Atmos. Chem. Phys. Discuss., 13, C11347–C11352, 2014
www.atmos-chem-phys-discuss.net/13/C11347/2014/

Atmospheric Chemistry and Physics Discussions
Open Access

Interactive comment on "Secondary organic aerosol formation during June 2010 in Central Europe: measurements and modelling studies with a mixed thermodynamic-kinetic approach" *by* B. Langmann et al.

B. Langmann et al.

baerbel.langmann@zmaw.de

Received and published: 22 January 2014

Answers to anonymous referee #3:

Reviewer: 1. Based on my reading of this manuscript, the authors have two major objectives: (1) to extend the SOC formation scheme and evaluate the impact on simulated SOC mass concentrations; (2) to explore the effect of organic vapour nucleation. I feel that the manuscript can be improved if the authors focus solely on the first objective, for the following reasons. (1) As the authors pointed out, nucleation events at

Puy-de-Dôme were rare during the chosen period of investigation. There are lots of measurements of nucleation events at a number of sites in Europe (within the domain covered by the REMOTE simulation). To present a convincing case, the authors need to look into periods when or sites where significant and frequent nucleation events occurred.

Authors: This will be done in future research with REMOTE. However, the current manuscript focuses on the measurement campaign in France during the second half of June 2010. The manuscript is modified putting a clear focus on the extension of the SOC formation scheme and the impact on simulated SOC mass concentrations. The nucleation study will only appear as a sensitivity case study.

(2) The nucleation scheme used in the REMOTE (e.g., Vehkamaeki et al., 2002) is for binary homogeneous nucleation of H2SO4-H2O. To treat LV-SOG as H2SO4 in binary nucleation parameterization calculation is completely unjustified and does not offer much scientific insights.

Authors: As written above, the manuscript is modified putting a clear focus on the extension of the SOC formation scheme and the impact on simulated SOC mass concentrations. The nucleation study will only appear in a sensitivity study.

(3) REMOTE is a climate-chemistry model. The authors should and can readily extend objective 1 by looking into the impacts of the extended SOC formation scheme on cloud properties and regional climate.

Authors: This is indeed an interesting topic, but it is out of the scope of the present manuscript.

Reviewer: 2. H2SO4 gas and LV-SOG concentrations are key components of this study. The authors should provide their values. The sensitivity of LV-SOG (and maybe also SVSOG and MV-SOG) concentrations to major assumptions (e.g., threshold values, VOC emission rate, etc.) should be presented and discussed.

Authors: We note that an evaluation of the species concentrations with measurements is not possible, as no measurements are available. In addition, we are only aware of one publication about SOA modelling (Yu, 2011) showing annual mean values of H2SO4 gas and LV-SOG concentrations and their ratio in Fig. 3 of the paper. The ratio of LV-SOG / H2SO4 gas over Europe in Yu (2011) ranges from 1-5, with the highest values over Northern Europe. Globally, the highest ratios of about 40 are determined in South America in the Amazon region, a photochemically active region with high biogenic VOC emissions. The measurement campaign analysed in the manuscript under review focuses on a high-pressure summer period, so that it cannot be directly compared with the annual mean values in Yu (2011). In addition, in the short period of time (about 10 days) the spatial distribution is rather inhomogeneous compared to Yu (2011). Therefore, we report here the range of the ratios of LV-SOG / H2SO4 gas for the late afternoon near surface layer, but do not include them into the manuscript, as we do not think they add a valuable contribution. The ratio of LV-SOG / H2SO4 gas over Europe in the current manuscript ranges from 0-50 in the fresh air masses (note, that due to the inhomogeneous spatial distribution very small concentrations occur as well) and increases locally up to 150 in the aged air masses. Taking into account that these ratios are modelled during a high pressure summer period in the late afternoon surface layer, the conditions and ratios are comparable with those shown in Yu (2011) for the Amazon region and can be regarded as reasonable.

Reviewer: 3. Page 26765, line 26. Please explain in more detail how you update the whole model domain with ECMWF data. Restart the simulation? How did you deal with chemistry fields when you force the model to stay close to the observed weather situation?

