

Hygroscopic growth and droplet activation of uncoated and coated soot particles

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This discussion paper is/has been under review for the journal Atmospheric Chemistry and Physics (ACP). Please refer to the corresponding final paper in ACP if available.

Hygroscopic growth and droplet activation of soot particles: uncoated, succinic or sulfuric acid coated

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Received: 5 October 2011 – Accepted: 5 October 2011 – Published: 21 October 2011

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Published by Copernicus Publications on behalf of the European Geosciences Union.

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Abstract

The hygroscopic growth and droplet activation of uncoated soot particles and such coated with succinic acid and sulfuric acid were investigated during the IN-11 campaign at the Aerosol Interaction and Dynamics in the Atmosphere (AIDA) facility. A GFG-1000 soot generator applying nitrogen, respectively argon as carrier gas and a miniCAST soot generator were utilized to generate soot particles. Different organic carbon (OC) to black carbon (BC) ratios were adjusted for the CAST-soot by varying the fuel to air ratio. The hygroscopic growth was investigated by means of the mobile Leipzig Aerosol Cloud Interaction Simulator (LACIS-mobile) and two different Hygroscopicity Tandem Differential Mobility Analyzers (HTDMA, VHTDMA). Two Cloud Condensation Nucleus Counter (CCNC) were applied to measure the activation of the particles. For the untreated soot particles neither hygroscopic growth nor activation was observed, with exception of a partial activation of GFG-soot generated with argon as carrier gas. Coatings of succinic acid lead to a detectable hygroscopic growth of GFG-soot and enhanced the activated fraction of GFG- (carrier gas: argon) and CAST-soot, whereas no hygroscopic growth of the coated CAST-soot was found. Sulfuric acid coatings lead to an OC-content dependent hygroscopic growth of CAST-soot. Such a dependence was not observed for activation measurements. Coating with sulfuric acid decreased the amount of Polycyclic Aromatic Hydrocarbons (PAH), which were detected by AMS-measurements in the CAST-soot, and increased the amount of substances with lower molecular weight than the initial PAHs. We assume, that these reaction products increased the hygroscopicity of the coated particles in addition to the coating substance itself.

1 Introduction

Atmospheric aerosol particles interact with solar radiation directly by scattering and/or absorbing it and indirectly by acting as Cloud Condensation Nuclei (CCN) and by

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changing cloud microphysical properties. Due to human activities the number of aerosol particles in the atmosphere has increased, which led also to an increased number of potential CCNs. Cloud properties such as the number and size of droplets depend on the CCN number and also on water vapor available. This may cause also changes in precipitation, cloud lifetime (Albrecht, 1989) and cloud albedo, known as the Twomey effect (Twomey, 1974). The atmospheric aerosol consists of many different particle types, e.g., sea salt, pollen, mineral dust or soot. We here focus on the hygroscopic and CCN properties of soot particles and their respective modification in mixtures with sulfuric and succinic acid. The latter to simulate chemical ageing by inorganic and organic compounds in the atmosphere.

Soot particles are agglomerates of carbon containing particles, which are formed during incomplete combustion processes (Seinfeld and Pandis, 2006). Soot particles contain absorbing graphite like carbon (black carbon, BC) and at the same time many other carbon containing substances (organic carbon, OC). From a morphological point of view, soot particles are fractals. They consist of chains of primary particles with internal voids, which may compact due to wetting with water or other fluids.

Main sources of atmospheric soot particles are diesel engines, biomass burning and domestic heating (Horvath, 1993). Diesel engines are the main source for soot particles in urban areas. For example 25 to 30 %, 93 % and 47 % of the soot particles are emitted by traffic in Vienna/Austria, Los Angeles/USA and Detroit/USA, respectively (Horvath, 1993; Pratsinis et al., 1988; Wolf and Korsog, 1985).

The soot mass fraction is 1.1 to 2.5 % of the anthropogenically emitted aerosol and 0.2 to 1 % of the total atmospheric aerosol (Horvath, 1993). 98 % of the soot particles are removed by precipitation (Jacobson, 2004). The annual emission of BC is estimated to 4.3 and 24 Tg a⁻¹ (Bond et al., 2004; Cooke and Wilson, 1996; Cooke et al., 1999; Horvath, 1993; Ito and Penner, 2005; Juncker and Liousse, 2008; Kim et al., 2008; Kanakidou et al., 2005; Novakov et al., 2003; Penner et al., 1993; Tegen et al., 2000).

The residence time of soot particles in the atmosphere is about seven days. That is long enough to transport the particles hundreds to thousands of kilometers, but too short for an interhemispheric transport (Horvath, 1993).

By nature soot particles are hydrophobic and insoluble in water, but during their presence in the atmosphere, soluble substances condense on the particles and alter their cloud-forming potential (Andreae and Rodenfeld, 2008).

At relative humidities lower than 95 % investigations regarding the hygroscopic growth of soot particles were performed by several groups. Meyer and Ristovski (2007) and Weingartner et al. (1997) for example investigated diesel soot and Gysel et al. (2003) soot from aviation fuel. The reported growth factors (GF, defined as the measured wet diameter divided by the selected dry diameter) for these soot types ranged between 1 and 1.28. The GF increased with increasing sulfur content of the fuel. Weingartner et al. (1997) and Henning et al. (2010) used the same type of spark generator with nitrogen as carrier gas as was used in this study. They observed no hygroscopic growth for the uncoated spark soot, but after coating with soluble substances a hygroscopic growth occurred (Henning et al., 2010). Khalizov et al. (2009) and Zhang et al. (2008) investigated the hygroscopic growth of sulfuric acid coated flame soot up to 90 % relative humidity (RH). They found growth factors up to 1.5 and at a certain RH a decrease in GF with increasing selected mobility diameter.

