

Review of the paper «Evaluation of the sectional aerosol microphysics module SALSA implementation in ECHAM5-HAM aerosol climate model » by Bergman T et al.

This paper presents the integration of the sectional aerosol model SALSA in ECHAM5-HAM climate model and its evaluation.

English should be checked by a native English speaker. I corrected some mistakes, but there are too many. It is sometimes difficult to understand what the authors mean. The model presentation is confusing. For example, the separation between soluble and insoluble is not clear, as well as the partitioning of mass and number concentrations. Sea-salt emissions are calculated by using a radius at 80% humidity. This should be corrected to correctly account for variations in relative humidity for soluble particles. For wet deposition, dry deposition and sedimentation, it is not clear which diameter is used for each section. For soluble particles, the water uptake should be considered when computing the section diameter to be used for deposition. Sedimentation is very low compared to M7. It should be checked that this is not due to the numerical sedimentation limitation with the time step. In the evaluation, the authors are not always objective in the comparison. For example, in Fig 5, they compare models at one specific particle diameter (200nm), whereas the comparison would give opposite results if a diameter of 500nm was chosen. In Figure 6, one model seems to be better in some cases, while the other model seems to be better in other cases. Computing statistics would be a fairer way to decide which model is better. The model is evaluated by comparisons to M7 and observations. For surface concentrations, it would be valuable to compute metrics which are commonly used to evaluate models (see Boylan and Russell, 2006).

More detail on the meteorology should be given, and the length of the simulation should be given. I don't understand why there are large differences in meteorological fields depending on whether M7 or SALSA is used.

I recommend the article to be published if the authors can address the specific comments below.

### **Specific comments:**

#### Abstract

- L8: What do you mean by « 20 size sections with 10 size sections in size space »? What about the other 10 sections?
- L13: I do not understand what « modal » refers to here. Maybe better to remove it from this sentence.
- L14: replace « sea salt mass » by « sea-salt mass ».
- L15: What does AOD stand for? Please detail at the first occurrence.
- L22: Is it acceptable to have concentrations within a factor five of the observations?
- L22 and L23 are in contradiction. You first say that sea salt is within a factor of five of the observations, and then you say that sea-salt observations are reproduced less accurately (than sea salt!)
- L23: Replace « sea salt concentrations » by « sea-salt concentrations»
- L27: Replace « than the observed » by « than observed ».

#### Introduction

- L27, p3625: For examples of other modal models, you may add these 2 citations:  
Binkowski FS; Roselle SJ, Models-3 community multiscale air quality (CMAQ) model aerosol component. 1. Model description, JOURNAL OF GEOPHYSICAL RESEARCH-ATMOSPHERES Volume: 108 Issue: D6 Article Number: 4183 DOI: 10.1029/2001JD001409 Published: MAR 26 2003  
Sartelet KN; Hayami H; Albriet B; et al., Development and preliminary validation of a modal aerosol model for tropospheric chemistry: MAM. AEROSOL SCIENCE AND TECHNOLOGY Volume: 40 Issue: 2 Pages: 118-127 DOI: 10.1080/02786820500485948

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- L3, p3626: limitations of modal models related to the growth of particles are illustrated in Sartelet et al. (2006)
- L6, p3626: What do you mean by « separate retrieval »?
- L22-24, P3626: I do not agree. Sectional models do have a pre-defined assumption of the particle size-distribution shape: it is a section!
- L9-L11, P3628: Please rephrase. I could only understand after looking at the figure, I think the sentence is not clear enough (as well as the legend of the figure).
- L11, P3628: For clarity, please replace « external mixing » by « chemical composition »
- Figure 1. For clarity, it maybe better not to call « a, b, c » parallel sections but rather chemical composition. Then the legend of Figure 1 can be rewritten by something like « for each size section, particles are separated depending on their chemical composition, that is whether they are soluble, insoluble or insoluble with a soluble coating enabling cloud activation ».

