

Interactive comment on “Hygroscopic growth and droplet activation of soot particles: uncoated, succinic or sulfuric acid coated” by S. Henning et al.

S. Henning et al.

silvia.henning@tropos.de

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General comments:

I have some serious doubts on the usefulness of using LACIS for measurement of soot particles. How accurate are these measurements for particles that are nonspherical and where the effective refractive index is not known a priori? In my opinion this is a very important and critical point for the interpretation of the LACIS measurements in this paper. With the HTDMA you show that the soot particles can shrink which is a clear indication that they are not spherical. For a given refractive index and fixed volume equivalent diameter the measured optical diameters are dependent on the particles

C15967

morphology, fractal dimension, and the primary particle size. I assume that the growth curves from the LACIS are calculated for spherical particles with a certain (clearly specified?) refractive index. Please discuss carefully these assumptions, limitations and uncertainties and add realistic vertical error bars to the graphs. Would this explain the encountered differences in Fig 7?

See also our answer(s) below. We should say clearly that to calculate the size of the grown particles the layered sphere model has been used, which at least accounts for the absorption in the soot core. It can be a source for a systematic error for extremely thin water layers on soot, since the actual aggregate morphology and hence the effective refractive index is not known. This might be an explanation for the "LACIS min OC" case in the figure 7.

– The paragraph dealing with the retrieval of the wet diameter was rewritten, see below

In line 256 it is stated that (except for the sulfuric acid coated soot) all data lies within the error bars. However, only the uncertainty in the RH measurement is considered but not the (much larger) uncertainty in the measured diameters. The disagreement observed with the sulfuric acid coated soot could arise in this case from the fact that the morphology is different.

OK,

The uncertainty in the measured diameters is large only for dry or coated soot particles. As soon as the water layer starts covering the insoluble soot core, the layered sphere model starts being valid and the uncertainty goes back to the level of 10% to 15% (known from the PSL calibration)

AND YES, Morphology can be important but we have no chance to evaluate how much.

– no changes in the text

Another important criticism is that I do not agree with the explanation presented in Figure 6 (hygroscopicity suppressed by the coating). I believe it is wrong. If it were true, it would mean that the coating is not water-soluble and that more residence time in the instruments would be needed to reach equilibrium. Instead, I believe that the observa-

C15968

tions can be explained by the nonspherical nature of the particles. See Tritscher et al. (2011).

If the reviewer would be right in this point, we would face this problem in all measurements due to consistency of the measurement set-up. But this is not the case: for CAST soot coated with a stronger acid namely sulfuric acid, the particles became more hygroscopic. This together with the PAH data from the AMS, lead us to the conclusion that the PAH are not destroyed by the weak acid (succinic acid) but by the sulfuric acid. However, the illustrating figure 6 seemed to be misleading to the reader and was therefore removed.

– no changes in the text, but figure 6 was removed

Specific comments:

An overview of the most recent literature is missing in the introduction. Many statements and numbers are referenced to Horvath (1993). There is a lot of newer literature on the interaction of soot particles with water and I strongly recommend rewriting the introduction and referring to the recent literature.

We did cite Horvath in connection with the main sources of soot and not concerning the interaction with water. We agree with the referee that there is newer literature available. We removed the Horvath citation and we rewrote the affected paragraphs. Text changed to:

– "The annual emission of BC is estimated to 8.0 Tg for BC and 33.9 Tg for OC (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), with the largest source of both being biomass burning (42% of BC and 74% of OC). The remaining fraction of BC emissions is dominated by transport, industry, and the residential sector with 20%, 10% and 25%, respectively. Besides biomass burning, the emissions from residential solid fuels contribute nearly 20% and transport 4% to the global budget of OC (Bond et al., 2004). The residence time of soot particles in the atmosphere is about seven days and 98% of the soot particles are removed by precipitation (Jacobson, 2004). That

C15969

is long enough to transport the particles hundreds to thousands of kilometers, but too short for an interhemispheric transport (Horvath, 1993). During transport in the atmosphere soot particles undergo aging processes, e.g. due to condensation, which alters their cloud forming potential (Andreae and Rosenfeld (2008) and references therein)."

