Atmos. Chem. Phys. Discuss., 11, C4913–C4930, 2011
www.atmos-chem-phys-discuss.net/11/C4913/2011/

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.

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Received and published: 14 June 2011

Response to reviewer 1

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?

The reviewer is correct, given the multitude of available sea salt emissions schemes, and the uncertainties remain large (de Leeuw et al., 2011). A thorough answer would require a study where a simulation of the closed-to-open cell transition is repeated with different sea salt emissions schemes. Here we can discuss possible consequences of switching from the Clarke et al. (2006) sea salt emissions scheme to other schemes, and the shift in the sea salt aerosol spectrum due to the presence of organic matter in sea water. In the discussion, it is important to consider that the cloud processes in our simulations, which drive the transition from closed- to open cells, are far more sensitive to the number of available aerosol particles rather than to their size.

- The Clarke et al. (2006) sea salt emissions scheme is considered a "strong" source in terms of sea salt mass, but not necessarily in terms of number, owing to its higher flux of large particles compared to other schemes (see, e.g., figure 6 in Clarke et al. (2006) and figure 2 in Pierce and Adams (2006)). In global models, it delivers a higher sea salt burden than other schemes, but performs on par with these schemes when the global model runs are evaluated with observations (Pierce and Adams, 2006; Ma et al., 2008). The number flux of large sea salt particles ($> 1\,\mu$m) is only about 5 % of the number flux of smaller particles in our simulations. We expect a negligible or small effect in our simulations were these large particles neglected.

- Fuentes et al. (2010) report an increase in the production of sea spray particles with dry diameter $< 100\,\mathrm{nm}$ with increasing amounts of organic matter in sea water, and conclude that this effect is expected only to be significant in areas of high biological activity. They write, referring to the shift to smaller sizes of the sea spray aerosol spectrum observed in artificial sea water enriched with organic matter: *"These trends were in agreement with analogous size distribution measurements conducted with oceanic unfiltered seawater samples collected from productive and non-productive waters along the RHaMBLe cruise transect route."*

  MODIS-Aqua satellite derived chlorophyll imagery for the VOCALS-REx region, available from the VOCALS data sets page (http: //data. eol.ucar .edu/ master_list/ ?project=VOCALS), shows that even in the most productive areas near the South American coast, chlorophyll concentrations did not exceed $1\,\mathrm{mg\,m^{-3}}$. Farther away from the coast, chlorophyll concentrations were lower. This qualifies the VOCALS-REx region as non-productive compared to the productive regions sampled by Fuentes et al. (2010), which gave rise to the shift in the sea spray spectrum: Seawater from these regions contained far higher chlorophyll concentrations, 1–60 $\mathrm{mg\,m^{-3}}$ (Figure 1 in Fuentes et al., 2010). Given the low chlorophyll concentrations in sea water in the VOCALS-REx region, no effect on the sea spray spectrum due to organic matter should be expected. Text reflecting this consideration was added in section 4.3.3 (Sea salt emissions and entrainment from the free troposphere).

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?

Although the MADE aerosol scheme (Ackermann et al., 1998) has been available for a number of years, and has been implemented in widely used host models such as WRF (Grell et al., 2005) and ECHAM (Lauer et al., 2005), we have been unable to find a study in the published literature that would evaluate the ability of MADE to represent

aerosol microphysical processes such as coagulation and growth, e.g. in comparison with a highly resolved sectional aerosol scheme. We agree that the MADE modal aerosol scheme should have been tested in such a way early in its existence, as done e.g. by Vignati et al. (2004), and later by Kokkola et al. (2009) for the M7 aerosol module. These works give account on the abilities and deficiencies of the modal method. In spite of these deficiencies, we argue that the MADE aerosol scheme is adequate for the stated goals of the paper.

Has a sensitivity analysis been conducted, perhaps by comparison with the MOSAIC sectional treatment of aerosol within WRF-Chem?

Most commonly, sensitivity studies investigate the response of a result to a quantitatively known uncertainty or variability in the model input, and require varying the numerical values of that input. Here, the sensitivity study would compare the response of the results to switching the aerosol schemes. In WRF/Chem, the MOSAIC aerosol scheme has its own set of shortcomings whose effect in our simulations is quantitatively not well known: It is a coarse scheme with only 4 or 8 size sections, hence its connection to the two-moment cloud microphysics scheme and the aerosol nucleation scheme would be subject to artifacts. For example, the smallest MOSAIC size section represents aerosol particles down to a diameter of 39 nm. This is much larger than the particles produced by our aerosol nucleation scheme. The gap is usually bridged with a scheme that describes growth of freshly nucleated aerosol particles, their loss onto pre-existing aerosol, and self-coagulation (Kerminen and Kulmala, 2002; Lehtinen et al., 2007; Anttila et al., 2010). However, these descriptions all assume steady state, which may not be a robust assumption to describe growth and loss of the particles from their size at nucleation ($\sim 1.5\,\mathrm{nm}$) to 39 nm. On the other hand, modal aerosol schemes such as MADE are not subject to artifacts due to lower or upper size cutoffs (but suffer from other shortcomings).

