST Tech-

nology GmbH (http://www.sootgenerator.com). The CAST is a propane burner and soot particles are produced in a broad submicron size- and concentration range. According to the manufacturer, the characteristics of the soot particles are similar to those emitted from real combustion processes like diesel engines.
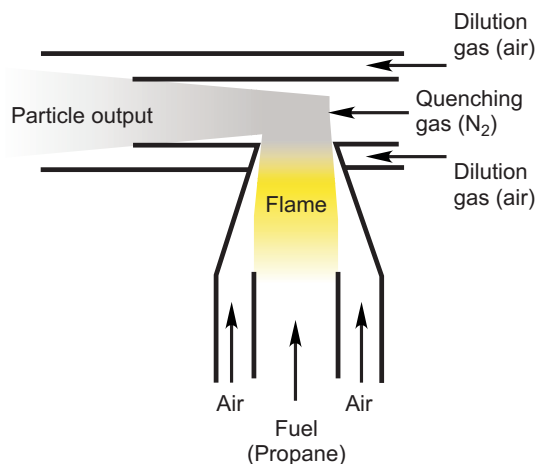

Figure 1: Propane and oxidation gas (synthetic air) are mixed and burned. The flame is extinguished by a flow of nitrogen. The resulting aerosol flow is diluted with synthetic air.

Within the CAST burner, soot particles are formed in a co-flow diffusion flame due to the hydrocarbon pyrolysis that takes place as a consequence of the heat provided by the oxydation at the flame front (see Fig. 1). The CAST burner enables the soot particles to escape from the flame without contact with oxygen. Subsequently the particle stream is mixed with a quenching gas in order to prevent further combustion processes in the particle stream and to stabilize the soot particles. The quenching inhibits condensation in the particle stream at ambient air condition. To dilute the particle stream, compressed air is supplied to the quenched particle stream.

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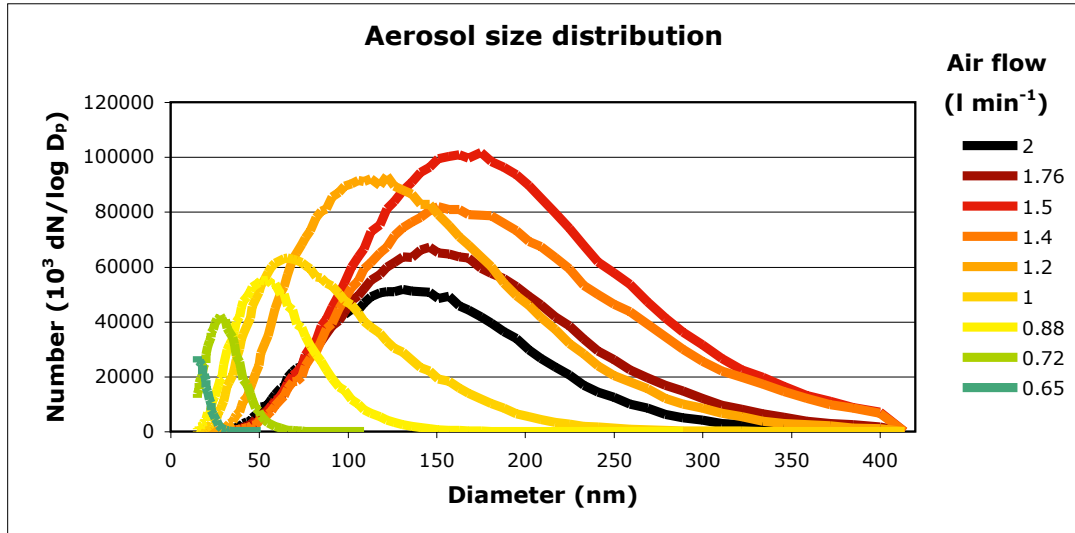e–mail: eszter.barthazy@env.ethz.ch

Figure 2: Size distribution of soot aerosol particles created with the CAST with dirrerent oxidation air flows.

By varying the oxidation air flow, flames of different heights can be generated. Therefore, the position within the flame can be varyied where the flame is extinguished and thus aerosols of different properties (number concentration, size distribution and organic/elemental carbon composition) can be generated.

Total number concentrations of the aerosol flow are measured with two condensation particle counters from TSI Incorporated. One is operating with butanol (CPC 3010) and one with water (CPC 3785). The size distribution of the soot particles are measured with a scanning mobility particle sizer (SMPS 3080 with DMA 3085 and CPC 3010, TSI).

3   EXPERIMENT

Soot particles with different properties have been generated with the CAST by varying the oxidation air flow. The operating conditions are summarized in Table 1. Right behind the exhaust, the particle output is diluted with two dilution stages (Palas VKL–10 and VKL–100) by a factor of 1000. The total length of the tubing between the exhaust of the CAST and the inlet of the CPCs (inluding the dilution stages) is in the order of 1.3 m. As shown in Fig. 2, the mean mobility diameter can be varied between about 160 and 20 nm by varying the oxidation air flow.

Aerosol total number concentrations of the diluted flow is measured with the two CPCs. The number counts of the butanol CPC were corrected according to the manufacturer since the counts are increasingly

Gas fluxes (l min^{-1})	
Propane	0.06
Oxidation air	0.5–2.0
Quenching gas (nitrogen)	7.5
Dilution air	20.0

Table 1: Operating conditions of the CAST

underestimated by the instrument because of coincidence above about 8000 particles per cubic centimeter. Results of the number counts with the two CPCs are shown in Fig. 3.
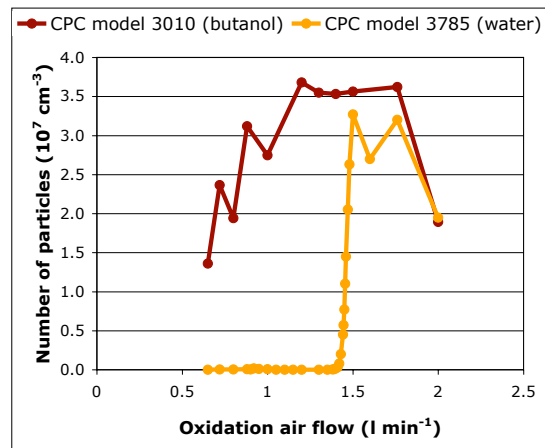

Figure 3: Total aerosol number counts as measured with a butanol and a water CPC.