Authors: According to Langmann (2000), Langmann et al. (2003) and Langmann and Heil (2004) the procedure is as follows: REMOTE can be applied principally in two modes with respect to the model physics, the so-called “climate mode” or the “forecast mode”. In the climate mode the model is initialised once and then run continuously until the end of the simulation period with an update of the meteorological analysis data every 6 h at the lateral model boundaries. Between these six hours intervals, the analyses are interpolated linearly in time. In the forecast mode, the model is started at 0 UTC every day to compute a 30 h forecast and the meteorological analysis data are also updated every 6 h. The first six hours of the consecutive meteorological forecasts are neglected to account for a spin-up time. The total simulation period is composed of 24 h simulation segments with a discontinuity in the physical state of the atmosphere at 6 UTC. But, by starting the model again every day the internal model variability is suppressed and the model is forced to stay close to the observed weather situation. Aerosol and atmospheric chemistry processes, however, are calculated continuously like in the climate mode. This is done by simulating meteorology only in the first six hours of each 30 h forecast. Then, combined chemistry, aerosol and meteorology calculations continue for 24 h starting with the aerosol and atmospheric chemistry information from the last time step of the previous forecast. Technically this is possible easily with a meteorological restart file at 6 UTC from a meteorology only simulation with REMOTE. To clarify this in the manuscript, the following text has been added to the manuscript: 'The model is started at 0 UTC every day to compute a 30 h meteorological forecast. The first six hours of the consecutive meteorological forecasts are neglected to account for a spin-up time. The total simulation period is therefore composed of 24 h simulation segments with a discontinuity in the physical state of the atmosphere at 6 UTC. But, by starting the model again every day the internal model variability is suppressed and the model is forced to stay close to the observed weather situation. Aerosol and atmospheric chemistry processes, however, are calculated continuously. This is done by simulating meteorology only in the first six hours of each 30 h forecast. Then, combined chemistry, aerosol and meteorology calculations continue for 24 h starting with the aerosol and atmospheric chemistry information from the last time step of the previous forecast.' The sentence: 'Every 30 h, ECMWF data are used for an update over the whole model domain to force the model to stay close to the observed weather situation.' has been deleted from the manuscript.

Full Screen / Esc
Printer-friendly Version
Interactive Discussion
Discussion Paper

Reviewer: 4. Page 26768, lines 1-5. Simplification generally compromises accuracy. Could you discuss how your simplification might have affected the results?

Authors: Air masses undergo temporal and spatial changes in the chemical and meteorological condition, so that an empirical approach based on a few days long time series may not reflect the overall variability. Therefore, the approach by Yu (2011) represents the more general applicable approach compared to the simplified approach suggested in the manuscript. To clarify this in the manuscript, the following sentence is added: ‘However, it should be emphasised that the optimal maximum threshold value may be variable in time and space so that an application to different locations and seasons requires further evaluation.’

Reviewer: 5. Page 26772, lines 1-6. Why the second aging step doesn’t modify OC mass concentration much? Based on my understanding, the condensation of LV-SOG is critical in the thermodynamic-kinetic approach. Without second aging step (SV-SOG to LVSOG), where did LV-SOG come from?

Authors: Without the second aging step (SV-SOG to LV-SOG), the concentrations of LV-SOG and LV-SOC remain 0. In this case only mass concentration of MV-SOG and SV-SOG contribute to the generation of MV-SOC and SV-SOC aerosols. Obviously, the general availability of secondary organic gases limits the modeled mass concentrations of SOC when considering the first aging step only or the second aging step in addition in the presented model simulations, because increasing biogenic emissions increases of SOG and SOC concentrations greatly. It should be noted, however, that even though the differences in SOC mass concentrations by considering the first aging step or the additional second aging step are small, the size distribution (Fig. 7) reveals modifications of the microphysical processes and the contribution of LV-SOG condensation.

References:

Langmann, B.: Numerical modelling of regional scale transport and photochemistry directly together with meteorological processes, Atmos. Environ., 34, 3585–3598, 2000.

Langmann, B., Bauer, S. E., Bey, I.: The influence of the global photochemical composition of the troposphere on European summer smog, Part I: Application of a global to mesoscale model chain, J. Geophys. Res. 108, 4146, doi: 10.1029/2002JD002072, 2003.

Langmann, B. and Heil, A.: Release and dispersion of vegetation and peat fire emissions in the atmosphere over Indonesia 1997/1998, Atmos. Chem. Phys. 4, 2145–2160, 2004.

Yu, F.: A secondary organic aerosol formation model considering successive oxidation aging and kinetic condensation of organic compounds: global scale implications, Atmos. Chem. Phys., 11, 1083–1099, 2011.

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 26761, 2013.