The activation of soot particles was investigated by Lammel and Novakov (1995) using diesel soot, by Hitztenberger et al. (2003); Hudson et al. (1991); Pitchford et al. (1991) using soot generated via the combustion of aviation fuel, by Hagen et al. (1989) using soot from white gas and ethanol combustion and by Hudson et al. (1991) using soot from different wood types and crude oil. The particles were found to activate better with increasing sulfur content of the fuel. No (complete) activation was found for soot from aviation fuel, wood and crude oil. Henning et al. (2010) observed, that coated spark soot acts as CCN. Zhang et al. (2008) investigated the activation of sulfuric acid coated flame soot at two supersaturations (0.2 and 0.4 %), but they found no activation for the uncoated soot at the same supersaturations.

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The IN-11 measurement campaign at the AIDA was carried out to get comprehensive data regarding hygroscopic growth, cloud droplet activation and ice nucleation potential, which is required to determine the importance of fresh and aged soot particles in cloud formation and freezing. The mobile version of the Leipzig Aerosol Cloud Interaction Simulator (LACIS-mobile, IFT home-built), a Hygroscopicity Tandem Differential Mobility Analyzer (HTDMA, Buchholz, 2007) and a Volatility Hygroscopicity Tandem Differential Mobility Analyzer (VHTDMA, LaMP home-built) were applied to investigate the hygroscopic growth up to 98.4 % RH as well as two Cloud Condensation Nucleus Counters (CCNC, DMT, Boulder, CO, USA, Roberts and Nenes, 2005) for activation measurements between 0.14 and 1.54 % supersaturation. The ice formation potential as discussed in Crawford et al. (2011), was investigated applying the AIDA-chamber (Möhler et al., 2001). Soot particles were generated by means of either a flame soot generator (miniCAST, Jing, 2000) or a spark soot generator (GFG-1000, Helsper et al., 1993). These particles were investigated with and without being coated with either succinic acid or sulfuric acid.

2 Experimental setup

The experiments were conducted in the framework of the AIDA infrastructure at the Karlsruhe Institute for Technology in order to collect comprehensive data for the hygroscopic growth, cloud droplet activation and ice forming potential of different kinds of soot particles. The setup is shown schematically in Fig. 1 and described in the following.

Experiments were typically conducted according to the following cycle: the soot particles were generated by utilizing either the miniCAST or the GFG generator. Afterwards the particles were either coated or used without any treatment. A small aerosol chamber (NAUA chamber) was used as aerosol reservoir for the whole experiment. Aerosol was feed into the NAUA chamber until a number concentration of around 10^4 – 10^5 cm^{-3} was reached. Afterwards the AIDA chamber was filled with a part of the aerosol from

the NAUA chamber, which was refilled with particle free dry synthetic air to regain atmospheric pressure inside. Thereafter, the NAUA chamber was available as aerosol source for all other instruments (Fig. 1). During aerosol sampling NAUA was permanently refilled with synthetic air to hold the pressure at atmospheric levels. The aerosol in the NAUA and AIDA chambers was polydisperse, but all measurements presented here were done for size-selected particles.

An overview of all conducted experiments including the generation and coating settings is given in Table 1.

2.1 Particle generation

Two soot types were investigated, generated by means of the two (see below) different soot generators. One was a graphite spark soot generator (GFG-1000, Palas GmbH, Karlsruhe, Germany, Helsper et al., 1993) with either nitrogen or argon as carrier gas (purity 6.0, from now on referred to as “GFG-soot”). The carrier gas was varied, because earlier measurements suggested an influence of the carrier gas on the hygroscopic properties of GFG-soot. The OC-content of the GFG-soot was 12 ± 9 wt. %.

The second was a miniCAST flame soot generator (miniCAST, Jing-CAST Technology GmbH, Switzerland, Jing, 2000, , from now on referred to as “CAST-soot”). The OC-content of the CAST-soot was adjusted by the fuel (propane) to air ratio, leading to particles with high OC-content (> 80 wt. %, max OC), medium OC-content (78 ± 2 wt. %, med OC) and low OC-content (32 ± 4 wt. %, min OC). The OC-content was determined following VDI 2465 (1999), considering material as organic carbon which is volatile in helium at temperatures below or equal 600°C .

2.2 Coating section

For the soot particle coating, a flow of particle-free synthetic air was saturated with the coating substance at the temperature given in Table 1, then further heated to the mixing zone temperature, which was about 10 K higher than the saturator temperature.

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The aerosol flow was heated to the same temperature and added to the mixing zone. Afterwards the temperature of the aerosol-gas-mixture was reduced continuously down to about room temperature to condense the coating substance on the particles.

Succinic acid and sulfuric acid were used as coating substances. Succinic acid was chosen as an example for an oxygenated organic substance to mimic soot aged in biogenically influenced environments. Succinic acid is found frequently in the atmospheric aerosol (Chebbi and Carlier, 1996). It features a low vapor pressure (10^{-3} Pa at 25 °C) and moderate solubility (83.2 g l^{-1} at 25 °C; Bilde et al., 2003; Howard and Meylan, 1997).

Sulfuric acid is formed in the atmosphere via oxidation of SO_2 , which is emitted by e.g. combustion of sulfur containing fuels (Seinfeld and Pandis, 2006). Soot particles coated with sulfuric acid are a model aerosol for particles, which are aged in anthropogenically influenced regions.

2.3 LACIS-mobile

The mobile version of LACIS (LACIS-mobile) was used in this study. This setup is similar to LACIS as described by Stratmann et al. (2004), but features a shorter flow tube of 0.8 m length. Aerosol and sheath air were humidified applying water-to-gas Nafion humidifiers (aerosol: MH-110-12S-4, sheath air: PH-30T-24KS, Perma Pure) to well defined dew point temperatures (reproducibility approx. 0.01 K) before entering the flow tube. The dew point temperatures and the LACIS-mobile wall temperature determine the water vapor saturation in the flow tube. For the hygroscopic growth measurements described here, LACIS-mobile was operated in its sub-saturated mode of operation (Wex et al., 2005). In this mode the wall temperature (20 °C) was above the dew point temperature (between 19.7 and 19.95 °C) of the aerosol and the sheath air. The residence time of the particles in the flow tube is around two seconds, which is sufficient for particles to reach their wet equilibrium diameter. At the end of the flow tube, an optical particle spectrometer detects the size of the grown particles (Kiselev et al., 2005).

