Atmospheric
Chemistry
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Discussions

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Interactive comment on "Modelling non-equilibrium secondary organic aerosol formation and evaporation with the aerosol dynamics, gas- and particle-phase chemistry kinetic multi-layer model ADCHAM" *by* P. Roldin et al.

P. Roldin et al.

pontus.roldin@nuclear.lu.se

Thank you for the relevant and constructive comments on the manuscript. They definitely will help us to improve it.

First of all we want to mention that in the revised manuscript we will change the formulation "particle surface layer" to "particle surface-bulk layer" when we refer to the

monolayer thick particle layer at the surfaces where the condensing molecules dissolves. And if we consider reversible adsorption we will always call this layer "sorption layer" and nothing else. When we model the condensation and dissolution of organic compounds into the particle surface-bulk layer we will change the formulation "surface mass accommodation coefficient" to "surface-bulk accommodation coefficient" which considers both the potentially non-unity probability of adsorption (sticking) and dissolution into the particle surface-bulk layer.

Answers to comments:

In the introduction and conclusion the authors promote the utility of ADCHAM for better understanding poorly constrained processes and their influence on SOA formation, and ultimately the representation of SOA formation in three-dimensional models. There is a generally recognized sentiment in the field that with advanced measurement and modeling techniques, we've moved beyond the need to accurately represent SOA mass concentrations and now need to accurately represent SOA mass concentrations, temporal behavior, and properties (and we need to "get it right for the right reasons"). While with each model application there was an effort to test sensitivity to various assumptions, given the significant uncertainty associated with so many of the model parameters, further sensitivity analyses is highly desired. While model validation is important, beyond being able to represent observations, a model such as ADCHAM could really provide insight into which of the model parameters need to be better constrained (and which processes need to be better understood). It is strongly suggested that the authors consider splitting this manuscript into one or more parts: one in which the model is generally described and sensitivity is assessed, and one or more describing application and validation. Given that this is the first publication on this complete version of ADCHAM, there were places where the more general and broader description of ADCHAM was confused with the specific applications described. Three examples: 1) p. 777, line 15 the authors describe the ADCHAM schematic and oxidation of SOA by O3. It isn't clear here why heterogeneous/bulk oxidation by only O3 is considered

and not OH for example. After reading further it is clear that ADCHAM could consider particle-phase OH reactions, but for the specific applications this is not relevant. And, 2) While the ADCHAM model can treat oligomerization, in this application only dimerization was considered. The authors need to clarify this point and remove discussion of oligomerization in discussion of results as necessary (e.g., p. 795, lines 13-14). The blurring between model capabilities and the specific applications could be remedied by separating the publication into one or more parts. It is the opinion of this reviewer that doing so would increase distribution and citations, and greatly enhance the contribution of this work to the field.

We prefer to keep the model description and applications as one paper because we think that the applications make the reader more prone to also read more about the model and to understand how it works. The KM-GAP model description and application paper is structured is a similar way as our paper (Shiraiwa et al., Atmos. Chem. Phys., 12, 2777–2794, doi:10.5194/acp-12-2777-2012, 2012).

We agree that there are places where the more general and broader description of ADCHAM is confused with the specific applications described. As pointed out by M. Shiraiwa a table which summarize the assumptions, processes and different parameter values used for the specific model applications will help to differentiate between the general model description and the specific applications.

Sections 2.4.2 which is called "O3 diffusion and reactions with SOA" will be named "Diffusion of oxidation agents and reactions with SOA" instead. In the text and all equations in this section we will replace O3 with Zox that represent any kind of oxidation agent (O3, OH, NO2, NO3).

On p. 777, line 15 we will change from: "oxidation of SOA with O3" to the more general "heterogeneous SOA oxidation".

Yes, we will clarify that we in this manuscript only consider dimerization but that ADCHAM in principle could consider oligomerization too. We will remove the discussion

of oligomerization in the results and discussion sections. In Sect. 2.3.4, P789, L9-11 we write:

"Currently ADCHAM only considers the reactions between monomers which form dimers and not the possible reactions between dimers and dimers with monomers."