Line 40: are these percentages in terms of mass or number?

Mass, number or volume percentages was not specified in Horvath 1993, but we removed that citation.

Line 52: "by nature soot particles are hydrophobic and insoluble". That's not generally true. For example, fuel sulfur content matters. Ship emissions (a very important BC source worldwide and not mentioned in the introduction) emit soot particles which are hygroscopic. Care should be taken with the term hydrophobic: "Pure" uncoated soot particle can be non-hygroscopic but it's unlikely that they are hydrophobic.

The statement was re-written in the following way:

– "During transport in the atmosphere soot particles undergo aging processes, e.g. due to condensation, which alter their cloud forming potential (Andreae and Rosenfeld (2008) and references therein)."

Line 59: specify RH. Giving a GF without RH makes limited sense.

Statement was clarified:

– "....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 1.19 (RH=95%, $d_{p,0}=50\text{nm}$)."

Line 73 and 11: Whether a particle "acts as CCN" depends on the SS. Same for "CCN activation". If SS is high enough all particles will act as CCN. Always indicate the SS when you discuss whether a particle will act as CCN or not.

Was corrected.

Line 90 et seq.: I did not understand the role of the NAUA and AIDA chamber. How much was the aerosol diluted? How long were the particles aged before measure-

C15970

ment? Did you observe an influence of aging on measured properties, e.g. evaporation of coating due to the dilution? Coagulation of agglomerates is expected to change the fractal dimension and these morphology changes are expected in turn to influence hygroscopic properties.

The NAUA chamber was used as an aerosol reservoir for continuous sampling with the hygroscopic growth and CCN instruments. This is directly stated in the experimental set-up section. The residence time of the aerosol in the NAUA chamber ranged depending on the experiment between 2 and 9 hours. Part of the aerosol was transferred to the AIDA chamber in order to investigate the cloud activation and ice nucleation behavior of the same aerosol. These measurements are discussed elsewhere.

The soot aerosol was diluted before entering the coating section 1/100 by means of two PALAS dilution systems (VKL 10, PALAS GmbH, Germany). The coated aerosol was fed into the NAUA chamber until a number concentration of around 10^4 to 10^5 cm^{-3} was reached. The coagulation alone would not change the fractal dimension of the soot particles (see e.g. Wentzel et al, 2003). We measured size selected and within the measurement time we did not observe an influence of ageing on the aerosol properties. For the measurements presented here we did not observe evaporation of the coating substances.

– We added in the experimental set-up section:

"The residence time of the aerosol in the NAUA chamber ranged depending on the experiment between 2 and 9 hours."

We added in the particle generation section:

"The soot aerosol was diluted before entering the coating section 1/100 by means of two PALAS dilution systems (VKL 10, PALAS GmbH, Germany)."

Line 127: ". . .succinic acid was as an example for oxygenated organics substance. . .". This compound is not a good choice for a representative model substance because it takes up no water at RH<99%. This deliquescence RH is high compared to most particle bound organics found in our atmosphere. Mention the hygroscopic properties (growth factors, deliquescence point) of succinic acid in the introduction.

C15971

We chose succinic acid because it represents a large fraction of the water soluble organic carbon (WSOC) found in atmospheric particles [Decesari et al., 2006; Martin et al., 2010; Saxena and Hildemann, 1996] and is one of the predominant OC forming CCN [Sun and Ariya, 2006 and references therein]. Beside, it is manageable under lab conditions unlike e.g. oxalic acid, which would evaporate from the particles due to its high vapor pressure. The hygroscopic properties of succinic acid are mentioned in the text.

– no changes in the text

Line 136 et seq. and 185 et seq. Mention that the LACIS and CCNC were operated downstream of a DMA. Did you employ a drier in front of the instruments?

Yes, we applied a diffusion drier in front of all instruments. We added this information also in the manuscript in the experimental section:

– "The aerosol in the NAUA was polydisperse, but all measurements presented here were done for diffusion dried size-selected particles."