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For the investigations presented here, the spectrometer was calibrated at least every three days using PSL particles with diameters between 274 and 1020 nm (Duke Scientific). The refractive index used to retrieve the droplet size from the measured scattered light intensities was $1.74 + 0.65i$ for soot. For the retrieval of the wet particle size, the coating on the soot was assumed to be highly diluted and thus the refractive index of pure water of 1.33 was applied. A sensitivity analysis proofed that the influence of the succinic or sulfuric acid coating on the refractive index and thereby on the retrieved wet sizes is negligible (not shown in this paper).

RH in LACIS-mobile was calibrated with ammonium sulfate particles of known dry sizes (size-selected by a DMA). The calibration measurements were repeated at least every third day.

2.4 HTDMA

The hygroscopic growth of the particles was also measured with a HTDMA (Buchholz, 2007). 150 or 200 nm particles were size-selected with a first DMA from the dried polydisperse aerosol. The monodisperse aerosol and the sheath air of a second DMA were humidified at room temperature (25 °C) using gas-to-gas humidifiers (Perma Pure Nafion, MH-Series) to almost the same RH value; with the sheath air being at slightly higher RH. The second DMA (measuring the size of the conditioned particles) is placed in an insulated box and was cooled to 20 °C. Before entering the second DMA both aerosol and sheath air were cooled down to this temperature and thus the RH increased to its final value. To determine the RH inside the second DMA, Vaisala HMP235 sensors measured the RH and temperature outside the insulation area. The temperatures of the air flows inside the insulation box were measured with PT100 sensors directly before entering and immediately after leaving the second DMA. With the temperature and RH measured outside the absolute humidity in the air streams is calculated. Assuming the absolute humidity being constant, the RH in the cooled (second) DMA can be calculated from the temperature measured inside the cooled area. For RH calibration ammonium sulfate aerosol was used.

2.5 VHTDMA

Also a VHTDMA was applied to investigate the hygroscopic and volatility behavior of soot particles (Villani et al., 2008). Particles with 150 nm in diameter were selected from the polydisperse aerosol using a first DMA. The RH in the first DMA was below 20%. The VHTDMA used has a thermodenuder section followed by a humidifier section between the first and a second DMA. The aerosol flow can be led through both sections or bypass either one, resulting in either volatility or hygroscopicity or combined volatility/hygroscopicity measurements. If the humidifier section was used, the sheath air of the second DMA was humidified; otherwise, it was kept dry. Both DMAs run as closed loop systems, and their flows are adjusted by means of flow controllers. Water-to-gas nafion humidifiers are used to condition the sheath air of the second DMA (Perma Pure PH-60T-12SS) and the aerosol flow (Perma Pure MD-110-12S-4). Capacitance humidity sensors (Rotronic, HygroClip SC05) are applied to monitor the RH, which are calibrated with ammonium sulfate particles.

2.6 CCNC

Two streamwise thermal gradient cloud condensation nucleus counters (CCNC, Roberts and Nenes, 2005) were applied to investigate the activation of the particles. In this instrument the inlet flow is split into a particle-free sheath air flow, which is kept particle-free via a filter and an aerosol flow. Only the sheath air is humidified before entering the flow tube and surrounds the aerosol at the centerline. The streamwise temperature gradient applied in the flow tube determines the supersaturation to which the particles are exposed. If the supersaturation is high enough to activate the particles into droplets, the number of these is detected at the end of the flow tube with an OPC. The CCNC is either used to measure saturations scans, meaning that the particle diameter is kept constant and the saturation is varied, or to measure diameter scans for which the saturation is fixed and the diameter is varied. The firstly mentioned saturation scan results in a critical supersaturation, which by definition is the saturation, at which

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50% of the particles become activated (activated fraction (AF) equal 0.5). Analogous a diameter scan allows for the detection of the critical diameter. However, this is only possible if full activation is reached within the available diameter and saturation matrix, which was not always the case for the pure soot particles. In the work presented here we run saturation scans.

Ammonium sulfate particles were used to calibrate the supersaturation reached in the CCNC. Calibration measurements were repeated at least every fourth day.

A HR-TOF-AMS (Aerodyne Research Inc.) connected to the NAUA chamber was used to characterize the coatings in a few cases. The AMS is described in detail in Jayne et al. (2000), and DeCarlo et al. (2006).

3 Results and Discussion

An overview of all experiments is given in Table 2 and their results are presented in the following sections according to the soot type. The hygroscopic growth of the different particle types is presented in the growth factor notation. No corrections regarding the particle shape were done, because no information on particle shape is available.

3.1 Uncoated soot

For uncoated CAST-soot particles hygroscopic growth was not observed for any OC-content.

Also the GFG-soot did not show hygroscopic growth in the LACIS-mobile measurements with either one of the carrier gases. Measurements with the HTDMA and VHTDMA (Fig. 2) indicated even a shrinking with increasing RH by up to 5% ($GF = 0.95$), which is likely due to compaction of the particles when getting into contact with water at $RH > 35\%$.

Activation was also not observed for the different CAST-soot types and for the GFG-soot with nitrogen as carrier gas at the highest supersaturation of about 1.5% reached

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in the CCN instrument. However, the activated fraction did reach up to 95 % at a supersaturation of 1.5 % if argon was employed as carrier gas in the GFG-soot production (see Fig. 3). At present we cannot explain the influence of the carrier gas on the activation of GFG-soot.

Neither hygroscopicity nor droplet activation of the uncoated CAST-soot particles depend on the OC content (see Table 2). The organic substances on the soot are possibly not wettable or miscible with water. This is consistent with findings by Dusek et al. (2006), who observed an increased activation supersaturation of soot particles after having coated them with an organic substance (hexadecanol, $C_{16}H_{34}O$). They attributed this to a reduced wettability of the particles due to the organic coating.

