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**Atmospheric Chemistry and Physics Discussions**

# *Interactive comment on* "Chemical and aerosol processes in the transition from closed to open cells during VOCALS-REx" *by* J. Kazil et al.

**Anonymous Referee #1**

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The manuscript provides a thorough analysis of the potential means of resupplying CCN to the MBL in the SEP region, comparing the modelled cloud structures with those measured during VOCALS-REx. The model description, the simulations and the model-measurement comparison are all suitable material for publication in ACP and I recommend publication after the following points have been addressed.

The methodology is generally very sound, but the authors might consider and comment on the following points:

The Clarke et al., 2006 scheme for SS is one of many. What is the possible sensitivity of the conclusions to the scheme chosen? Whilst the impact of the arguable amounts of organics in the seaspray on cloud activation in the clean MBL may be negligible, there will probably be a non-negligible sensitivity to size. Firstly, various SS schemes

generate different numbers. Secondly, any organics present can push the size distribution to smaller sizes by affecting the bubble bursting mechanism (e.g. Fuentes et al., 2010). Can the authors comment on the magnitude of the impact of such a reduction in the modal size of the seasalt emissions?

How well can a representation of the aerosol by 3 lognormal modes accommodate the aerosol microphysics across 6 orders of magnitude from nucleation to coarse SS? Has a sensitivity analysis been conducted, perhaps by comparison with the MOSAIC sectional treatment of aerosol within WRF-Chem? Whilst, as stated on line 18 on page 4696, the incorporation of the freshly nucleated particles into the Aitken mode is a viable simplification, the growth timescales for repopulation of the Aitken mode will likely be underestimated (e.g. does recalculation of the moments of the mode based on addition of the nucleated particles ignore the very high Kelvin term resistance to growth of the extremely small particles?). The authors should confirm whether this is the case and comment on the possible implications. It is noted and applauded that a separate nucleation mode is planned for incorporation.

On a related note, the assumption that all Aitken mode particles are cloud processed to the accumulation mode does not seem intuitively physically correct, even though the sensitivity has been evaluated previously. It would surely depend upon the distinction between the "modes" (composition, mixing state and how close each of them really is to a lognormal distribution), which will be defined by the model setup.) Could the authors comment on whether the conditions likely to disqualify the use of this assumption will apply in the region of interest?

RADM2 is already a very simple and rather dated chemical scheme. The further simplified scheme presented in tables C1 to C5 present an undoubtedly stable, but rather rudimentary chemical scheme, particularly with respect to DMS. Removing species from RADM2 without conserving reactivity will inevitably affect oxidant concentrations (perturbing the VOC limitation of O3 production and destruction and the primary and secondary radical reaction channels producing OH and reducing the OH and NO3 losses). Coupled to the simplified single stage oxidation of DMS to SO2, this will propagate through to the H2SO4 production rate. This will interact with the uncertainty in condensation sink (and with any errors in the semi-analytical nucleation treatment) to impact on the nucleation rate. Have the authors evaluated the reduced mechanism against any of the more comprehensive schemes available in WRF-Chem (e.g. CBMZ)? The authors should discuss these considerations and comment on how such uncertainties could impact on the conclusions of the study.

Can the authors please comment on the potential impacts of disabling dry deposition of gaseous species in the simulations and why they did not choose simply to reduce the depositional loss rate to more reasonable values rather than completely disabling the process?

On p4691, the sentence starting line 6 reads a little clumsily. In addition, whilst it is correct, it doesn't discuss the fact that the near-surface wind speeds will determine both DMS and seasalt emission flux. Indeed, the windspeed dependence of sea-air fluxes of DMS is well-documented and at the root of available parameterisations for oceanic DMS flux. It seems rather odd that the windfields are used to drive the seasalt parameterisation but not the DMS flux (perhaps normalised to reproduce the average observed DMS fluxes). Can the authors comment on the possible impact of wind-driven variation of DMS fluxes coincident with the wind driven fluxes of seasalt?

The results section is very clearly and appropriately presented, within the constraints of the assumptions in the model setup discussed so far. The model seems to be behaving physically and stably with intuitive results. I have a few comments that the authors might like to clarify:

Why were particles containing 15 H2SO4 molecules used to represent the nucleation mode? I presume it is the number of molecules in roughly nanometre sized particles, but this should be stated explicitly.

There is a discussion of the growth timescales for nucleation mode aerosol on p 4702

line 15 onwards, along with discussion of the coagulation sink. At this point, reference should be made to the impact of the modal methods on these timescales, based on the answer to the query raised previously.

The Uncertainties section is appropriate so far as it goes, though mention should be made of the other uncertainties discussed previously. There is one further comment that might be addressed:

On line 25 page 4703, it is stated that the fact that ultrafine aerosol are predominantly volatile supports the H2SO4-water nucleation mechanism. This is not correct. Nucleation is the formation of nanometre scale particles. Volatility of particles much larger than the critical cluster (one or two nanometres) yields no information about the nucleation mechanism.

The comparison with measurements presents a useful reality check for the simulations, but I have one or two comments:

Line 18 page 4707: volatility cannot be used to identify composition. Many compounds volatilise at 300C and it is dangerous to make such an inference on volatility alone.

As shown by figure 9, the model does a reasonable job of reproducing the aerosol and cloud evolution. Model-measurement comparison for such specific case studies are highly ambitious using the very idealised model inputs and necessary assumptions in the model, so it is a credit that such a lack of sustantial bias is observed.

The discussion of the DMS discrepancies is interesting and I am in agreement that the measurements cannot be readily reconciled. However, a discussion of DMS flux windspeed dependence would be appropriate here. The discussion of the SO2 measurements is similarly confusing, but again convincing. It would appear that the DMS and SO2 measurements, far from providing useful constraint on the model, ask more questions than they answer.

I fully agree with the concluding remarks of the paper that "The results presented here form the groundwork for future research on the behavior of aerosol sources and their determining factors in the marine boundary layer, and on their role for cloud properties"

Minor comments: p4689, line 8: ceteris paribus is insufficiently widely used to not be italicised, otherwise use "all other things being equal"

p4690, line 4; presumably the necessary 1/cc/hr replenishment rate was of CCN at a given supersaturation at cloud base, not just a replenishment rate of any aerosol (of any size or composition) at any point in the BL. Please clarify.

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Interactive comment on Atmos. Chem. Phys. Discuss., 11, 4687, 2011.