Line 146: How can you be sure that two seconds are sufficient to reach equilibrium? Depending on the aerosol this might not be sufficient. What are the corresponding residence times in the two TDMA's and the CCNC?

We are confident that two seconds residence time is sufficient to reach equilibrium. We calculated the equilibrium time for the most critical scenarios (sulfuric acid, high RH) with the CFD code FLUENT (ANSYS Inc., Canonsburg, PA, USA) together with the Fine Particle Model (FPM; Particle Dynamics GmbH, Leipzig, Germany). The equilibration time for pure H_2SO_4 is 0.3 s and for a H_2SO_4 coated particle less than 0.2 s ($T=20^\circ\text{C}$, $\text{RH}=98.4\%$). The residence times of the TDMA's are 20 s for the Jülich instrument (Lang-Yone et al., ACP, 2010, p. 7256) and 12 s for the Clermont instrument. In the CCNC instrument the duration of the particles exposure to the maximum supersaturation in the column is about 2 s (Snider 2010).

– The residence times were added in the instruments descriptions.

VHTDMA

C15972

"The aerosol flow can be led through both sections or bypass either one, resulting in either volatility or hygroscopicity or combined volatility/hygroscopicity measurements. The residence time in the conditioned section is 12 s."

HTDMA Jülich

"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 which they were exposed for approximately 20 s (Lang-Yona et al., 2010)."

CCNC

"The exposure time to the maximum supersaturation in the column is about 2 s (Snider et al., 2010)."

Line 150: indicate wavelength as refractive indices are wavelength dependent.

The OPC (WOPS) is a white light optical particle counter. The spectral transfer function of the instrument is located between 350nm and 800nm and has a maximum around 550nm. The calculation of the optical response function takes the wavelength dependence of refractive indices of the particle materials into account where necessary. Most of the particle materials (water, black carbon, ammonium sulphate, succinic acid, sulphuric acid) do not exhibit strong variation of the refractive index within the visible wavelength range (being dielectrics). Therefore, it is sufficient to use a single value of refractive index corresponding to 550 nm.

– The paragraph dealing with the retrieval of the wet diameter was rewritten, see below

Line 151: for dry soot a refractive index of $1.74+0.65i$ was used. The diameter of "wet particles" is retrieved with $n=1.33$. How do you know if a particle is wet or dry? Which criterion did you apply? Which refractive index is used for a particle which experiences restructuring?

We agree that the retrieval of droplet and aerosol particle size from the optical measurements should have been described more thoroughly. The following paragraph has been added to the section 2.3 of the manuscript:

"At the end of the flow tube, the White-light Optical Particle Spectrometer (WOPS)

C15973

measures the size of the grown particles (Kiselev et al., 2005). WOPS uses the scattering of the broad band light (350 to 800 nm) and the wide range of scattering angles to avoid the ambiguity of size determination (the so-called Mie oscillations). The size of the single particles crossing the illuminating beam is retrieved from the response characteristic, which is calculated with the Mie theory for a multi-layered sphere (C. F. Bohren and D. R. Huffmann, "Absorption and Scattering of Light by Small Particles", Wiley, New York, 1983) with account for size and complex refractive index of the dry aerosol, amount of soluble material (if known) and the thickness of water shell condensing on the particle. To calculate a response characteristic, the size of the non-soluble core (soot) is kept fixed and the size of the droplet is changing by condensation of a water shell. For the water shell, the refractive index was calculated as a volume proportional mixture of water and soluble material. The effective refractive index of light absorbing soot core could be calculated using Maxwell-Garnett approach for porous media (Dobbins, R. A., and C. M. Megaridis, "Absorption and Scattering of Light by Polydisperse Aggregates", Appl. Optics, 30, 4747-4754, 1991) assuming the characteristic size of primary particles, number of primary particles per agglomerate (which is known from the mobility diameter measurement) and assuming that a soot aggregate undergoes incomplete restructuring while being immersed into a water droplet. The calculated response curve is then corrected using the calibration of the instrument with PSL microspheres (Duke Scientific).