3.2 Succinic acid coated soot

CAST-soot with medium OC-content and GFG-soot with argon as carrier gas were coated with succinic acid (see Table 1). No hygroscopic growth was observed for the coated CAST-soot by all three instruments. As depicted in Fig. 4, a shrinking of the coated GFG-soot was observed with the HTDMA. Hygroscopicity measurements of the same soot with LACIS-mobile gave a growth factor of 1.11 at 98.4 % RH for particles with 375 and 500 nm dry diameter. The observed difference is most likely due to the different “humidity history” of the particles. Gas-to-gas humidifier are applied in the HTDMA, whereas water-to-gas humidifier are utilized in LACIS-mobile. Using the latter, particles are wetted at up to 100 % RH in the water-to-gas humidifier and deliquescence of succinic acid occurs (at 99 % RH, Wex et al., 2007). In contrast, gas-to-gas humidifiers do not reach RHs high enough to reach deliquescence of succinic acid. Therefore LACIS-mobile measured probably soot with a succinic acid solution and the HTDMA soot coated with solid succinic acid.

For succinic acid coated GFG-soot an increase of the hygroscopic growth (applying LACIS-mobile) and the activated fraction compared to the uncoated soot was observed. In contrary, the coating of CAST-soot with succinic acid increased only the activated fraction and had no influence on the hygroscopic growth. Also the coated

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GFG-soot depicted a higher activated fraction at the same supersaturation than the coated CAST-soot (Fig. 5). For both soot types the coating section was operated under similar conditions (see Table 1), thus should have led to a similar coating thickness of the particles. The difference between CAST- and GFG-soot could be explained by the formation of a mixed liquid layer of succinic acid and organics on the CAST-soot particles in contrast to adsorption of succinic acid to the almost pure black carbon surface of the GFG-soot, as illustrated in Fig. 6. As a result of such a process, the contribution of succinic acid to hygroscopic growth and activation could be reduced or suppressed leading to undetectable hygroscopic growth and only partial activation.

3.3 Sulfuric acid coated soot

CAST-soot with low, medium and high OC-content was coated with sulfuric acid (see Table 1). Hygroscopic growth increased with increasing OC-content, as depicted in Fig. 7. This general trend is in agreement for all instruments. However, considering the instrumental error bars discrepancies are remaining. At the moment we have no explanation why some measurements of sulfuric acid coated CAST-soot deviate, while all others agree within the error bars. We can exclude malfunction of any instrument, since repeated calibrations of the relative humidity (and for LACIS-mobile the OPC) showed a high reproducibility for all instruments. Therefore one might speculate, that the chemical composition (OC-content, amount of sulfuric acid, ...) is a function of the particle size. Then the measured GF's would be different, because the instruments selected different mobility sizes (HTDMA 150 nm, VHTDMA 100/150 nm and LACIS-mobile 250 nm), which was due to instrumental limitations.

We observed an increasing hygroscopic growth with increasing OC content. This could be caused by a reactive uptake of sulfuric acid by the OC, leading to an increasing amount of soluble material on the particles with increasing OC-content. This could have caused enhanced growth factors for the higher OC-contents due to the dependence of the formation of the soluble material on the OC-content. Our results are consistent with findings from Khalizov et al. (2009), who investigated flame soot. They observed no

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water uptake below 100 % for untreated particles, but water uptake for the sulfuric acid coated particles. Zhang et al. (2008) reported a size dependent uptake of sulfuric acid at flame soot, which caused a decrease in hygroscopic growth with increasing particle size. This is in contrast to our measurements, however they used a different burner type resulting in lower OC-contents (about 1 %, Pagels et al., 2009).

Coating with sulfuric acid caused complete activation of the CAST-soot without dependence on the OC-content (cf. Fig. 8).

Measurements of the chemical composition were carried out by means of an HR-ToF-AMS. Polycyclic aromatic hydrocarbons (PAH) were found in the fraction of the OC of the CAST soot volatile at 600 °C. PAHs are slightly soluble or even insoluble in water and the solubility decreases with increasing molecular mass. The comparison of the mass spectra of untreated soot and sulfuric acid coated soot showed that the fraction of mass peaks with $m/z > 150$ dropped and the fraction with $m/z < 150$ increased, as illustrated in a difference mass spectrum (Fig. 9). Probably the sulfuric acid reacts with the PAHs (as illustrated in Fig. 10) and forms products with lower molecular weight than the initial PAHs. These products could have a higher solubility in water than the initial PAHs. As a consequence, the hygroscopic growth and activated fraction increased due to (a) the products of this reaction and (b) the unconsumed coating fraction itself.

Reaction of sulfuric acid with PAH have been observed before (Granello and Clonfero, 1991). Ester are possible products of this reaction (Banoglu and Duffel, 1999, and references therein). Sulfuric acid coated flame soot was also investigated by Zhang et al. (2008). In contrast to our results, they found no hints for interactions of sulfuric acid with any substance on the soot. The particle mass before coating was identical to the mass found after the removal of the coating. However, note that Zhang et al. (2008) generated the soot particles with a different burner type, which produces flame soot with a lower OC-content (about 1 % Pagels et al., 2009) than used in our experiments.

In contrast to the CAST-soot, no PAHs were found on the GFG-soot. A substance condensed on the GFG-soot would increase the activated fraction and hygroscopic

growth by itself. A comparison of the AMS spectra of untreated and succinic acid coated GFG-soot gave no hints for an interaction of succinic acid and substances at the GFG-soot as the difference mass spectrum was identical to the mass spectrum of succinic acid.

4 Summary and conclusions

The hygroscopic growth and cloud droplet activation of uncoated and coated soot particles were investigated. A flame soot generator (miniCAST) and a spark soot generator (GFG-1000) were utilized to generate soot particles. CAST-soot particles with either low, medium or high OC-content were produced and nitrogen or argon were used as carrier gases for the GFG-1000. Investigations were conducted on untreated soot particles and on soot coated with either succinic or sulfuric acid.

As summarized in Table 2, neither hygroscopic growth nor activation were observed for the untreated soot particles – with one exception; for GFG-soot with argon as carrier gas activation was observed. We have no explanation for the influence of the carrier gas on the activation behavior.

