**Atmospheric
Chemistry
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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 2

In this paper authors investigate effect of the aerosol emission from the ocean surface, aerosol formation from sulphuric acid and entrainment of the aerosol from above the boundary layer on the stratocumulus cloud evolution using numerical model. In particular authors are trying to answer the question if the mentioned above aerosol sources provide enough aerosol to keep open cell circulation within the marine boundary layer; with the sink of the aerosol being removal by the drizzle droplets. Numerical model (WRF/chem) is modified by authors, and the two moment aerosol MADE module is

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coupled with the two-moment microphysics, resulting in the scheme capable of simulating aerosol formation, aerosol growth to the CCN sizes and later used as a CCN in microphysics parametrization; and the transport/reaction of the chemical species by/with the cloud/rain water. Authors find that combined effect of the aerosol from the surface, nucleation of the aerosol from the H2SO4 and entrainment is sufficient to maintain open cell circulation and explain in detail aerosol formation in the cloud free regions. Authors also find agreement between numerical model results and an observations for the concentration of the aerosol bigger than 120 nm. There are differences in the modelled and observed SO2 concentration, especially inside the cloud, which at this stage authors can not explain.

After reading the manuscript I have few questions authors might consider to address in revised version: Specific comments: a)Part where the model is described is split into main text and appendix what makes reading difficult and some information is repeated in both.

In an early version of the manuscript, the detailed model description was part of the manuscript body. However, it was tedious and distracting. We have therefore placed the model description in the appendix. Selected parts were retained in the manuscript body, to provide a basis for the understanding of the model. We agree that the model description is difficult to read.

b)It's not clear from the description how exactly aerosol is activated. How number of activated CCN is related to the supersaturation (P4715L14)?

In our model the supersaturation is calculated much as it is in a bin method (for details, see Feingold et al. (1996); Stevens et al. (1996)), except for the fact the drop size spectrum conforms to a lognormal size distribution derived from two moments (mass and number). The ambient supersaturation is used to determine the size of the minimum aerosol particle size that will be activated. All particles larger than that size are activated to drops.

c)I'm concerned about aerosol conservation. Since microphysics keeps (redundant) information about aerosol concentration and mass (mass can be calculated by integration of the: $N(r_a)r_a^3$ over the r_a) ...

Our cloud microphysics keeps track of two fundamental aerosol properties: mass and number. Aerosol size (the geometric mean diameters of the log-normal modes) is a diagnostic quantity in the model, and is used only for activation in the cloud microphysics routines.

... is mass and number of the aerosol conserved in this scheme during deactivation?

The aerosol and cloud microphysics code has balance equations for aerosol mass, which is reduced only by wet removal, and aerosol number, which is reduced by coagulation, collision-coalescence, and wet removal. Upon evaporation of a drop, one particle is returned to the atmosphere (regenerated) for each drop evaporated.

Or maybe conservation is ensured by varying geometric mean diameter for each mode. But in the latter case is coarse mode geometric mean diameter always bigger than accumulation mode geometric mean diameter?

The geometric mean diameters of the log-normal aerosol modes are diagnostic quantities, and are calculated from the aerosol mass and number after cloud processing. There is no mechanism that ensures that the coarse mode size is always larger than the accumulation mode size. The coarse mode may assume a smaller geometric mean diameter than the accumulation mode, e.g. due to depletion by precipitation.

d)I'm not sure authors discussed in enough details entrainment as a source of aerosol.

We have limited the discussion of entrainment in this work in acknowledgment of the fact that at the relatively coarse resolution of our simulations, entrainment is likely overestimated (Stevens et al., 2005). We have added an explanatory note and reference in section 4.3.3 (Sea salt emissions and entrainment from the free troposphere).

C4933

In this paper authors state that entrainment contribution is 50 % of that by the surface fluxes. However in Wang et al. 2010 apparently the contribution by entrainment was not enough to maintain boundary layer (open cell) circulation. Under what conditions aerosol from entrainment will contribute enough to keep open cell circulation?