We will change this formulation to:

"For the applications in this article, we only consider the reactions between monomers which form dimers and not the possible reactions between dimers and dimers with monomers."

pp. 773-774, discussion of equilibrium: While the authors note on p. 774, line 7-10 that there are a number of possible reasons why predicted evaporation does not agree with measured evaporation, the preceding discussion emphasizes (perhaps erroneously) only the mass transfer limitations. It is important to clarify that the Dzepina et al. (2009) and Vaden et al. (2011) predictions are for some specific assumed product distribution, which both lack a significant fraction of low-volatility material. The authors suggest that particles will never be in equilibrium with the gas phase: : :doesn't this depend on the temporal and spatial scale of interest? What about the findings of Saleh et al., ES&T, 2013?

On P774, L3-8 we write: "In well controlled laboratory experiments Grieshop et al. (2007) and Vaden et al. (2011) have illustrated that the evaporation of SOA particles formed from α-pinene ozonolysis is a slow process (hours to days). Vaden et al. (2011) showed that this is orders of magnitude slower than expected from equilibrium partitioning. The slow evaporation of SOA can be due to mixing effects, mass transfer limitations, presence of oligomers (Grieshop et al., 2007 and Vaden et al., 2011)."

We, agree that the second sentence here is a too strong statement. We will change the text to:

"In well controlled laboratory experiments Grieshop et al. (2007) and Vaden et al.

(2011) have illustrated that the evaporation of SOA particles formed from α-pinene ozonolysis is a slow process (hours to days). Vaden et al. (2011) showed that this is orders of magnitude slower than expected from the 7-product VBS parameterization from Pathak et al., 2007, which lack substantial fraction of low-volatile material. The slow evaporation of SOA can be due to presence of low-volatile oligomers in combination with mass transfer limitations and mixing effects (Grieshop et al., 2007 and Vaden et al., 2011)."

We are aware of that without any low-volatile compounds in the particle phase, solid-like particles will also evaporate rapidly (as illustrated in Fig. 9).

On P773, L27-P774, L1 we write: "However, in the atmosphere the aerosol is exposed to much more variable concentration, temperature and humidity conditions. "Hence, the atmospheric aerosol will never be entirely in equilibrium with the gas phase."

We don't think this a too strong statement to say. It is also important to consider that the size of the particles also influence the saturation vapour pressure above their surface. Hence, if the large particles are in equilibrium with the gas-phase the smallest particles will evaporate.

We will change this sentence to:

"However, in the atmosphere the aerosol particles span over a broad size range and is exposed to much more variable concentration, temperature and humidity conditions. Hence, the atmospheric aerosol particles will never be entirely in equilibrium with the gas phase."

We will mention that Saleh et al., ES&T, 2013 did not observe a strong evaporation inhibition because of diffusion limitations in the particle phase of α-pinene SOA particles at low RH. But we will also write that this may be because in Saleh et al., 2013 only a relatively small fraction (10-30 %) of the SOA particle mass was evaporated before equilibrium was reached. Hence, low-volatile bulk-phase oligomers may not have

formed a complete monolayer thick surface layer coverage, which inhibit further evaporation. Although, the evaporation experiments in Saleh et al., 2013 are atmospherically relevant, the atmosphere is more complex and the particles may be exposed to temperature and concentration fluctuations during several episodes which eventually lead to accumulation of low-volatile viscous oligomer SOA at the particle surface. Additionally, traditional gas-particle partitioning cannot explain the observed nearly size independent evaporation of SOA particle observed by Vaden et al. (2011). To our knowledge Saleh et al. (2013) do not present experimental evidence for size dependent evaporation of SOA particles.

pp. 775-776, discussion of development of ADCHEM from ADCHAM should be moved to the conclusion.

Yes, we agree. We will move this text to the conclusions.

p. 776, the list of model applications here does not match the list provided in the abstract. 4 applications should be listed in the abstract, and mentioned first sentence last paragraph?

Yes, we will also list the DOP evaporation experiments in the abstract and introduction.

Model description: ADCHAM "consists of", add "s"

Thank you, we will do that.

Section 2.2.1, are the molar growth rates calculated iteratively? If not, how does this affect predictions?

No the molar growth rates are not calculated iteratively, because we consider condensation and evaporation as a dynamic process and do not assume equilibrium partitioning. The exception is the uptake of NH3 into the particle surface-bulk layer, which is treated as an equilibrium process after the condensation of all other compounds, according to the method developed by Jacobson Aerosol Sci. Technol., 39, 92–103, 2005. We will add a reference to Jacobson, 2005. To treat the non-equilibrium gas-

particle partitioning of many different organic and inorganic compounds require that the model takes short time steps. For each model application we have tested how short the time steps need to be in order to get results which are not biased by the length of the time step. For condensation and evaporation we use an internal time step of 1-10 ms (see P777, L21-22).