Also, from the figure, it seems that the possible chemical components of particles of one subrange are detailed below the subrange. Is this correct? For example, may soluble particles contain dust or black carbon and may insoluble particles contain water soluble fraction?

- Table 1: For sections 3b3 and 3c3, there may be typos: the max diameter are probably 10 rather than 1
- L15, P3628: What do you mean by « external mixing of particles is limited to subranges 2 and 3 »? Is it that insoluble particles are not considered in the first subrange? If it is so, it is clearer to say it explicitly.
- Table 2: What is dry deposition not considered for 1a and 2a particles, if it is considered for others? The influence of dry deposition is minimum for particles of diameters about 1 micrometer, but the influence increases as the diameter increases and decreases.
- Table 3: What do you mean by « the number tracer »?
- L20-21 and L22-23 are in contradiction. In the third subrange, number and mass are considered according to L21. Then it says that mass is considered only for particles below 700nm. Do you mean above 700nm? Considering mass as a prognostic variable is crucial for large particles as they account for most of the mass of particles.
- L1, P3629: Can you detail which water soluble compounds you are referring to? Sulphate, sea salt are water soluble content, as can also be organic carbon.
- L6, P3629: Can't particles in subrange 1 coagulate with themselves? Is only coagulation with larger particles considered? Why?
- L13, P3629: Does the soluble bins include dust? If so, why? Because dust is not soluble. Does the insoluble bins included sulfate? If so, why? Because sulfate is soluble. Black carbon is also insoluble but it also goes into the soluble bins?
- L16, P3629: For clarity, replace « three parallel bins » by « three parallel chemical composition ».
- L18, P3629: What is soluble dust?! Is it dust mixed with water soluble compounds? What are these compounds? Why are they not considered for subrange 2?
- L20-21, P3629: in contradiction with L25-26 P3628.
- L6-7, L11, P3629. For clarity, you should consider moving all the processes description to section 2.4.
- L25-27, P3629: Why is intra-sectional coagulation omitted? Especially for nanoparticles, intra-sectional coagulation will make the section diameter grows, and it could be an important process. This should be discussed in the manuscript.
- L10, P3630: What organic compounds do you consider? Do they partition between the gas and the particulate phases?
- L19-21, P3630: Why us binary nucleation parameterisation not applied everywhere? How are the cases defined?
- L7-15, P3631 and L23-24, P3631: This part can be shortened. You don't need to detail the Lehtinen et al and the Antilla et al parameterisations here, as they are not used. You don't need

to detail the subject of future studies, that's to be done in the conclusion.

- L14-15, P3632: Is it possible to have cloud droplets and produce sulphate in-cloud without pre-existing particles? How are the cloud droplets formed?
- Paragraph 2.4.2: Why are heterogeneous reactions at the surface of particles not modeled? Some parameterisations such as the one of Jacob (2000).  
Jacob DJ, Heterogeneous chemistry and tropospheric ozone, *ATMOSPHERIC ENVIRONMENT* Volume: 34 Issue: 12-14 Pages: 2131-2159 DOI: 10.1016/S1352-2310(99)00462-8 Published: 2000
- L17-23, P3632: What is done in this study then to transfer insoluble sections to soluble sections?
- Table 1: Add a column with the mean diameter, as it is often used to transfer from number to mass
- L26, P3632 and L27, P3633: Why is 2a4 not transferred to 3a1 rather than 3b1 (soluble section should be transferred to soluble rather than insoluble section?). Why do you need to compute the number mixing ratio from the mass mixing ratio? Is it that the subranges 1 and 2 only have mass mixing ratio and the subrange 3 only number mixing ratio?
- L4, P3633: Why is 2b4 not transferred to 3c1?
- Table 4: What do you mean by « has been defined separately ». How were they defined?
- L5, P3635: For what kind of calculation do you refer to Kerkweg? Is it for more detailed model presentation?
- L12-15, P3635: For soluble mode, do you consider the water uptake of particles for computing the bin mean radius?
- L17-18, P3635: Rather than limiting the sedimentation velocity, why do you not limit the time step and subcycle the temporal resolution for sedimentation? If you keep the time step constant and limit the sedimentation velocity, wouldn't you sometimes under-estimate sedimentation?
- L20, P3635: It has been rewritten from what?
- Paragraph 2.6.1: Is there a reference which describes in more details the different carbon emissions?
- L7-8, P3637: How are primary emissions of SO<sub>4</sub> divided between the subranges 1a, 2a and 3b?
- P3638: All these parameterisations use a radius at 80% relative humidity (r<sub>80</sub>), which is probably different from the mean radius of a SALSA section. I assume that the mean radius of a SALSA section corresponds to the dry radius (r<sub>d</sub>) of particles. Then, for soluble particles, to calculate sea-salt emission, you need to use  $dF/dr_d$  rather than  $dF/dr_{80}$ . To do so, you can approximate  $dF/dr_d = dF/dr_{80} * dr_{80}/dr_d$ .  $dr_{80}/dr_d$  can be approximated using empirical relationships, such as those of Hanel.