The activated fraction of CAST- and GFG-soot was increased by coating with succinic acid. For succinic acid coated CAST-soot, no water uptake at subsaturation was observed. For GFG-soot coated with succinic acid hygroscopic growth was observed with LACIS-mobile, but not with the HTDMAs. The instrumental discrepancy is explained by the “humidity history” of the particles, i.e. LACIS-mobile measured soot with deliquescent succinic acid whereas the HTDMAs measured solid succinic acid on the soot. The difference between CAST- and GFG-soot could be explained by the formation of a mixed liquid layer of succinic acid and organics on the CAST-soot particles in contrast to adsorption of succinic acid to the almost pure black carbon surface of the GFG-soot.

Sulfuric acid was applied to coat CAST-soot. An increase of the hygroscopic growth was observed with increasing OC-content. However, no dependence of the cloud

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droplet activation on the OC-content was observed. AMS-measurements showed the presence of PAHs, the amount of which was decreased by coating with sulfuric acid. At the same time the fraction of lighter mass peaks increased. This led to the conclusion that the amount of PAHs and their chemical processing by the uptake of reactive substances like sulphuric acid may play a key role in understanding hygroscopic growth and droplet activation of flame soot. Sulfuric acid acts in two ways: (a) it reacts with PAHs forming thereby lighter molecular weight products with changed hygroscopic properties and (b) the amount of sulfuric acid which is not consumed in the reaction increases the hygroscopicity by itself.

Acknowledgements. We are grateful for financial support by the Helmholtz Virtual Institute "Aerosol Cloud Interaction" (VI-ACI). Traveling expenses of the LaMP group as well as the VHTDMA transport were funded by the ESF programme INTROP, which we would like to acknowledge here. Furthermore, we would like to thank Stefan Benz for coordinating the IN-11 campaign at the AIDA facility.

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Table 1. Experimental settings of NAUA experiments during IN-11.

NAUA Experiment	Generator	Carrier gas	OC-content	Coating substance	Aerosol flow saturator (l min ⁻¹)	T saturator (°C)
1,2,4	GFG	Argon		Uncoated		
3	GFG	Nitrogen		Uncoated		
6	GFG	Argon		Succinic acid	5	90
7	miniCAST		minOC	Uncoated		
15	miniCAST		medOC	Uncoated		
10	miniCAST		maxOC	Uncoated		
16,17	miniCAST		medOC	Succinic acid	5	90/95
9	miniCAST		minOC	Sulfuric acid	5	55
13,14	miniCAST		medOC	Sulfuric acid	2/5	70/95
12	miniCAST		maxOC	Sulfuric acid	1	45

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Table 2. Overview of experimental conditions and results of NAUA experiments during IN-11. The growth factor (GF) at 98.4% RH is given for LACIS-mobile. The activated fraction (AF) at a certain supersaturation (SS) is given, if no complete activation was observed. For complete activation the critical diameter (d_c) is given at a supersaturation of 0.24, 0.27 or 0.30 %, respectively.

Generator	Carrier gas/OC-content	Coating	Hygroscopic growth	Activation	OC fraction from filter analysis in wt. %*
GFG	Argon	Uncoated	Not observed, shrinking	AF = 0.27 at SS = 0.97 ± 0.04 %, $d_{p,0} = 175$ nm	12 ± 9 %
GFG	Nitrogen	Uncoated	Not observed, shrinking	AF = 0.04 at SS = 0.97 ± 0.04 %, $d_{p,0} = 175$ nm	
GFG	Argon	Succinic acid	GF = 1.11 (RH = 98.4 ± 0.1 %) shrinking (HTDMA)	$d_c = 175$ nm, SS = 0.30 ± 0.01 %	$12 + x$ %
miniCAST	Min OC	Uncoated	Not observed	AF = 0.002 at SS = 1.38 ± 0.06 %, $d_{p,0} = 175$ nm	32 ± 4 %
miniCAST	Med OC	Uncoated	Not observed	AF = 0.006 at SS = 1.03 ± 0.04 %, $d_{p,0} = 175$ nm	78 ± 2 %
miniCAST	Max OC	Uncoated	Not observed	AF = 0.007 at SS = 1.03 ± 0.04 %, $d_{p,0} = 175$ nm	>80%
miniCAST	Med OC	Succinic acid	Not observed	AF = 0.3 at SS = 1.03 ± 0.04 %, $d_{p,0} = 175$ nm	$78 + x$ %
miniCAST	Min OC	Sulfuric acid	GF = 1.15 (RH = 98.4 ± 0.1 %)	$d_c = 215$ nm, SS = 0.27 ± 0.01 %	32 ± 4 %
miniCAST	Med OC	Sulfuric acid	GF = 1.37 (RH = 98.4 ± 0.1 %)	$d_c = 183$ nm, SS = 0.24 ± 0.01 %	78 ± 2 %
miniCAST	Max OC	Sulfuric acid	GF = 1.91 (RH = 98.4 ± 0.1 %)	$d_c = 151$ nm, SS = 0.27 ± 0.01 %	> 80 %

* OC values: personal communication C. Linke, OC fraction determined following the method VDI 2465 (1999).

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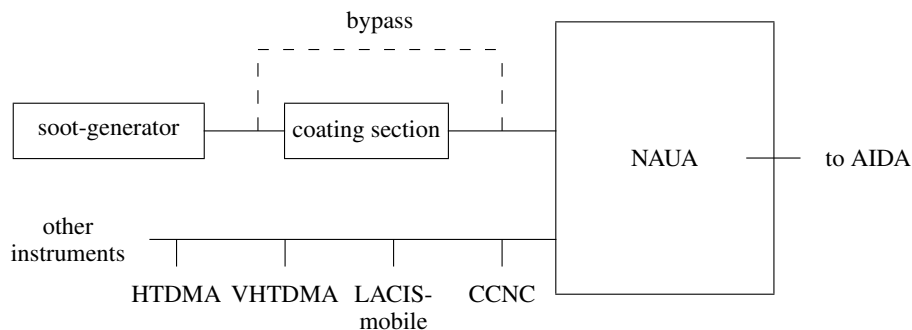


Fig. 1. Experimental setup.

