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Comment

## ***Interactive comment on “MATCH–SALSA – Multi-scale Atmospheric Transport and CHemistry model coupled to the SALSA aerosol microphysics model – Part 1: Model description and evaluation” by C. Andersson et al.***

**Anonymous Referee #3**

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The implementation of the aerosol dynamics model SALSA in the regional atmospheric chemistry transport model MATCH for modeling of particle number concentrations on European scale is presented. Importantly, the manuscript identifies issues with the model and areas of future development. The rather comprehensive evaluation of the model with observation data of number size distribution, total PNC, EC, OC, PM<sub>1</sub>, and PM<sub>2.5</sub> is greatly appreciated. The manuscript gives an overview of the new model, but many details on the respective processes are not provided in the manuscript. Instead, it is referred to the Supplement, which turns out to be a rather long SMHI report by the

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same authors. This impairs the readability of the manuscript since one has to search for the corresponding parts in the Supplement. Even so, many details on the aerosol dynamics modelling are still missing. For instance, the description of condensation in the Supplement does not extend over what is already stated in the manuscript text. Unfortunately, all sensitivity tests that could help to evaluate the assumptions made in MATCH-SALSA are presented in part 2 of the manuscript, not accessible to the reviewer.

The SALSA model has some focus on the prediction of activating cloud droplets. However, the prediction of activating cloud droplets is not included in the current evaluation of MATCH-SALSA. It is mentioned that a more advanced cloud activation scheme can be coupled online, but I got the impression that this is computationally too expensive to be applied operationally. On the other hand, if the prediction of PNC and size distributions is the focus of the new model, then maybe an aerosol dynamics model better suited for prediction of new particle formation should have been preferred for the implementation in MATCH.

Before I can recommend publication in GMD, the authors should address the following concerns and also the technical corrections.

1.) The authors state that they do not expect to model BSOA formation in a realistic way and justify this by the need to make progress in the model development. Instead of consequently using the best yield estimate available, the authors chose 30%. By doing so, they ignore yield values currently applied in other models or recommended in literature. The value of 30% would not be so critical if they had decided to use a reasonable saturation vapor pressure for the semi-volatile compounds, instead of setting it to zero. The decision for treating SOA formation in this way might be motivated by the wish to match observed OC, but that is not a justification for making unrealistic assumptions. The SOA parameterization in the model should be revised for example by using the simple parameterization as applied in GLOMAP (Scott et al., 2014), with a fixed molar yield of 13% (equivalent to a 14.3% mass-based yield) of SOA from the

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oxidation of monoterpenes.

2.) Why is a rather outdated isoprene chemistry scheme used instead of the detailed scheme which is included in the EMEP MSC-W model's EmChem09 scheme? Further, isoprene is not included as SOA precursor in MATCH-SALSA. The authors should justify the use of an outdated isoprene scheme and state whether it is planned to include SOA formation from isoprene oxidation.

3.) A so-called "nitrogen gas-particle partitioning" is introduced in this manuscript. However no explanation on the nitrogen gas-particle partitioning and how it is solved in MATCH-SALSA can be found in the manuscript or in the Supplement. Furthermore, it needs to be stated which species are involved in the nitrogen gas-particle partitioning.

4.) More details on the treatment of the emission of primary particle components, in terms of mass and number need to be provided, and how consistency between mass and number of emitted particles is ascertained. Elemental Carbon (EC) is not defined in the first size range (nucleation and Aitken modes) despite EC is emitted from various combustion sources (mainly from residential biomass burning and traffic) in the Aitken size range. The attribution of EC should be revised for the PNC modelling on the European scale where many regions are under the influence of combustion sources.

5.) Nucleation is solved concurrently with condensation using the methodology by Jacobson (2002). Coupling nucleation with growth avoids that one of these processes is favored over the other in the operator splitting. However, the manuscript states that several nucleation options exist in MATCH-SALSA, including for example the activation of sulfuric acid and organic vapors, while the original methodology by Jacobson (2002) was derived for homogenous binary nucleation. Despite the authors mention that the alternative nucleation options are only used in part 2 of the manuscript, it is recommended to present the detailed algorithm for the coupling of sulfuric acid - organic nucleation with growth (give equations!) in part 1.

6.) It is suggested to split section 2.3 into two sections, one that deals with deposition

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and one that deals with cloud droplet activation. If there is any interaction between the two processes in MATCH-SALSA, it has to be stated more clearly as it is the case now. Based on the current description it is not obvious why there is a need for online calculation of cloud condensation nuclei, since the fraction of activated cloud droplets in the standard version of MATCH-SALSA is only inferred from the fraction of particles that are located in grid boxes covered with cloud. That can be done equally well in a simple post-processing of the model output.