In section 2.2.1 and throughout the paper the discussion of the surface layer is somewhat confusing. On p. 780, lines 7-8 the authors state that compound i "partitions into (dissolves)" in the surface layer. However, on lines 12-13 the authors discuss the adsorption of oligomers at the surface. This is confusing throughout. Is the surface layer an absorption matrix for monomers but a solid for dimers?

Yes, we agree that this is sometimes confusing. We will go through the whole paper with this in mind, and make it easier for the reader to understand what we mean by "particle surface layer" (which will be called particle surface-bulk layer if we consider absorption and sorption layer if we consider adsorption).

We will also clearly state that in this paper we treat the gas-particle-partitioning of all organic compounds as an condensation/evaporation process and not as an reversible adsorption process followed by mixing into a particle surface-bulk layer. We will also clearly explain that ADCHAM in principle could treat the uptake of the organic compounds as a reversible adsorption process; analogues to the way the uptake of oxidation agents are treated. The main reasons why we treat the uptake of organic compounds as a condensation (dissolution) process and not a reversible adsorption process is that: (i) ADCHAM build on the atmospheric chemistry transport model AD-CHEM, which use Eq. 1 to consider condensation and dissolution into the particle phase, and (ii) reversible adsorption require knowledge about the desorption lifetime of the organic molecules, which to our knowledge generally is not well known.

On P780, L12-L15 we write:

"However, adsorption of low-volatility oligomers at the surface layer still affects the gas-

particle partitioning of monomer SOA, by altering the surface mass accommodation (Sect. 2.4.1) and by decreasing the monomer SOA fraction in the surface layer."

Based on the comments received from M. Shiraiwa we will no more assume that the surface-bulk accommodation is affected by the amount of oligomer (dimer) found in the particle surface-bulk layer. Hence, we will remove this sentence.

The particle surface-bulk layer is always an absorption matrix both for the monomers and the dimers.

p. 780, line 16: does setting a volatility limit on the saturation vapor pressures preclude consideration of more volatile species as reactants in dimerization? Please clarify.

Yes, in principle this could be the case if the more volatile compounds are adsorbed on a particle sorption layer and then react at the sorption layer and form dimers. We also tried to consider this by allowing pinonaldehyde to partitioning to the particle phase by a simplified reactive uptake mechanism (Eq. 33). Our idea was that pinonaldehyde adsorb to the sorption layer and rapidly reacts with other molecules and form dimers. However, according to M. Shiraiwa Eq. 33 is probably too simplified and he doubt that this kind of Eley-Rideal mechanism is applicable for aldehydes (the dimerization rate is probably too slow compared to the desorption-lifetime). Thus in the revised manuscript we will not consider this mechanism and instead only consider dimerization as a process occurring purely in the particle surface-bulk layer and the other particle bulk-layers (see comment by M. Shiraiwa concerning P790, L12).

If the compounds need to dissolve into the particle surface-bulk layer before it will have time to react and form dimers, then our volatility limit of 1 Pa will most likely not cause any strong limitations on the dimerization. This is because the concentrations (mole fractions) of these volatile organic compounds will be extremely low.

p. 781, lines 18-24: the discussion of the particle wall effects corrections and their influence on predicted mass seems reversed (or needs further clarification). With method

1, particles deposit onto wall and can continue to take up gas-phase species. Give that both particles and gas-phase constituents move to the walls, it seems like this would give a lower-bound estimate of mass formed (assuming mass formed is the total suspended in the bag, and does not consider that formed on walls). Maybe the authors are considering mass formed to be the total suspended+walls. A note here about terminology: technically, "aerosols" refers to the gas+particle phase. While "aerosols" is often used to refer to the particles themselves, it can be quite confusing (and in this case the authors appear to be using it to refer to the gas phase). The authors state that the particles "take up SOA" (line 15). I think the authors mean that the particles take up volatile/semivolatile gas-phase compounds (and not both particles+gases). See also p. 783, line 6.

