**Atmospheric
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Discussions

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Interactive comment on "Does the POA-SOA split matter for global CCN formation?" *by* W. Trivitayanurak and P. J. Adams

Anonymous Referee #3

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General comments

The paper summarizes a new treatment of carbonaceous aerosol in the TOMAS microphysics module and presents an application of the model to simulations of relative impacts of POA and SOA emissions on CCN formation. The treatment of microphysical processes is compelling and provides a good basis for modeling effects of POA and SOA emissions on CCN formation.

However, a substantial concern is that the objectives of the study are not sufficiently clear. The authors state in very broad terms that results of their study address the relative importance of POA and SOA emissions for CCN. This is misleading given that they make very specific and highly idealized model assumptions about emissions and

processes. The rather brief review of previous studies in the introduction lacks specific context and clear motivation of this approach. See comments below for details.

Specific comments

Throughout the text: It should be clarified whether the model simulates elemental carbon (EC) or black carbon (BC). BC is the non-volatile and refractory component of the carbonaceous aerosol whereas EC is normally determined by thermal measurements. Emissions data sets for models are typically for BC.

P. 10563, l. 15 and p. 10567, l. 22 and elsewhere: If OA and OM are considered to be equivalent in this study it is not clear why different terms are used for the same type of aerosol?

Table 1: OM emissions in the current study are about 60% higher than the more recent CMIP5/RCP emissions. This difference reflects fundamental biases and uncertainties in emissions which are relevant to the current study. Uncertainties in emissions should be considered.

P. 10568, l. 13-17: References need to be added. The assumed sizes of primarily emitted aerosol particles are rather small (a brief review is available from Bond et al., 2013, for instance). Model results can be expected to be sensitive to the assumed sizes of the emitted particles. Given the choices made here, one may expect the model to produce CCN concentrations that are too high. Therefore, the sensitivity of CCN to relative POA emissions is perhaps unrealistically high in the model. What is the impact of the assumed size on these sensitivities?

P. 10569, l. 17-18: Please clarify whether the surface area of the hydrophilic aerosol component is used here?

P. 10570, l. 16-17: Are the same assumptions are applied to all types of emissions, including open fires? The original approach proposed by Cooke et al. (1999) only applies to fossil fuel emissions. It seems unlikely that these assumptions are generally

applicable to all types of emissions.

P. 10570, l. 29-P. 10571, l. 1: It seems that there will almost certainly be a systematic effect on aerosol burdens and the level of significance depends on circumstances such as the length of the simulation etc., even if the difference is small. Perhaps the sentence should be reworded?

P. 10571, l. 20-23: A rather problematic assumption is to take the POA emission rate as the total (POA+SOA) aerosol source and to artificially vary the contributions of POA and SOA emissions. This implies that the SOA emissions can be represented by POA, which is a poor assumption for various reasons. Fundamental sources of uncertainty are very different for POA and SOA emissions. Spatial patterns and diurnal cycle of SOA emissions differ markedly from emissions of POA. Furthermore, the conversion of organic precursors gases to SOA mainly occurs in the troposphere above the surface. This increases the lifetime of the aerosol relative to aerosol that is emitted at the surface. For instance, Zhang et al. (2012) attribute an increase in POM burden between ECHAM-HAM1 and ECHAM-HAM2 by about 50% to vertical transport of condensable gases by convection and SOA formation in the upper troposphere in HAM2, which has been omitted in HAM1. Consequently, effects of POA emissions on CCN concentrations can be expected to be underestimated.

Although sources of POA and SOA are generally different, primary POM emissions in current inventories are likely to go through a cycle of evaporation, oxidation, and recondensation as oxidized, lower-volatility products (Hallquist et al., 2009). The author should clarify the purpose of their sensitivity study by identifying specific processes that produce POA and SOA in the atmosphere. Information about causes of uncertainty for these processes needs to be included so that the reader can understand why such a very wide range of SOA/OA source ratios (0%, 10%, 50%, 90%, and 100%) needs to be considered.

P. 10573, l. 24: Missing "emissions"?

Table 3: Why is the nucleation rate greater for BASE? This seems counterintuitive given that the aerosol burden is higher for this simulation. This should lead to less efficient nucleation of sulfate aerosol owing to increased condensation of sulfuric acid on pre-existing aerosol.

P. 10575, l. 5 and following: Why are only comparisons for Europe included? Data from other networks should be added, e.g. from IMPROVE. Comparisons for Europe are unlikely to be representative of results in other regions, which makes these comparisons rather pointless.

P. 10575, l. 22-23: This speculation seems vague. The relationship between CCN concentrations and aerosol mass is highly nontrivial. It can be argued that a skillful representation of POM particle sizes is more important for CCN concentrations than an accurate simulation of aerosol mass yet little attention is given to aerosol size in this study. Furthermore, comparisons for mass are based only on a very small number of model grid points, which does not provide any useful constraints for global results.

P. 10576, l. 23: Do the authors rule out other effects? It is somewhat surprising that only three different effects of changes in emissions on CCN concentrations can be identified. For instance, as shown in Table 3, OA aerosol emissions generally affect nucleation rates, the growth of the aerosol through coagulation, and the deposition. All of these changes clearly have important consequences for aerosol number and CCN concentrations. It would be useful to include results similar to Table 3 here.

P. 10580, l. 15: Please provide mean values instead of a range of values.

References

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