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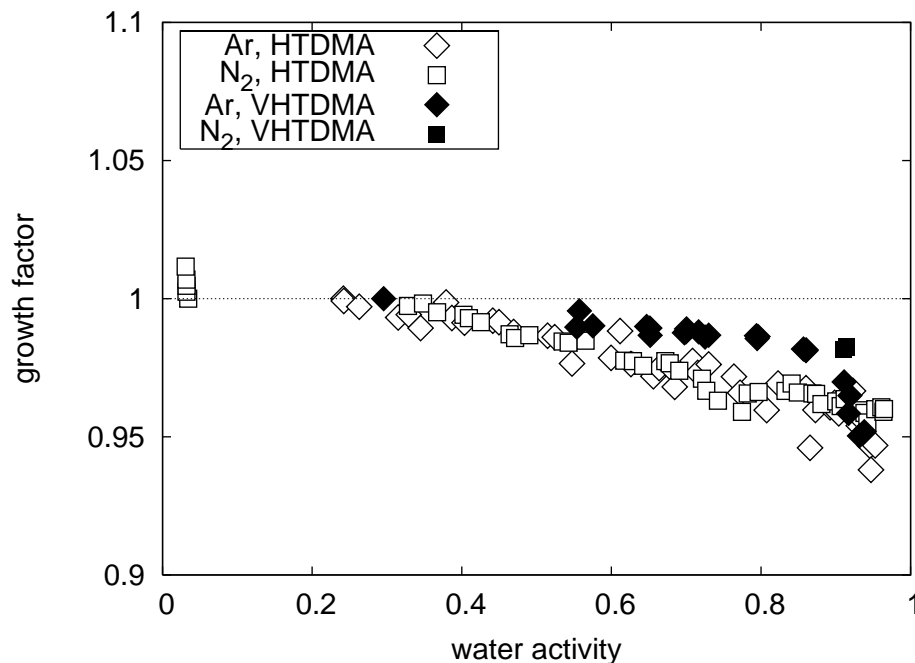


Fig. 2. Hygroscopic growth of GFG-soot measured with HTDMA (open symbols) and VHTDMA (filled symbols), carrier gases argon (diamonds) and nitrogen (squares).

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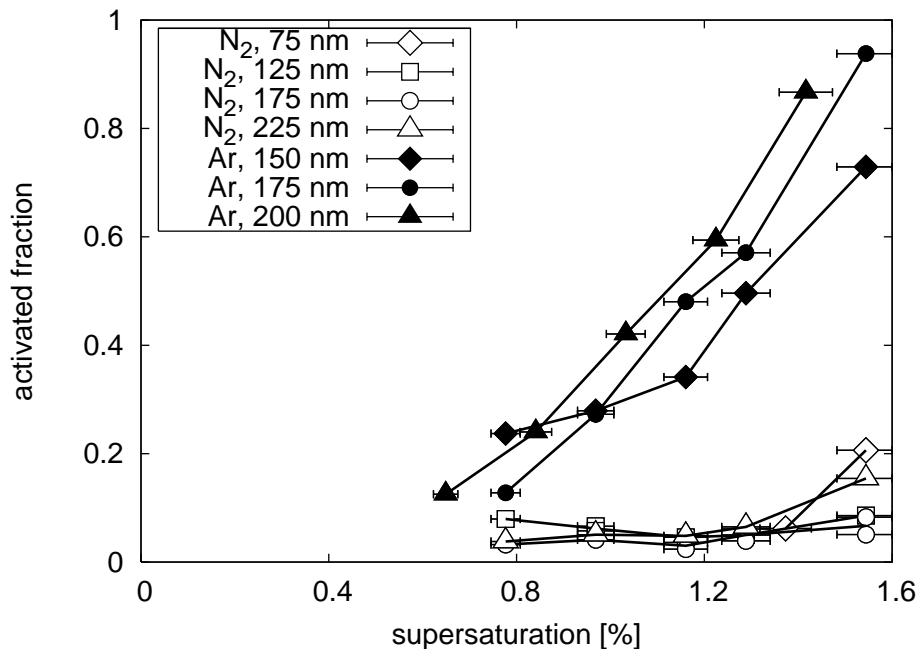


Fig. 3. Activation of GFG-soot measured with CCNC, carrier gases argon (filled symbols) and nitrogen (open symbols).

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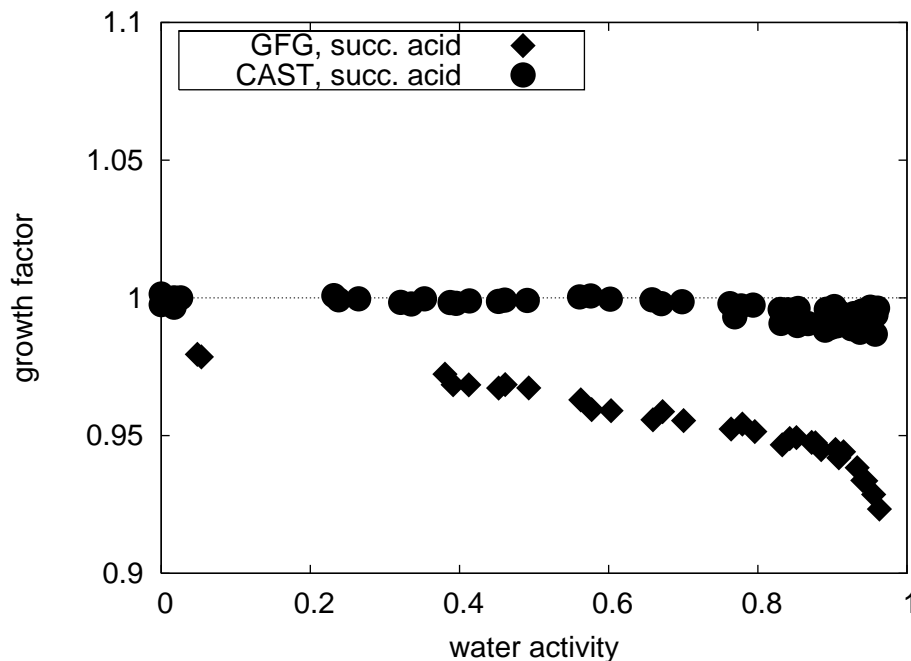


Fig. 4. Hygroscopic growth of succinic acid coated GFG-soot (diamonds, carrier gas argon) and CAST-soot (circles, medium OC-content) measured by the HTDMA.