For the study, the MOSAIC aerosol scheme would have to be connected to

- the simplified version of the RADM2 scheme gas phase chemical scheme used in our simulations
- the Feingold two-moment cloud microphysics scheme
- the aerosol nucleation scheme

After completing the required model development and running the simulations, the difference in the results obtained with the two aerosol schemes would have to be analyzed and discussed, and possibly traced back to the specific treatment of aerosol processes in each scheme. Such a study would be a great value for aerosol modeling, but would be unlikely to illuminate the processes taking place in the transition from closed to open cells.

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.

The Kelvin effect is neglected in the calculation of the growth of the aerosol modes by condensation in the MADE aerosol scheme (Ackermann et al., 1998). It is therefore correct that as long as the nucleation/Aitken mode has a very small geometric mean diameter (say < 3 nm), growth of its particles will be likely overestimated, in spite of the extremely small vapor pressure of sulfuric acid. We have added a reference to this in the “Uncertainties" section.

However, the nucleation scheme which passes freshly nucleated aerosol to MADE does account for the Kelvin effect, and hence in the recalculation of the modal parameters after the addition of these particles, we expect no consequence by neglecting the Kelvin effect.

It is noted and applauded that a separate nucleation mode is planned for incorporation.

Unfortunately, adding a nucleation mode can solve one problem and create others. The reason is that there is, to our knowledge, no fully consistent criterion that determines when a particle should be moved from the nucleation- to the Aitken mode, resulting in some level of arbitrariness. One approach, e.g., is to prescribe a diameter at which particles are transferred from the nucleation to the Aitken mode. This can have a significant effect on model results (Kazil et al., 2010).

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?

Activation and cloud processing are the mechanisms by which the accumulation mode comes about in the first place. In the model, it is assumed that the growth of cloud-borne aerosol particles resulting from collision coalescence of drops (reduction in number at constant mass) or aqueous chemistry (addition of mass at constant number concentration) will produce accumulation mode particles *upon evaporation of the drops*. Hence the criterion to place Aitken mode particles into the accumulation mode upon evaporation is based on the consideration of the involved physical processes as well as on observations (e.g. Hoppel et al., 1989). We consider this sufficient justification. We have made changes in Section 2.1 and appendix A1 to clarify this.

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).*

In the clean marine boundary layer, the major (photo-) chemical processes controlling the ozone mixing ratio are losses by photolysis and reaction with the HO_2 and OH radicals (Ayers et al., 1997). Given the very low NO_x levels during the VOCALS-REx campaign, which are indicated by the low observed CO, and supported by calculations by Yang et al. (2009), the reduction by NO of peroxy radicals from volatile organic compound (VOC) oxidation is unlikely to contribute to ozone generation. Therefore, in order to enable the computationally demanding simulations, the chemical scheme has been simplified by removing non-methane hydrocarbons. Another reason for excluding VOC reactions is that VOC levels were not measured during VOCALS-REx, and estimates of VOC levels (or zero VOC levels) would have to be used for model initialization. The oxidation of the ubiquitous methane to CH_3O_2, the reduction by NO of the latter followed by photolysis of NO_2, which results in ozone formation are, however, accounted for in our chemical scheme. The resulting chemical scheme captures the essence (zeroth and first order effects) of the gas phase chemistry in the clean MBL, and the text here has been added to section 2.2 of the manuscript to explain the choice of chemical scheme in more detail. We agree that the RADM2 scheme contains rate coefficients that have been superseded by more recent work. We can't but acknowledge that bringing all model components up to standard would occupy several scientists for a significant period of time, but would not change the key results of this paper.

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.

The one-stage oxidation scheme of DMS to SO_2 can be justified by the consideration that in the DMS → SO_2 formation chain, the rate limiting step is the initial DMS + OH

reaction (Tyndall and Ravishankara, 1991), because the reactions of its products are very efficient, as they involve the abundant O_2 (or H_2O). It is a widely used simplification of the fairly complex oxidation scheme of DMS, e.g. in chemical transport models such as MOZART (Emmons et al., 2010) as well as in the evaluation of field measurements (Faloona et al., 2010). Similar considerations apply to the oxidation of SO_2 + OH to sulfuric acid, where the initial reaction is the rate limiting step (Lovejoy et al., 1996). We have added a corresponding note in section 4.3 ("Uncertainties").