Method 1 and 2 are only assumptions which are used in order to correct the measurements of SOA in the gas-phase and are not what is happening in reality. You are correct that if either method 1 or 2 would be true, the expected SOA concentration in the gas-phase would be higher in case 2 and lower in the case 1. However, the truth may be somewhere in-between case 1 and 2. If you then correct the data assuming that case 1 is true (method 1) you correct too much (overestimate the SOA formation) while with method 2 you underestimate the SOA formation.

To make it more clear that we referee to the measured SOA and nothing else when we talk about these correction methods we will change the sentence on L13-L15 from:

"A commonly used method (see e.g. Hildebrandt et al., 2009 and Loza et al., 2012) is to scale the formed SOA mass with the measured relative seed aerosol (typically ammonium sulphate) loss rate."

to

"A commonly used method (see e.g. Hildebrandt et al., 2009 and Loza et al., 2012) is to scale the measured SOA mass with the measured relative seed aerosol (typically ammonium sulphate) loss rate."

<div align="center">C1486</div>

We will change the sentence on L15-L17 from:

"With this method it is assumed that the particles deposited on the chamber walls continue to take up SOA as if they were still present in the gas phase."

to

"With this method it is assumed that the particles deposited on the chamber walls continue to take up condensable gas-phase compounds as if they were still present in the gas phase."

We will go through the manuscript and check that we always use "aerosol particles" if we referee to the particles and "aerosol" if we referee to particles+gas.

p. 782, line 15: sensitivity: : :how sensitive is the model to these uncertain parameters?

We will add a figure to the supplementary material where we vary the mean electrical field strength from 0-100 V/cm and the friction velocity from 0.01-1 m/s when we simulate the wall deposition losses experiment with ammonium sulphate seed aerosols in the 6 m3 Teflon chamber.

p. 786, line 22: "is" should be "are"

Thank you, we will change this.

p. 787, line 18-19: why are Henry's law constants used for the carboxylic acids? I can see how partitioning coefficients might be difficult to obtain for the inorganic compounds. However, even for those compounds it is important to consider the implications on modeling predictions. For example, it seems that NH3 would be much more soluble in water than in an organic+water phase (at low RH), and thus the influence of the NH3 uptake overestimated in the model applications. (e.g., as described on p. 802).

No, we do not use Henry's law constants for carboxylic acids but model their condensation uptake based on their estimated vapour pressures. We will change the sentence on P787, L18-L20 from:

<div align="center">C1487</div>

"Henry's law coefficients (KH) and dissociation rates (Ka) of the inorganic compounds and carboxylic acids, are (if at all) usually only available for aqueous solutions."

to:

"Henry's law coefficients (KH) of inorganic compounds and dissociation rates (Ka) of inorganic compounds and carboxylic acids, are (if at all) usually only available for aqueous solutions."

We will run sensitivity tests where we also consider that NH3 probably have a much lower solubility in the organic rich phase compared to pure water (e.g. by decreasing the Henry's law solubility coefficient with 1 or 2 orders of magnitude).

p. 789, line 11: Monomers restricted to of or below a specified volatility?

Yes, if the monomer vapour pressure is larger than 1 Pa we assume that it will not participate to the particle phase (see P780, L17). However, by default there are no other restrictions.

p. 789-790, discussion of oligomerization: What is the difference between the generic dimer formation (R7, R8) and the specified dimerization reactions (R1-R6)? Are R7 and R8 additional dimerization pathways? And if so, what is the justification of the additional pathways? If all dimer reactions are represented by R7 and R8, maybe it is better to list the specific reactions in a table? There have been a number of studies reporting on the thermodynamics and kinetics of the specific dimerization reactions considered and/or oligomer formation (e.g., Barsanti et al., AE, 2006; Walser et al., PCCP, 2008; Atkinson and Ziemann, Chem. Soc. Rev., 2012; DePalma et al., PCCP, 2013) that suggest that the kinetics/thermodynamics are not equally favorable. Thus the assumption that the dimers revert to monomers based on the relative abundance of the monomers (p. 791, line 15) does not seem justified (and is not necessarily "reasonable" as noted by the authors). At a minimum, some discussion or investigation of this assumption on the model results is needed.

C1488

No R7 and R8 are not additional dimerization pathways but the generic dimer formation rates which R1-R4 all belong to. We will remove R1-R4 and just discuss different types of particle phase dimerization mechanism with a reference to each.