Hanel, G.: The properties of atmospheric aerosol particles as functions of the Relative humidity at thermodynamic equilibrium with the surrounding moist air, *Adv. Geophys.*, 19, 73–188, 1976.

- L25-26, P3640: why? Is aqueous chemistry not considered in both SALSA and M7?
- P3641: For wet deposition, dry deposition and sedimentation, which diameter is used for each section? Does the diameter include the water uptake for soluble particles?
- P3641: sedimentation is very low compared to M7. This may be due to numerical sedimentation limitation with the time step?
- L 19, P3641: Why would the cloud cover be different between the two model runs? I thought that M7 and SALSA are based on the same meteorology?
- L1-3, P3642: Why are there windspeed differences when using SALSA rather than M7?
- L11-12, P3642: OC mass is in particles below 700microm in the model SALSA used. That may not always be the case in reality. Therefore, the term “this suggests that” is inappropriate.
- L18-19, P3643: “As dry .... both models.” This sentence is not finished.
- Section 3.1.4: Why is sedimentation large for sea-salt, while it is low for sulfate and BC? Please explain why sedimentation and deposition are important sinks for sea salt but comparatively they are not for black and organic carbon.
- L23, P3644: Why would the coagulation of particles be more important for black carbon than

for other chemical compounds?

- L24, P3644: Not clear: lower burden and lower removal than what?
- L3, P3645: Processes such as sedimentation depends on diameter rather than chemical composition. Why is it more important for sea salt than for black carbon?
- L22-24, P3646: What do you mean: most of H<sub>2</sub>SO<sub>4</sub> is used for nucleation in M7 and none is left for condensation? But there is enough H<sub>2</sub>SO<sub>4</sub> for nucleation and the parameterized condensation in SALSA? Is it because of process splitting in M7?
- Paragraph 4.1.: Boylan and Russel (2006) propose model goal and performance to evaluate how models simulate PM mass. Please compute the statistics described in their paper and include them in a table.

Boylan James W.; Russell Armistead G., PM and light extinction model performance metrics, goal, and criteria for three-dimensional air quality models. *ATMOSPHERIC ENVIRONMENT* Volume: 40 Issue: 26 Pages: 4946-4959 DOI: 10.1016/j.atmosenv.2005.09.087 Published: AUG 2006