In the open-cell cases by Wang et al. (2010), the free-tropospheric aerosol (all in accumulation mode) number concentration was 30 mg^{-1}, about 10 time lower than in this study. Even with the same entrainment rate, the contribution to MBL aerosol by entrainment is about 0.1 $mg^{-1} h^{-1}$. Without surface emission and new particle formation, this was obviously not enough to maintain the open-cell clouds. It is worth noting that the entrainment contribution in this study was estimated using passive tracers, which are not subject to removal during the 48-hour simulation. In reality, entrained aerosols are directly subject to cloud scavenging in the cloud layer, which may lower their accumulated contribution.

We will address the very important question on the importance of entrainment into the MBL in future studies. A generally valid answer to this question will be challenging to deliver, because of the numerous parameters to consider: Sea surface temperature/fluxes, background wind speeds, subsidence rate, aerosol number in the free troposphere, to name the most relevant. In addition, at the relatively coarse resolution of our simulations, entrainment is likely overestimated (Stevens et al., 2005). We have added an explanatory note and reference in section 4.3.3 (Sea salt emissions and entrainment from the free troposphere).

e) And related to the previous point: is aerosol nucleation from the gas phase and entrainment (without the sea salt emission from the surface) enough to keep open cell circulation? As authors state in the article it takes time to grow nucleated aerosol to the CCN sizes, as a result cloud may dissipate before nucleated aerosol reach CCN size.

The reviewer's assessment is supported by preliminary results of our simulations, but more work is needed before publication.

f) In 2.2 authors state that some reactions were removed from the WRF/chem. Can these reactions affect DMS → SO2 transition in the cloud (figure 10b and 10d).

The reactions that were removed from the scheme RADM2 chemical scheme involve volatile organic compounds (VOCs). These are not known to react with DMS. However, VOCs are oxidized by OH to peroxy radicals, which generate ozone in a cycle in which NO_x acts as a catalyst. The higher ozone may result in higher OH levels, because every ozone molecule can potentially produce two OH molecules. This would accelerate the conversion of SO_2 to DMS. However, this would require presence of NO_x at levels found in polluted air masses. 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), peroxy radicals from VOC oxidation are more likely to form the more stable peroxides rather than to contribute to generation of ozone. 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.

Text here has been added to section 2.2 of the manuscript to explain the choice of chemical scheme in more detail.

g)P4717,L3: It's not clear why authors decided to include removal of the nucleation mode aerosol by collision with drops, but neglected accumulation and coarse mode removal by this process.

We have added text in appendix A1 that explains the reason.

The rate coefficient for Brownian coagulation of accumulation and coarse mode particles with cloud droplets is significantly smaller than for Brownian coagulation of Aitken (nucleation) mode particles with cloud droplets. In addition, the accumulation and coarse mode particle concentrations inside clean MBL clouds are strongly depleted due to activation, while Aitken (nucleation) mode particles are commonly activated to

a much smaller extent. Since the coagulation rate scales with the product of the concentrations of the coagulating particle populations, it will be small for the accumulation and the coarse mode, and larger for the Aitken (nucleation) mode.

Technical corrections: 1)Authors should consider adding "Numerical simulations" in title and abstract (first sentence) to make it clear that paper is about numerical simulations of chemical and aerosol processes.

The title was changed to "Modeling chemical and aerosol processes in the transition from closed to open cells during VOCALS-REx". The numerical character of the simulations is now disclosed in the abstract and in the manuscript body.

P4688L11 Remove sentence "We introduce ...."; and change next sentence to "Results from the numerical model ...".

We have removed "... introduce the new features of the model and ...". We have kept the remainder of the sentence, which lays out the main focus of the study, namely the transition from a closed- to an open-cell state. The revised passage reads

"We conduct numerical simulations of the marine boundary layer in the transition from a closed- to an open-cell state. Results are compared with observations in the Southeast Pacific boundary layer during the VAMOS Ocean-Cloud-Atmosphere-Land Study Regional Experiment (VOCALS-REx)."

P4689,L15: Shouldn't authors refer Stevens et al. 2005b instead of 2005a?

The reviewer is correct. The correct reference is given in the revised manuscript.

P4690: Consider rewriting first paragraph starting from "Aerosol nucleation has been ...". First sentence in this paragraph is about nucleation and removal, second about emission and entrainment, third again about nucleation

Done.