We will add a discussion about the assumption about the dimers reverting to monomers based on the relative abundance of the monomers, why we do this, why it may not be correct (with added references as suggested above) and how the model potentially could treat this in a different way.

p. 791, line 20-27: sensitivity: : :how sensitive is the model to each of these factors? Which are the most certain/uncertain? Which of these need be better constrained?

All of these factors are uncertain and need to be better constrained. We will add a sentence which clarifies this.

The factors which have largest influence on the modelled oligomer content are the oligomerization formation and degradation rates.

On P787, L26-L29 we describe why the hydrogen ion concentration is uncertain and a motivation why we still use it and try to calculate the ammonia uptake and formation of organic salts.

On P791 we will add a sentence which clearly states that for the model applications in this work we do not use the hydrogen ion concentration to estimate the dimerization rates, and why we don't. This is because the dimerization rates are anyhow very uncertain and need to be fitted with sensitivity tests.

For the dimer formation rates we will add a reference to (Ziemann and Atkinson, Chem. Soc. Rev., 41, 6582-6605, 2012) where they discuss the range of reactions rates expected for several different oligomerization mechanisms, and compare the values we use with theirs.

For the oligomer degradation reaction rates we will discuss this according to the answer to your previous comment.

C1489

On P805, L12-L19 we refer to Barley and McFiggans, 2010 which has shown that different methods to calculate pure-liquid saturation vapour pressures can differ substantially. For that reason we use to different methods to calculate the pure-liquid saturation vapour pressures (SIMPOL and the methods from Nannoolal et al. (2008)) in Sect 3.2-3.4.

The diffusion coefficients of monomers and dimers are also very uncertain but there are at least two experimental studies of diffusion in SOA particles formed from α-pinene ozonolysis: Abramson et al. (2013) and Zhou et al., Faraday Discuss., 165, 391-406, (2013). The Abramson et al. (2013) study we have considered when we made our simulations but we will also consider the Zhou et al. (2013) study and do some sensitivity studies with diffusion coefficients in agreement with their study too. During the SOA formation phase in typical precursor oxidation experiments (e.g. α-pinene +O3) the mixing within the particles are not a factor which have strong influence on the monomer SOA formation and probably not the oligomerization either.

The ozone, uptake, diffusion and reaction rate with organic compounds are probably only important for the oligomer content if the SOA are composed of substantial amount of unsaturated organic compounds.

p. 799, line 19: "or" should be "and"

Thank you, we will change this

p. 801, line 20-25: It is not clear what the authors are trying to convey by the description of the observed RH effects by Kuwata and Martin, particularly starting with "However".

By however we convey that all NH3 uptake may not be attributed to diffusion of NH3 into the particles, but can be caused by the formation of additional organic salts from ammonia and the carboxylic acids initially found in the gas-phase.

We will remove however.

p. 803, line 14-16: What is the difference between ignoring the activity effects of the

C1490

ammonium salt on the organic compounds and treating it as a separate phase?

By ignoring the effect which ammonium salts have on the organic compounds but still allowing the other organic compounds to dissolve in the ammonium salt matrix, the ammonium salts will allow more of the other organic compounds to partition to the particle phase. While if the ammonium salts form a separate phase no other organic compounds will dissolve in this matrix and hence, they will instead limit the dissolution of other organic compounds into the particle surface-bulk layer by taking up part of the surface-bulk volume.

p. 807, lines 1-15: Could these results also be explained or be sensitive to characterization of wall effects?

For these model simulations we did not consider wall effects. We will add a new table which clearly describe which assumptions, parameters and processes that were used for the different simulations in Sect 3.1-3.4. But, yes the results could be sensitive to the wall effects but we do not known how the results could be explained by these effects. Where it something specific you were thinking of? The experiments by Na et al., 2007 were performed in a relatively large chamber of 18 m3, relatively high concentration and rapid oxidation of α-pinene, thus the chamber wall effects are probably not as influential as in the 6 m3 chamber which was used for the experiments simulated in Sect. 3.4.

p. 808, lines 1-8: Could the over-/underprediction be due to uncertainties in input parameters, missing compounds rather than suggestive of some non-ideal mixing? (not sure incomplete phase separation happens)

Yes, bearing in mind the many uncertain parameters this could also be the case. We will hedge a bit more and change from:

"This indicates that in reality, there will neither be perfect (ideal) mixing between NH4RCOO and the other condensable organic compounds, nor a complete phase

C1491

separation."