Such calculations have been carried out for several test cases where aggregate size, fractal dimension, number and size of primary particles, degree of restructuring by immersion and amount of soluble coating have been varied within realistic limits. We found, that due to the broad spectrum of illuminating light and wide range of scattering angles used for the size measurement in WOPS, the influence of soot core morphology and presence of the soluble coating have a negligible effect on the particle optical properties as soon as the particle has acquired a water shell. Therefore, we have abandoned the fractal formalism of the soot core aggregate replacing it with the simplified model of two-layers sphere. For the complex refractive index of soot core the value of

C15974

1.74+0.65i has been assumed, while the refractive index of water shell has been kept at 1.33."

Line 156: LACIS calibration with ammonium sulfate: What exactly is calibrated? The size selection of the DMA, the optical sensor or the RH measurement? Which theory describing the water uptake of ammonium sulfate did you use?

The description of the calibration was improved as follows:

– The relative humidity (RH) in LACIS-mobile was calibrated as described in detail in Wex et al.(2005). Briefly, ammonium sulfate particles of known dry sizes (size-selected by a DMA) were sent through the flow tube for a certain fixed wall temperature. The grown particle size at the end of the flow tube was detected with the WOPS for several set dew point temperatures. The wet particle diameters were determined applying a refractive index following the volume mixing rule for the diluted ammonium sulfate particles. For the so determined wet diameter the related RH was calculated via Köhler theory applying the water activity after Young and Warren (1992) and for a droplet molality above 0.8 mol/kg a fit to the data of Low (1969).

Line 162 et seq.: Which inversion was applied to the TDMA data to retrieve GF?

Ideal case: wet HGF distributions are mode-fitted with one or more modes. More than one mode was retrieved if the standard deviation of the HGF distribution was larger than during the dry scans (representation of the transfer function of both DMAs). In this case: just a mode fitting was applied and the modal HGF used as an average.

– We added in the VHTDMA section:

"The size of the grown particles was determined applying a mode fitting routine."

Line 178: It's described that the VHTDMA is able to measure the particles volatility. Could this be used to derive information on the (size dependent) coating thickness?

Yes, ideally the information could be used. However, the VHTDMA was run with a volatilization temperature of 200 °C and part of the soot also volatilized at this temperature (for the high OC soot at least), so the sulfuric acid coating thickness could not be retrieved.

C15975

– no changes in the text

Line 184: How can you calibrate a humidity sensor with ammonium sulfate particles?

Statement was rewritten as follows:

"The RH in the VHTDMA instrument was calibrated analogously to LACIS-mobile with ammonium sulfate particles (see above). Capacitance humidity sensors (Rotronic, HygroClip SC05) are applied to monitor the RH."

Line 196: Mention the total particle counter which is needed to calculate the AF.

The information was added in the CCNC section, also in Figure 1.

– "In parallel to the CCNC a total particle counter was run, so that the activated particle fraction (AF) can be calculated by dividing the activated by the total particle number."

Line 201: Its good that you calibrate you instrument frequently. But it's also important to report the encountered variability from calibration to calibration because it contains information on the instrumental stability (drifts). This comment applies also to the other instrument calibrations.

The frequent calibrations were of course used for the interpretation of the instruments stability. In the calculation of the effective supersaturation in the CCNC a mean value from the calibration was applied as the instrument worked very stable with variability below 5% relative (for 0.1 of +-0.005% absolute and for a supersaturation above 0.1% +- 3% of the set value). In the data analysis the mean value of the calibration was applied.

– "Ammonium sulfate particles were used to calibrate the supersaturation reached in the CCNC (Snider et al., 2010). Calibration measurements were repeated at least every fourth day. The instrument worked very stable with variability below 5% relative. In the data analysis the mean value of the calibration was applied."

Line 204: The AMS was used to "characterize the coatings". Please give more information, be more specific. Does it allow for a chemical and physical characterization? Together with some BC mass concentration measurements one could derive the OC

C15976

fraction.