As measured with the butanol CPC, aerosol number concentration is highest for oxidation air flow between about 1.2 and 1.7 l min^{-1}. For both, oxidation air flow smaller than 1.2 or larger than 1.7 l min^{-1} the aerosol number concentration is reduced. A completely different behaviour of the number concentration can be seen, when the aerosol flow is analysed with the water CPC. For high oxidation air flow, the results are comparable to the number counts with the butanol CPC. But for oxidation air flow below 1.5 l min^{-1} the number counts with the water CPC decrease rapidly until virtually no particles are counted anymore when the oxidation air flow is below 1.4 l min^{-1}.

To better understand the origin of the differences in the number counts with the two CPCs, samples for organic carbon/elemental carbon analysis were collected on quartz fibre filters and were analysed with a standard thermal method (VDI 2465/2). Figure 4 and Table 2 show that soot particles generated with a high oxidation air flow are almost entirely composed of elemental carbon (EC) whereas an oxidation air flow of below 1.5 l min^{-1} leads to the growth of soot particles with an amount of 40 to 50% of organic carbon (OC). The OC/EC composition of the present study is close to the values obtained with a different model of a CAST by Möhler et al. (2005). They report an OC content of 16% at an oxidation gas flow of 1.5 l min^{-1} and an OC content of 40% at 1.09 l min^{-1}.
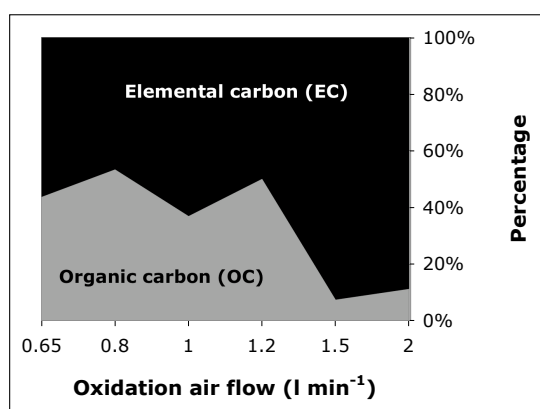

Figure 4: OC/EC composition of the soot particles in dependence of the oxidation air flow.

4 CONCLUSION

By combining the information shown in the Figs. 3 and 4, one can conclude that as long as the soot particles have a low OC content of about 10% they can be activated within a CPC with water. However, when the OC

Oxidation air flow (l min^{-1})	EC (%)	OC (%)
2.0	88.7	11.3
1.5	92.5	7.5
1.2	49.8	50.2
1.0	63.0	47.0
0.8	46.6	53.4
0.65	56.3	43.7

Table 2: OC/EC composition of the soot particles in dependence of the oxidation air flow.

content rises to about 40%, activation of the soot particles with water is not possible anymore. The OC content of the soot particles seems to be solely responsible for this observation since particle sizes do not change significantly as can be seen in Fig. 2.

In the literature it is well documented that, generally, elemental carbon particles are hydrophobic but a coating of organic carbon increases the hygroscopicity of the particles and transforms them to CCNs (Andrews and Larson (1993), Rissler et al. (2005), Ogren and Charlson (1983)). However, the particles generated with the CAST do not exhibit the same behaviour. Instead, high OC content renders the particles extremely hydrophobic. A similar behaviour is reported by Möhler et al. (2005) where ice nucleation on soot particles with an OC content of 16% is observed, but nearly inhibited for particles with OC content of 40%. The authors speculate that ice nucleation of soot aerosol with low OC content is due to the deposition nucleation mode on soot particles that are only partly covered with organic carbon. Higher OC fractions produce almost complete OC coverage and thereby suppress the deposition nucleation mode. The same process as described by Möhler et al. may be resposible for the observed inability for water uptake of the soot aerosols with high OC content in the present study.

The OC content of the freshly generated soot particles of the CAST cannot be compared to the OC content of soot particles aged in the atmosphere. Saxena et al. (1995) found that in urban areas the OC fraction of the aerosols was predominantly primary (and hence hydrophobic) whereas in rural areas the OC fraction was secondary (and thus likely to be hydrophilic). The aerosol particles found in the rural setting were aged and they may have undergone coagulation, condensation of secondary aerosol compounds and oxidation which left the now so-called secondary organic aerosols (SOA) with the OC coating observed to enhance hygroscopicity. How long this aging would take place for the CAST particles is not known and should be investigated with further experiments. Furthermore,

it seems that the OC fraction that renders soot aerosols good CCNs does not originate from burning clean fuels like propane in the present study, or, e.g., aircraft fuel with low sulfur content (Hitzenberger et al., 2003), but has to condense on the soot particles from atmospheric trace gases. This would require a certain aging time before freshly emitted soot particles could act as cloud condensation or ice nuclei.

5 OUTLOOK

Some problems with the soot generator demanded recently for a revision of the instrument. During this revision the geometry of the burner unit was changed slightly. First results show that the particle output of the generator has increased since but time was too short to obtain a thorough overview on all possible changes.

The aging process will be investigated as well as electron microscope pictures will be taken.

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