To

"This may indicate that in reality, there will neither be perfect (ideal) mixing between NH4RCOO and the other condensable organic compounds, nor a complete phase separation."

p. 809, line 1-2: Why weren't simulations with lower input concentrations performed? Isn't this the benefit of having a model?

Yes, you are correct. We will also run the model with more atmospheric relevant NH3 concentrations (e.g. 1 ppb NH3) and based on these results expand the discussion about the importance of ammonium-carboxylic acid salt formation in the atmosphere.

p. 813-814: Are the model assumptions and parameters (e.g., assumptions about oligomer coated surface, dimer formation/decomposition rates) consistent with chamber studies/reported values (e.g., previously listed references for dimerization/ oligomerization reactions)? The dimers considered in this work seem to be about the size/polarity of sesquiterpene oxidation monomers. Do such chamber studies provide support for the assumptions about dimers in the current work?

Based on the comments from M. Shiraiwa, which questioned our proposed mechanism and way of modelling the sorption and rapid dimerization of pinonaldehyde with other particle phase organic compounds, we will redo all model simulations where we did include this oligomerization mechanism (Eq. 33). Instead we will include a mechanism where a substantial fraction of the SOA is composed of short-lived dimers, which form relatively rapidly in the particle phase and then rapidly are degraded back to monomers again (within minutes).

We will consider the references which you suggested and compare our dimerization rates with the values reported by Ziemann and Atkinson (2012).

We are not really sure about which assumptions about dimers which you were thinking

of. Since you mention their molecular size maybe you think of their saturation vapour pressures and mixing within the amorphous semi-solid SOA phase. One should bear in mind that the dimers in our model runs are only a simplified representation of the oligomers that probably have a wide range of molar massed.

In the new table which we will add in the revised manuscript we will clearly write out which diffusion coefficients and vapour pressures (methods) which we use for the monomers and dimers, for all different model runs. We have run simulations where we test if the slow evaporation of the α-pinene particles from Vaden et al., 2011 could be described by the saturation vapour pressure driven dimer loss rates. However, as expected the model cannot capture the observed size independent evaporation rates. Thus we expect that at least some of the dimers (and oligomers in reality) are very low volatile and will not evaporate, but instead gradually chemically degrade to more volatile compounds which then can evaporate. The absolute values and relative difference in diffusion rates of dimers and monomers are also very uncertain. One way of estimating these are just to use the Stoke-Einstein relationship and the reported values of SOA viscosity. But then the relative difference in diffusion rates between the monomers and dimers are very small (see P818, L4). This was also addressed by M. Shiraiwa in one of his comments. The Stoke-Einstein relationship cannot explain the observed many order of magnitude larger diffusion rates of water and O3 in SOA compared to the diffusion rates of the organic compounds themselves. Thus, we expect that the diffusion rates of dimers (representing oligomers) are order of magnitudes slower than the monomers. In Shiraiwa et al., ACP, 12, 2777–2794, 2012 the model KM-GAP is used to simulate the oxidation and volatilization of oleic acid. In this work they estimate that the diffusion coefficient of the dimers is 2-4 orders of magnitude lower than for the monomers.

p. 832, line 17: "on" should be "of"

Thank you, we will change this.

Figure 4: Does the skill of model performance as a function of particle size give any insight into processes or model assumptions/parameters?

Yes, we think that information about size dependent growth or evaporation is very important to consider when drawing conclusions concerning which mechanism that are explaining the growth and evaporation of particles. E.g. if the evaporation is driven purely by the chemical degradation of low-volatile oligomers, and the chemical composition of the different particle sizes are identical, we would expect to see size independent mass loss rates. While if the evaporation is driven by the vapour pressures of the particle phase compounds we would expect the small particle to evaporate faster than the large ones. And if the particles are solid-like we would expect the evaporation rate to be limited by the least volatile organic compounds vapour pressures or chemical degradation rates, once they have accumulated and formed a monolayer thick surface-bulk layer.

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/14/C1478/2014/acpd-14-C1478-2014-supplement.pdf

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Interactive comment on Atmos. Chem. Phys. Discuss., 14, 769, 2014.

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