Quantitative measurements of non-refractory coatings on a refractory and corrugated material like soot are difficult and require extra measures and calibrations e.g. to exclude matrix effects (compare Reitz et al., ACP, 7839-7858, 2011). We calibrated the HR-AMS only in the usual way with Ammonium Nitrate and PSLs and applied standardized evaluation procedures. Nevertheless, in the few cases mentioned in the manuscript the AMS observations helped to elucidate differences in the composition of the coated particles and helped to explain their microphysical behavior. The AMS results will thus be only used in qualitative fashion as given in the manuscript. We prefer not to include any further discussion of the AMS results in this manuscript.

– We changed the statement in the manuscript to:

“A HR-TOF-AMS (Aerodyne Research Inc.) connected to the NAUA chamber was used to qualitatively characterize the coatings. The AMS is described in detail in Jayne et al. (2000), and DeCarlo et al. (2006). Briefly the AMS allows the mass spectrometric on-line investigation of aerosol particle composition after substantial reduction of the gas phase. The AMS was calibrated for ionization efficiency with NH_4NO_3 aerosol and particle size calibrated using polystyrol latex (PSL, Duke Scientific Corporation, Palo Alto CA) standards of different sizes between 81 nm and 596 nm. For evaluation of the mass spectra the standard software package Squirrel provided by Aerodyne Research Inc. was applied “

Line 214: I expect that the observed shrinking in Fig. 2 is dependent on the coating thickness and on the aging time in the chamber. In addition, it's known that this shrinking is strongly dependent on particle size. Please provide more information.

The data presented in Figure 2 are given for pure soot generated with the GFG - only the carrier gas differs (either Argon or Nitrogen). This information is given in text and figure caption. A coating thickness is for this reason not given. We did not observe an influence of aging time on the particle properties.

– The investigated diameter dp is stated in the text, but was now also added in the figure caption.

C15977

Line 216: “Activation was not observed for the different cast soot types...”. So what? This information is not very useful because activation strongly depends on particle size. Report the dry size and discuss this activation with respect to the corresponding Kelvin diameter. I propose to report and compare e.g. apparent kappa values (a parameterization of the particles hygroscopicity) derived from CCNC and HTDMA as done in Tritscher et al. (2011). Looking at Table 2 this should be done for all experiments. If no growth is observed with the HTDMA, the apparent kappa at subsaturation is =0. A complication arises for nonspherical particles: the mobility diameters selected by the DMA are generally larger than the corresponding diameter which is needed for calculating kappa.

Here the reviewer is contradicting him-/herself concerning the role of kappa. We prefer to omit a discussion of the kappa value, as we can not recognise an advantage in the interpretation of a value, the calculation of which requires further assumptions.

– no changes in the text

Fig. 3 and Fig. 5: This is a follow-up remark on the request for more physical parameterization of the hygroscopic behavior. For some activation curves the 50% activation point can be derived. E.g.: Can the observed shift in Fig 3 (filled points) be explained by the Kelvin effect or is it due to size dependent chemistry or morphology?

The observed shift could be due to the Kelvin effect. However, neither morphology nor size dependent chemistry can be excluded completely, as the measured and calculated values lie within the accuracy of the CCNC instrument (5% relative).

– We added in the discussion of figure 3:

“With increasing particle diameter the activation curves did shift to lower supersaturation. The observed shift could be due to the Kelvin effect. However, neither morphology nor size dependent chemistry can be excluded completely, as the measured and calculated critical supersaturation values lie within the accuracy of the CCNC instrument (5% relative).”

Fig. 4: The presented difference can either be caused by different coating thickness

C15978

or by different morphology. I suspect the 2nd reason because the GFG soot is more “fluffy” (lower fractal dimension, smaller primary particle size) compared to the CAST soot. Can you confirm this hypothesis with ESM or SEM pictures?