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.

The possibility to use the CBMZ chemical mechanism with the MADE/ SORGAM aerosol scheme is not available in the WRF/Chem version 3.1 used in our simulations, but has been implemented recently in WRF/Chem 3.3 (released April 2011). A sensitivity study using the CMBZ scheme would therefore require that we implement all changes we have applied to WRF/Chem 3.1 so far to version 3.3, running our current simulations again, as well as the numerically more expensive simulations with the CBMZ scheme, which contains more than twice the chemical species than the simplified scheme used in our simulations. Since VOC levels were not measured during VOCALS-REx, estimates of VOC levels (or zero VOC levels) would have to be used, resulting in an uncertainty in the sensitivity study. We see a contrast between the effort required to conduct the sensitivity study and its potential informative value, which would be subject to the study's uncertainties.

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?

We have added text to section 2.6 (Ocean sources and sinks) to reflect the following considerations that led to neglecting dry deposition in our simulations. The chemical species for which dry deposition is most relevant (in our simulations) are SO_2 and H_2O_2.

- Physically, SO_2, with a Henry's law constant of $\sim 1.4\,\mathrm{mol\,L^{-1}\,atm^{-1}}$ (Mondal, 2007) is far less soluble in water than H_2O_2 with a Henry's law constant of $1.1\times10^5\,\mathrm{mol\,L^{-1}\,atm^{-1}}$ (Staffelbach and Kok, 1993). However, SO_2 has a much higher dissociation constant than H_2O_2, and its effective Henry's law constant for uptake in sea water (pH $\approx$ 8.3) is $\sim 5\times10^7\,\mathrm{mol\,L^{-1}\,atm^{-1}}$. Both will be therefore taken up very efficiently by the ocean, and the limiting process for the loss is gas phase diffusion. Indeed, in the cloud-free MBL, dry deposition is the second most important loss process of SO_2 after loss onto aerosol (Faloona et al., 2010). In the cloud-capped MBL during VOCALS-REx, however, the strongest loss process of SO_2 is aqueous oxidation, while dry deposition is a minor sink (Yang et al., 2011).
- Observations suggests that current parameterizations overestimate dry deposition: Faloona et al. (2010) found an average dry deposition velocity of SO_2 in the (cloud-free) MBL that was about 30 % lower than calculated with parameterizations related to that of WRF/Chem (Wesely, 2007). Similarly, Gray et al. (2010) conclude from analysis of observations that “the current generation of global models may be significantly overestimating SO_2 deposition rates over some tropical marine areas".

In summary, slowing down dry deposition as calculated in WRF/Chem to an ad hoc estimate would have been a viable option. However, we have determined that switching it off is a realistic approximation on the grounds that it is a subordinate loss process in the cloudy MBL. This conclusion has been made independently of the possibility brought forward by Gray et al. (2010) that SO_2 uptake by the ocean is slowed by an ocean surface layer with reduced pH.

The overestimation of CO_2 dry deposition has turned out to be the result of WRF/Chem assigning a default deposition velocity to newly added species (which is too high for CO_2). CO_2 was added as species to our chemical scheme for technical reasons. We have corrected the text regarding dry deposition of CO_2 in section 2.6.

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.

We have changed the sentence to read as follows:

“The strength of MBL aerosol sources will be determined by various factors: Oceanic emissions of DMS, which provide the gas phase precursor molecules for nucleation and growth of aerosol particles, depend on near-surface wind speeds and seawater DMS concentrations (e.g. Huebert et al., 2004, and references therein). Sea salt emissions are driven by near-surface wind speeds as well, and while providing new CCN, suppress aerosol nucleation by increasing the sink for nucleating molecules on the other. Entrainment of aerosol from the free troposphere depends on the strength of the inversion, and on the presence of enhanced aerosol concentrations above it, e.g. from long-range transport of pollution.“

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 present study was designed for a specific observed case, hence using the DMS fluxes measured on board of the NOAA RV Ronald H. Brown for that case seemed straightforward. Using a parameterization would need to be justified in such a situation, and may in the end draw criticism as well. A parameterization normalized to the observations would indeed represent short-term and small-scale variability in DMS flux around the observed mean. However, we compare domain-averaged DMS profiles with aircraft profiles, and since DMS is not miscible in water, it can spread efficiently with the MBL circulation. It can hence be argued that the variability due to the wind-speed dependence would be suppressed in the averaged profile. However, we agree that this is a subject to be investigated. This topic is currently under investigation.