- L25-26, P3648: I don't understand how Table 7 gives information on the mass included in particles of diameter over 700nm.
- L12, P3649: What do you mean by « limited transport »?
- L18, P3640: Why looking at a single diameter of 200nm? If you look at a diameter of 500nm, the opposite to what is said here is observed. If one model performs better for a narrow diameter range than another, but if the opposite is observed for another diameter range, the conclusion should be that both models have difficulties. Changing the model configuration would probably change the narrow diameter range in which one model performs well.
- L26-28, P3650: Not true for Figure 5f and Figure5a, for which M7 is clearly better than SALSA. It seems that there is a shift between SALSA and M7 in the peak of the distribution corresponding to the largest diameters.
- L8-10, P3651: How do you differentiate marine and polluted air from the graph?
- I don't agree from the results presented here that SALSA performs better than M7 for cloud activation studies. To have a fair comparison, you should compute different statistics of comparison to observations, and take a larger range of diameter: 100-700nm for example.
- L25-26, P3651: How long are the simulations performed for? Do they represent a typical year? More detail on the meteorology should be given. Why don't you use the Heintzenberg data for one specific year that you simulate?
- L20-23, P3652: From Figure 7, I do not agree that SALSA is better than M7. To look at the data objectively, you could see which of the two models give lower root mean square error.
- L26-27: No, between 0.01 micrometers and 0.1 micrometers, it seems that M7 is rather better.
- L1, P3653: To which graph (latitude band) are you referring to?
- L1-7, P3653: What is the point of this paragraph? You already mentioned that SALSA has high low-diameter particle concentration.
- L7-9, P3654: If we look at the scale of Figure 9, it seems that both models AOD are within 0.02 of the satellite retrieval. Is it correct?
- L18, P3654. « M7 has AOD at least 0.05 higher than SALSA ». Why is this information interesting?
- L1-3, P3655: This does not show from Figure 9. At high latitudes, M7 seems to perform better than SALSA. Also, SALSA seems to strongly underestimate AOD in the North half part of Africa, India and South East Asia.
- Please modify the conclusion following the previous comments.

#### **Minor comments:**

- L11, P3626: Replace « aeosol » by « aerosol ».
- L17, P3626: Replace « from size bin » by « from one size bin »
- L28, P3626: Replace « and has been applied in » by « and which has been applied to »
- L12, P3627: Replace « used spectral » by « used a spectral »

- L20, P3627: Replace « with SALSA » by « with the SALSA »
- L5, P3628: Replace « by the differences » by « by differences »
- L5, P3628: Replace « within ECHAM5-HAM » by « within the ECHAM5-HAM »
- L7, P3628: Replace « SALSA model » by « The SALSA model »
- L16, P3628: Remove the last s from « processess »
- L18, P3628: Replace « in the given size » by « in a given size »
- L19, P3628: Replace « compounds have been » by « compounds are »
- L20, P3628: Replace « number concentrations and mass concentration » by « number and mass concentrations »
- L26, P3628: Replace « for 3<sup>rd</sup> subrange » by « for the 3<sup>rd</sup> subrange »
- L27, P3628: Replace « constrained to » by « constrained by »
- Table 3: Replace « consist seasalt » by « consist in sea salt »
- L5, P3629: sulfate or sulphate is used in this manuscript. Please be consistent and keep only 1 spelling.
- L6, P3629: Replace « are grown » by « grow » and « lost » by « are depleted ».
- L23, P3629: Replace « processes » by « process »
- L25, P3629: Replace « will » by « can »
- L27, P3629: Replace « collisions.... is omitted, » by « Coagulation is neglected when both colliding particles have diameters exceeding 700nm »
- L3, P3630: Replace « set » by « sets »
- L3, P3630: Replace « For collision rate.... in » by « For the coagulation collision rate, we use the expression of »
- L19, P3630: Replace « to whole » by « to the whole »
- L19, P3630: Replace « with an exception in the cases » by « except for cases »
- L1, P3631: Replace « 1 for activation (or 2 for kinetic) nucleation scheme » by « 1 for the activation and 2 for the kinetic nucleation schemes »
- L20, P3632: Replace « to the soluble ... diameter » by « to a soluble bin in the same diameter range ».
- L25, P3632: Replace « 2<sup>nd</sup> and 3<sup>rd</sup> subrange » by « the 2<sup>nd</sup> and the 3<sup>rd</sup> subranges »
- L3, P3633. For clarity, replace « material » by « mass mixing ratio »
- L4, P3633. Replace « bin mean diameter » by « the mean diameter of section XX »
- L18-19, P3633: Replace « using solubility of different compounds using » by « using the solubility of different compounds following the »
- L22, P3633: Replace « Ri is the size » by « Ri is a size »
- L24, P3633: Replace « liquid and ice fraction » by « liquid and ice fractions »
- L3, P3634: Replace « Coefficients Ri for SALSA follow » by « The coefficients Ri for SALSA are obtained from »
- L8-10, P3634: Please rephrase.
- L13, P3634: Replace « inverse of ... surface » by « the inverse of the resistance at the surface »
- L21, P3634: Replace « resistance of » by « resistance at »
- L25, P3634: Replace « by parameterisation of pH » by « a parameterisation depending on pH »
- L26, P3634: Replace « Total resistance » by « The total resistance ».
- L12-13, P3635: Replace « For particle radius and diameter ... mode mean radius » by « The particle radius is assumed to be equal to the bin mean radius. »
- L3, P3636: Replace « for year 2008 the emissions from year 2000 may cause discrepancies » by « for the year 2008, the emissions from the year 2000 may be a cause of discrepancies ».
- L21, P3637: Replace « show » by « estimate »
- L23, P3637: Replace « has found out » by « estimates »
- L23, P3637: Replace « using » by a comma.
- L1-2, P3638: Replace « For the smallest ... 400nm » by « For radii between 50nm to 400nm, »
- L19, P3638: Replace « where parameter C » by « where the parameter C »
- L22, P3638: Why is there an upper limit of 32 m s<sup>-1</sup> set for the wind speed?