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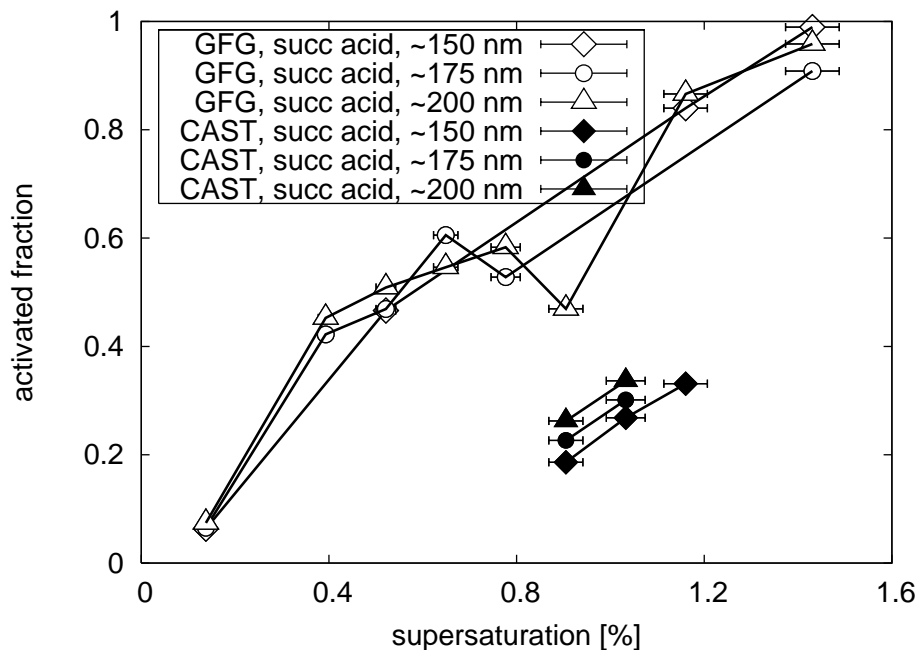


Fig. 5. Activation of succinic acid coated GFG-soot (carrier gas argon) and CAST-soot (medium OC-content).

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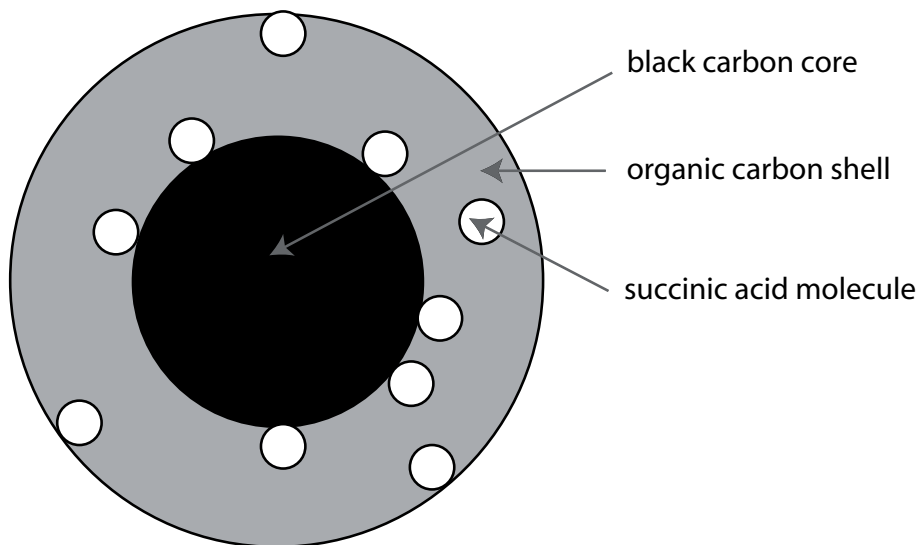


Fig. 6. Illustration for covering succinic acid molecules by OC, as assumed for CAST-soot.

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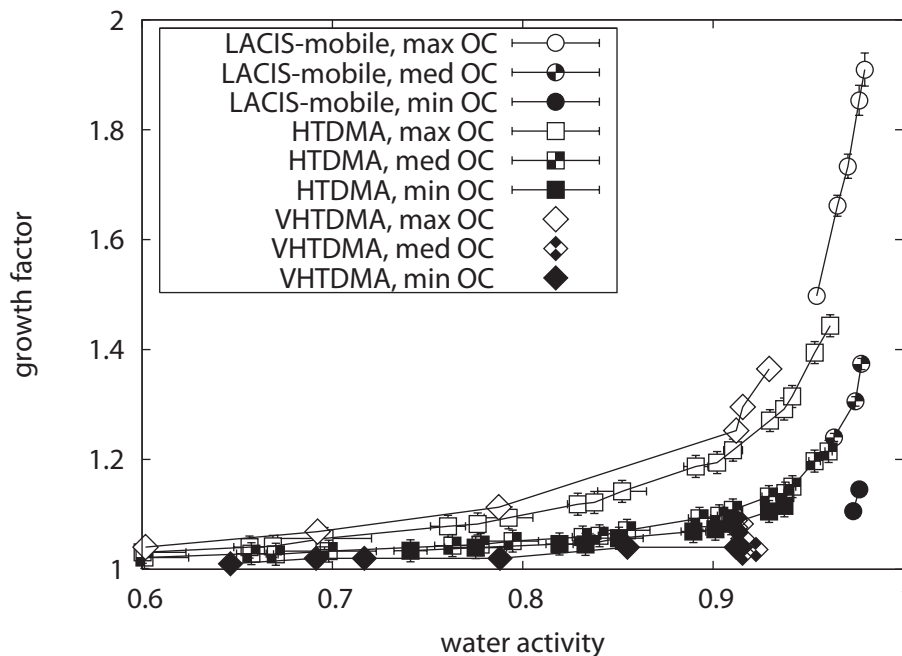


Fig. 7. Hygroscopic growth of sulfuric acid coated CAST-soot with low (filled), medium (partial filled) and high (open) OC-content. Applied instruments: LACIS-mobile (circles), HTDMA (squares) and VHTDMA (diamonds).