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It also contains concept of "aerosol surface area" I'm not familiar with and a although it was used in Tomlinson (2007) I didn't find physical meaning of it.

Condensable molecules are removed from the gas phase by collisions with surfaces (cloud droples, ice crystals, aerosol). The total surface area is an indicator of how rapidly condensable molecules are removed from the gas phase: The smaller the total surface area, the slower condensation proceeds. At sufficiently low surface areas, when the production of the condensable molecules exceeds their condensation, their concentration can increase to a level at which they commence to stick to each other and form clusters. Growth of these clusters to a stable size is called nucleation. Hence, conditions with low aerosol surface area are considered conducive for nucleation.

However, the surface area is only an approximate measure of how rapidly condensable molecules are removed from the gas phase, because it does not account for the geometry and size of the particles, their surface properties, and the nature of the condensing molecules. An accurate measure is the condensation sink (see, e.g., Kazil et al., 2007), which, however, is commonly not observed. Therefore, "aerosol surface area" is often used informally in place of "aerosol condensation sink", in particular in the context of field measurements. We added a reference to Kulmala et al. (2001) in the revised manuscript which introduces the sulfuric acid condensation sink onto an aerosol size distribution.

P.4691, L9 Sentence "Photo-, gas ..." seems to be unfinished. Maybe "It follows photo-, ... and transport will play important roles in determining aerosol properties in the MBL".

The paragraph was replaced with

"The strength of MBL aerosol sources will be determined by several 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 on the

one hand, they suppress aerosol nucleation by increasing the condensation loss of nucleating molecules on the other. Entrainment of aerosol from the free troposphere depends on the stability of the inversion, and on the presence of enhanced aerosol concentrations above it, e.g. from long-range transport of pollution."

P4692,L7: Starting from "In recent years ..." to the end of the paragraph seems to be out of context in this place. Shouldn't this part be in Motivations?

This passage is meant as justification that we only work with oceanic emissions of sea salt, but neglect a possible organic component of the emissions. We have moved the passage to the "Uncertainties" section, where we also elaborate on the appropriateness of the approach.

P4697,L23: "Dry deposition ..", It's not clear what authors are trying to say in this sentence. It may be too technical and unnecessary.

We have changed this passage in the revised manuscript.

P4698,L23: In the table 1 there is no aerosol in coarse mode, and Aitken mode aerosol is specified only in the free troposphere. Please specify it in the text too.

Done.

P4700,L16: First 2 sentences in this paragraph are difficult to understand. What is H2SO4 condensational sink? Is it a transfer of the H2SO4 from the gas phase to the water droplets?

We have rewritten the paragraph. The condensation sink is a measure of how quickly molecules of a given species are removed from the gas phase by condensation onto surfaces, such as of aerosol particles, cloud droplets, or ice crystals (see, e.g., Kazil et al., 2007). Formally, the rate of condensation of H_2SO_4 can be written as

$$\left. \frac{d[\text{H}_2\text{SO}_4]}{dt} \right|_{condensation} = -[\text{H}_2\text{SO}_4]\sigma \tag{1}$$

where σ is the condensation sink. A reference was added in the revised manuscript to Kulmala et al. (2001) who introduce the condensation sink.

And where is sea salt emission on the figure?

The sea salt emissions time series is shown in Fig. 7. We have changed the paragraph in question so that the presented information is self-contained.

P4701,L15: Maybe ?... in the cloud free MBL top region?.

Done.

Later authors explain why SO2 within the cloud is low.

P47505,L24: Is it really true that there is more water in sizes d>40 um than in d<40 for the drizzling stratocumulus? Typically mean droplet diameter is less than 40 um.

Our main point is that under drizzling conditions, the King probe (range 5–40 μm in diameter) includes much less of the total liquid water (cloud plus rain) than does the 2D-C probe (range 25–800 μm in diameter).

We have reformulated the passage so that it reads as follows:

"In drizzling conditions, the 2D-C probe includes a more significant fraction of the total liquid water than does the King probe and so 2D-C data are used in the following."

b) It?s not clear why author discuss DMS and SO2 in different sections (5.3 and 5.4), at the same time having plots on the same figure.

The DMS and SO$_2$ plots have been split up in the revised manuscript.

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