- L23-24, P3638. Does the sentence end after separately?
- L24, P3638: Replace « using sectional » by « using the sectional ».
- L18, P3639: Replace « we have r.... wavelength » with « , r is the mean radius of a section and lambda is the wavelength ».
- Please be consistent in the notations used in the paper. For clarity, it is better to always use « section » rather to switch between « section » and « bin ».
- L21, P3639: Do you mean mixing ratio by « amounts ».
- L27, P3639: Does « level » refer to « vertical level »?
- L15-16, P3640: Replace « unchanged ../../ M7. » by « similar between SALSA and M7 »
- L17, P3640: Replace « nearly identical » by « very similar »
- L28-29, P3641: Remove « However, the more detailed .... study »
- L10, P3642: Replace “differs only” by “differs by only”
- L22, P3644: Replace “found in” by “found for”.
- L16, P3647: Replace « within » by « of the »
- L25, P3647: Replace « and all 11 » by « although all 11 »
- L26, P3647: Replace « for EMEP » by « at EMEP »
- L1, P3648: Replace « data includes the mass .... in diameter » by « data corresponds to the mass concentrations of particles of diameter low than 2.5microm. »
- L5, P3648: Replace « In the organic » by « For the organic »
- L6, P3648: Replace « Out of the 117 comparison pairs 45 (36.5%) the simulated concentrations are » by « Out of the 117 comparison pairs, 45 (36.5%) are »
- L7, P3648: Replace « factor of two within » by « factor of two of ».
- L14, P3648: Remove the sentence « However, there are .... average ».
- L20, P3648: Replace « differs » by « differs by ».
- L20, P3648: Replace « observed » by « observation »
- L28, P3648: Replace « The concentration of SO4 has a mean of » by « The mean simulated concentration of SO4 is »
- L8, P3649: Replace « Of the 117 » by « Out of the 117 »
- L13, P3649: Replace « in Europe » by « over Europe »
- L17, P3649: Remove « simulated » before « size distributions »
- L18, P3649: Replace « together for » by « at ».
- Figure 5: the diameters should have the same units in the Figure and in the explanations.
- L16-17, P3655: Replace « AOD of 12. M7, however, .... SALSA or AERONET » by « AOD of 12, and than M7 AOD which is equal to 14 ».