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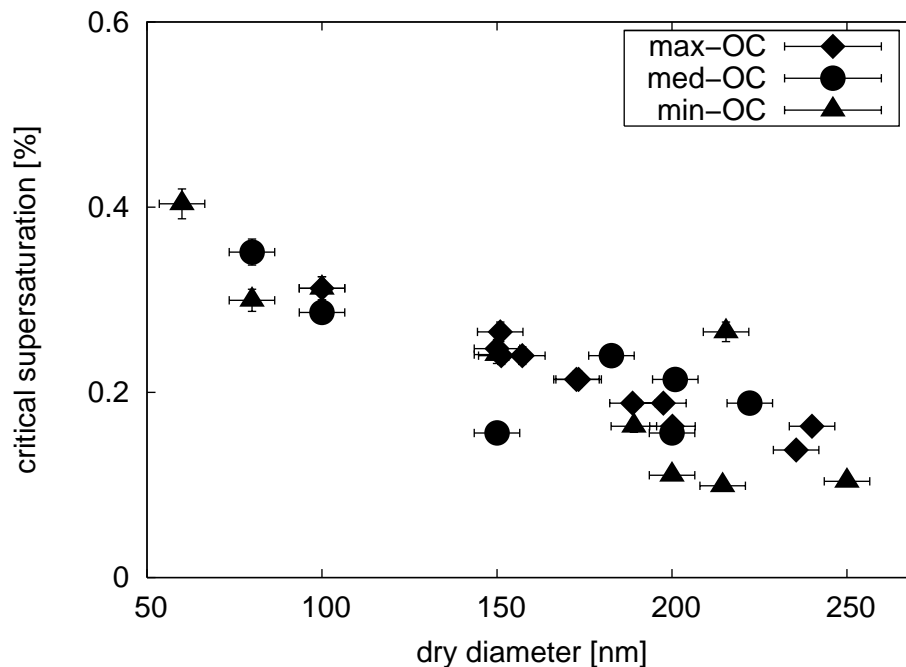


Fig. 8. Activation of sulfuric acid coated CAST-soot with low (triangles), medium (circles) and high (diamonds) OC-content.

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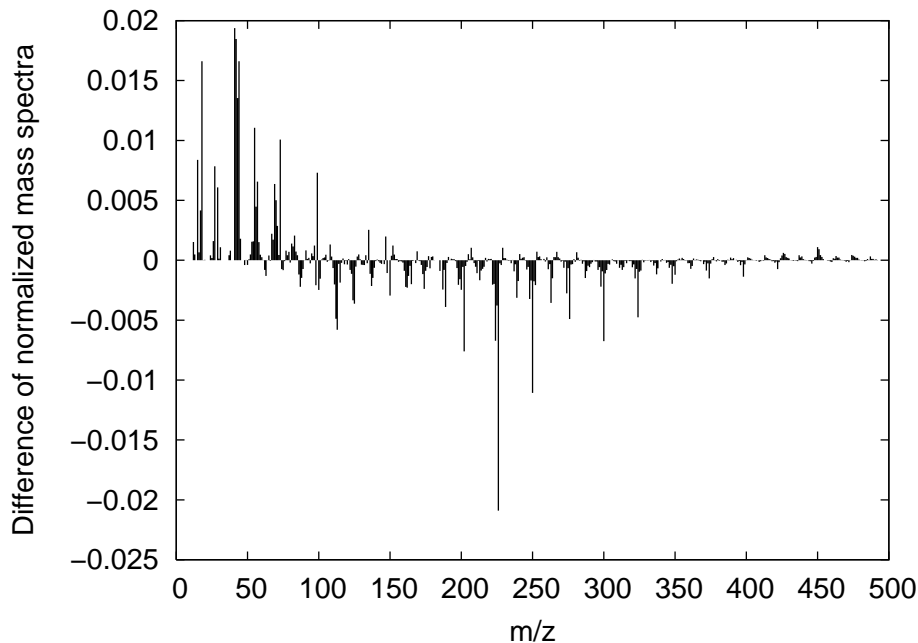


Fig. 9. Difference mass spectrum of sulfuric acid coated CAST-soot (medium OC-content) and uncoated CAST-soot.

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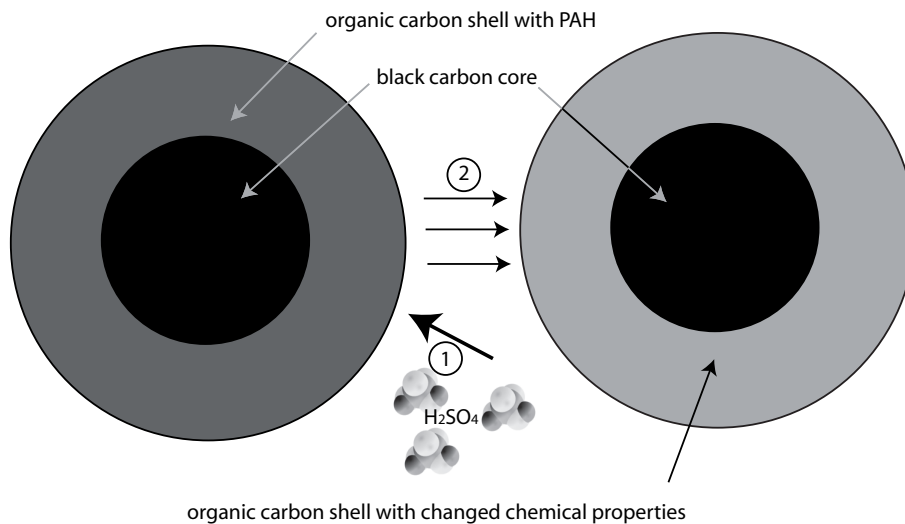


Fig. 10. Illustration for reactive uptake of sulfuric acid and change of chemical composition of the OC due to the reaction of sulfuric acid and PAHs.