7.) Provide reasons for the underestimation of total PNC at Northern and Central European sites and of accumulation mode particle numbers at all sites, in the Abstract and in the Conclusions. The list of planned developments given at the end of the manuscript is useful, but it does not replace a discussion on how missing processes or inadequate parameterizations have affected the presented model results. Specifically, it has to be assessed how the obvious shortcomings in the treatment of BSOA and nucleation mechanism affected the prediction of total PNC.

#### Technical corrections

P. 3268, line 17-20: when discussing modal vs. sectional schemes, examples for the application of both in aerosol dynamics models and the corresponding literature references should be given. How does the sectional approach compare with the modal approach? Mention some advantages and disadvantages of both.

P. 3268, line 21-23: PM-CAMx and GLOMAP are mentioned as two examples of CTMs that include aerosol dynamics and are applied on the European scale. Give a short summary of the shortcomings of these models with respect to their capability to predict observed PNC in Europe.

P. 3269, line 2: Exemplify briefly what the new model features of MATCH-SALSA are.

P. 3269, line 22-23: "makes it possible to describe PNC and the mixing state of the particles." Revise language. It has to be explained more precisely what this entails:

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physical treatment, computation, model output, etc. It should also be stated briefly which size distributions are defined, with reference to section 3 where this is explained in more detail.

P. 3269, line 26: "New emissions are emitted"; revise language of this sentence.

P. 3270, line 2: Replace "model chemistry" by "model gas-phase chemistry".

P. 3270, Footnote 1: The footnote should be included in the main text because otherwise it is difficult for the reader to comprehend the statement on P. 3286, line 1; which explains the underestimation of OC and PM peaks at Melpitz by a vegetation fire episode.

P. 3271, line 5: Describe the coupling between gas-phase chemistry and aerosol dynamics. How frequently are gas-phase concentrations of the relevant species (e.g. sulfuric acid) updated by the changes due to condensation and nucleation?

P. 3271, line 17-19: A complete list of the reactions of the MATCH-SALSA model is missing in the manuscript and in the Supplement.

P. 3273, line 5: The reference to the paper by Lehtinen et al. (2007) is missing in the list of References.

P. 3273, line 17: "accurate over time step length of 7200 s" - presumably this accuracy is only achieved with condensation is the only operative aerosol dynamical process.

P. 3277, line 7: Replace "(PM<sub>2.5</sub> and PM<sub>1</sub>)" by "(PM<sub>1</sub> and PM<sub>2.5</sub>)".

P. 3277, line 18: Why was Mace Head chosen as a station for evaluation of PNC? It is known that new particle formation at Mace Head occurs via nucleation of iodine oxides (e.g. Saiz-Lopez et al., 2006). Therefore it cannot be reproduced by a model that uses nucleation parameterizations for sulfuric acid clusters.

P. 3278, line 9: High PNC in nucleation mode along shipping lanes are probably artificial since kinetic and activation nucleation parameterizations tend to overestimate the

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nucleation rate in the ship plume (e.g. Metzger et al., 2010).

P. 3280, line 4-5: This explanation is in contradiction with the fact that the formation of <3 nm particles is parameterized as J3 according to Lehtinen et al., 2007 (see P. 3273, line 4-5).

## References

Jacobson, M. Z.: Analysis of aerosol interactions with numerical techniques for solving coagulation, nucleation, condensation, dissolution, and reversible chemistry among multiple size distributions, *J. Geophys. Res.*, 107, 4366, doi:10.1029/2001JD002044, 2002.

Metzger, A., Verheggen, B., Dommen, J., Duplissy, J., Prevot, A. S. H., Weingartner, E., Riipinen, I., Kulmala, M., Spracklen, D. V., Carslaw, K. S. and Baltensperger, U.: Evidence for the role of organics in aerosol particle formation under atmospheric conditions, *P. Natl. Acad. Sci. USA*, 107 (15), 6646-6651, 2010.

Saiz-Lopez, A., Plane, J. M. C., McFiggans, G., Williams, P. I., Ball, S. M., Bitter, M., Jones, R. L., Hongwei, C., and Hoffmann, T.: Modelling molecular iodine emissions in a coastal marine environment: the link to new particle formation, *Atmos. Chem. Phys.*, 6, 883-895, www.atmos-chem-phys.net/6/883/2006/, 2006.

Scott, C. E., Rap, A., Spracklen, D. V., Forster, P. M., Carslaw, K. S., Mann, G. W., Pringle, K. J., Nivekäs, N., Kulmala, M., Lihavainen, H., and Tunved, P.: The direct and indirect radiative effects of biogenic secondary organic aerosol, *Atmos. Chem. Phys.*, 14, 447-470, doi:10.5194/acp-14-447-2014, 2014.

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