Morphology can be important but for different reason than suggested by the reviewer: The fractal dimension of GFG soot is, in fact, HIGHER than that of CAST-soot: $D_f = 2.0$ compared to 1.8 which is normally assumed for CAST soot, even though GFG soot aggregates appear more “fluffy”. This is not the only difference between GFG- and CAST-generated soot: GFG aggregates comprise larger number of smaller primary particles (mean diameter 6 to 8 nm, see Kiselev et al. JGR-A 2010, compared to 20 nm and more for CAST soot primary particles, see Malik et al., Aerosol Sci. Tech., 2011). Also so-called mean coordination number (the mean number of interparticle joints per primary particle, see for example Brasil et al., J. Aerosol Sci., 2001) is higher for GFG soot than for CAST soot (as implied by the higher fractal dimension) and therefore GFG soot is more porous and thus offers more space for capillary condensation under given RH. One have to consider also that primary particles in a GFG agglomerate are smaller (see above) and therefore the contact area between the primary particles is also smaller than between larger primary particles within a CAST soot aggregate. That means, that the mechanical strength of such a joint is lower and therefore the GFG soot agglomerates are more inclined to restructure under the action of capillary forces. Another point to consider:

the soot aggregate of both types (GFG and CAST) would most likely compact already during the coating (Stratmann et al., JGR-A, 2010, Kiselev et al., JGR-A, 2010) same mobility diameter an average GFG soot aggregate would contain much more primary particles compared to CAST; therefore at the stage of coating the GFG would compact only partially leaving some space for the further compaction during the hygroscopic growth, whereas the smaller and very chain-like CAST soot aggregates would collapse to the bulk agglomerated globes already at the coating stage. This point is indirectly supported by the observation that the GFG soot did not show total collapse even after condensation of propanol (see Kiselev et al., JGR-A, 2010). This would explain why no

C15979

shrinking of the CAST soot has been observed.

The review of the possible morphology effects as given above shows that it is not possible to take all these effects into account quantitatively without providing additional structural information (TEM / SEM before and after coating). Therefore the major quantitative merit of these experiments can be gained in those cases, where soot aggregates have shown detectable hygroscopic growth or activation, because in these cases the water solute shell around the soot aggregates dominates their properties.

– no changes in the text

Fig. 3 and Fig. 5: I assume that the presented activation curves are less steep than the curves for a pure model aerosol (like ammonium sulfate). Can this be explained by a variability of the composition (mixing state) of the soot particles? An analogous broadening of the growth spectra should also be seen by the HTDMA.

In deed, the activation curves are less steep than for a pure substance such as ammonium sulfate, the broadening of the step might be due to the variability in the particles composition. However, a better quantification of the effect is beyond the scope of this paper.

– no changes in the text

Figure 5: Why did you add a ~ in front of the mobility diameter in the legend?

Diameter was taken from diameter scan and d_p is for this reason not exactly 150, 175 or 200nm. We state now the precise diameter in the figure.

Table 2: With the HTDMA the hygroscopic growth is classified into “not observed, shrinking”, “not observed” and a growth which can be quantified. Can this classification be quantified a bit more? When is a behavior classified as shrinking? Could it be that a particle undergoes a certain shrinking and simultaneously takes up water and is therefore classified as “not observed”?

In table 2 the hygroscopic growth as quantified with LACIS-mobile at the highest set relative humidity of 98.4% is given. This is stated in the caption. However, for the GFG

C15980

soot shrinking was observed with the HTDMA instruments, but not with LACIS. For this reason the statement shrinking (HTDMA) was added. We clarified this in the table by adding HTDMA in parentheses after "shrinking".

– We changed the caption to:

"The growth factor (GF) of 250 nm particles at 98.4% RH is given for LACIS-mobile, if not mentioned otherwise."

Table 2, 3rd experiment: 12+x%; same for minicast experiment: 78+x%.

The x in table 2 stands for the additional, but not quantified, OC fraction due to the succinic acid coating. We clarified this with a footnote in the table.

– **** x stands for the additional, but not quantified, OC fraction due to the succinic acid coating.*

Line 254: "instrumental error bars": In the figures, only horizontal error bars representing the uncertainty in the RH measurement are presented. I guess that the main uncertainty in the presented data arises from errors in vertical direction (uncertainties and bias in calculating activated fractions and growth factors). Please add these as well.

The error bars in GF and water activity are given, however they are small.

– no changes in the text

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C15981

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C15982