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.

Nucleation from the gas phase is quantified in the model with a lookup table that gives the formation rate of particles containing 15 H_2SO_4 molecules (approximately 2.1 nm in dry diameter at 290 K) as a function of environmental parameters (Kazil et al., 2010). The particles are placed in the Aitken mode of the MADE aerosol module, whose geometric mean diameter is assumed to be sufficiently small in conditions in which nucleation occurs so that it can act as a nucleation mode. The quantities that are passed to the Aitken mode of the MADE aerosol module are the number of particles and their total H_2SO_4 content.

There is a technical reason for working with the formation rate of particles containing 15 H_2SO_4 molecules: A priori, describing the nucleation rate requires knowledge of two quantities: The H_2SO_4 content of the critical cluster (which changes depending of temperature and gas phase sulfuric acid concentration), and the number of supercritical particles produced per volume and time interval. This would require two lookup tables. The number of lookup tables can be reduced to one by working with the formation rate

due to nucleation of particles with a fixed H_2SO_4 content, which must be larger than the H_2SO_4 content of the critical cluster in the conditions in question. The scheme of Kazil and Lovejoy (2007) can be used to calculate the formation rate of particles from the gas phase at a given size, under consideration of loss due to self-coagulation and coagulation with pre-existing particles. It was used to construct the lookup table of formation rates of particles containing 15 or more H_2SO_4 molecules (Kazil et al., 2010). In the construction of the lookup table, it was determined that in atmospheric conditions in which the critical cluster for nucleation of sulfuric acid and water contains 15 or more H_2SO_4 molecules, the nucleation rate is so small that it can be safely neglected (set to zero).

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.

We have no resources at this time to investigate the impact (over-/ underestimation) of the MADE modal aerosol scheme on the growth time scales of the aerosol particles in our simulations. We have therefore removed the parts on the growth timescale from the discussion in question, which now reads as follows:

“However, the relative strength of the two particle formation processes does not quantify their relative effect on CCN and cloud drop number concentrations: The freshly nucleated particles measure $\sim$1.5-2 nm in diameter, and need to grow to sizes of tens of nanometers before they can participate as CCN. The larger sea salt aerosol particles on the other hand can be activated to cloud droplets soon after emission."

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 reviewer is correct. The passage

"This consideration is backed by the predominantly volatile nature of ultrafine aerosol particles during VOCALS-REx RF06 (see Sect. 5.1), which supports the use of the sulfuric acid/water nucleation scheme (Kazil et al., 2010)."

was removed.

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.

This is correct. At the same time, previous aerosol volatility measurements over the remote Pacific in the MBL (Clarke et al., 1998) and free troposphere (Clarke et al., 1999; Clarke and Kapustin, 2002), made in conjunction with measurements of gas phase sulfuric acid, implicate the latter as the primary component in these nuclei. We have therefore changed the text to read as follows:

"Heating of these particles to 300 °C revealed that they consisted of volatile compounds at this temperature. Similar volatility measurements over the remote Pacific in the MBL (Clarke et al., 1998) and free troposphere (Clarke et al., 1999; Clarke and Kapustin, 2002), all made in conjunction with gas phase sulfuric acid, implicate the latter as the primary component in these nuclei."

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 substantial 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.

We have added a note on the wind-speed dependence of DMS emissions in section 1 (Motivation). However, we compare domain-averaged DMS profiles with the aircraft observations, and it is not immediately obvious that the wind speed dependence in a parameterization of DMS emissions would appear in the averaged profiles we show, since DMS is not miscible in water, and can therefore spread efficiently with the MBL circulation.

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.

We agree with this assessment. We are optimistic that over time, the discrepancies and open question will be resolved, as the understanding of the system both in terms of measurements and modeling deepens.

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"

We have changed this in the revised manuscript, but the final typesetting is done by the editorial staff according to ACP guidelines.

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.

In the determination of the aerosol source strength required to maintain open cell circulation, Wang et al. (2010) used a source of accumulation mode particles uniformly distributed over the depth of the boundary layer. The log-normal parameters of the accumulation mode were based on observations during VOCALS-REx. We have changed the text passage in question to read as follows:

"Wang et al. (2010) showed in a case study of the Southeast Pacific stratocumulus deck that replenishment of accumulation mode particles at the rate of $\sim 1\,\mathrm{cm}^{-3}\,\mathrm{h}^{-1}$, uniformly distributed over the depth of the boundary layer, was sufficient to maintain an open-cell circulation."

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