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Atmospheric Chemistry and Physics Discussions

Interactive comment on "A GCM study of organic matter in marine aerosol and its potential contribution to cloud drop activation" *by* G. J. Roelofs

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We want to thank both reviewers for evaluating our manuscript and for their questions and comments. We have altered the manuscript accordingly. Most important changes are the addition of a brief comparison of model results with in situ measurements of CDNC and organic matter, an explanation of our assumption that the organic mass and DMS are emitted with similar distribution and seasonality. We will answer the questions of the reviewers and outline the changes made in the manuscript in detail below.

REFEREE #1

1) Referee #1 mentions that processes or issues other than marine organics may be (partly) responsible for the underestimation of CDNC in our previous simulation study

without marine organics (Roelofs et al., 2006; see manuscript for full reference). Referee #1 specifically mentions the updraft velocity. In the relatively coarse T21 resolution the simulated updraft velocities were found to be rather high compared to observations when using the relation between updraft velocity and TKE proposed by Lohmann et al. (J. Geophys. Res., 104, 1999). Therefore we applied a different relation that yields smaller vertical velocities, which produced realistic and representative simulated cloud drop effective radius (reff) over the NH continents. Simulated reff in marine regions was severely overestimated compared to observations, corresponding with a underestimation of CDNC up to a factor of three. In our model, marine CDNC is predominantly limited by CCN availability: the coarse and accumulation modes aerosol concentrations are much smaller than observed (Stier et al., 2005). Using a larger updraft speed, e.g., following Peng et al. (J. Geophys. Res., 110, 2005), would therefore not result in significantly larger CDNC in remote marine regions given this limitation and may even worsen the representativity over the continent. The Aitken mode aerosol is a potential additional reservoir for cloud drops but their activation requires relatively high supersaturation values that are not reached in the marine environment. In Roelofs et al. (2006) we concluded that specific emissions are missing from our simulation, or that the size distribution of aerosol species is not representative. In the present study we have investigated the first option, specifically the potential role of marine organics for CDNC in marine environments. It is an exploratory study: if the discrepancies were due to marine organic aerosol, what can we say about their abundance and size distribution? However, a qualitatively and quantitatively consistent theory for emissions of organics, chemical transformations, SOA production and solubility does not exists at present because many important parameters needed for this are unknown or highly uncertain. Further, our model is not equipped to explicitly calculate SOA formation and primary production of marine organics. Therefore we applied a top-down approach in which organics are added to specific aerosol modes. The simulated aerosol organic mass and size distribution is evaluated using the measurements of O'Dowd et al. (2004) and ISCCP cloud effective radius as constraint. We have added a discussion of other

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model uncertainties and inaccuracies, but these can not fully explain the discrepancies between simulated and observed CDNC and effective cloud drop radius. We remark that mixing/entrainment is not simulated in the cloud processing routine; the computed CDNC represents the amount of droplets activated at the cloud base.

2a) Referee #1 asks about the accuracy of remotely sensed reff, and for comparison of model results with in situ observations. We have used cloud drop effective radius (reff) from ISCCP to assess the representativity of the simulated influence of marine organic aerosol on marine clouds. Retrieved reff has a typical uncertainty of 2 micron (Han et al., 1994). The discrepancy between observed reff and reff from the simulation without consideration of marine organics is much larger, 6ś3 micron (see Figure 6) and can therefore not fully be explained by measurement uncertainties. When taking marine aerosol organics into account, the simulated reff is much closer to the retrieved reff, thereby demonstrating the potential importance of this aerosol specie. We have included a brief comparison of the simulated CDNC with in situ observations. In situ measurements of CDNC in North Atlantic clouds are presented in Bower et al. (2000; ACE2 HILLCLOUD June-July Tenerife; clean 50-300 cm-3, polluted $>$ 2000 cm 3; Borys et al. (1998; Tenerife June-July 1995-1996; clean ˜125 cm-3, poll: ˜ 785 cm 3); Leaitch et al. (1996; North Atlantic coastal Canada; 100 - 400 cm-3). Without consideration of marine organics the simulated CDNC over the NH Atlantic Ocean is between 10 and 60 cm-3, whereas with consideration of marine organics CDNC is between 50 and 200 cm-3. The latter is in much better agreement with the observations. Not many measurements of organic content of marine aerosol have been published. We have compared our results to observations close to Amsterdam Island (SH) that indicate concentrations of organics between 200-300 ng/m3 (avg: 220 ng/m3) for the bloom period, and between 50-100 ng/m3 (avg:85 ng/m3) for the season with low biogenic activity (J. Sciare, personal communication). These data agree with calculated aerosol organic mass derived by Meskhidze and Nenes (2006) as mentioned in the manuscript. Observations by Phinney et al. (2006) will be discussed in our answer to comment 3b. We remark that a more detailed validation of emissions and organic aerosol forma-

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tion is required which also involves precursor gases and the contributions of primary and secondary aerosol formation. This requires a detailed calculation of emissions, chemical processes and gas-to-particle conversion of which our model is not capable yet.

2b) The marine organics are emitted into the lowest model layer, and upward transport is obtained mainly through mixing within the boundary layer (BL). The model BL in the extratropical marine atmosphere is relatively stable so that detrainment of aerosol organics into the free troposphere is moderate. The model results show that the CDNC increase due to the added organics at altitudes below 1000 m is on average about threefold. The increase is significantly smaller at higher altitudes, about 10-20% at 2 km altitude. It must be noted that our model considers all organic aerosol to be primary. When also SOA formation is represented, the effect at higher altitudes may become larger due to transport of organic precursor gases out of the BL. Above three kilometres altitude large scale cloud occurrence is very rare.

3a) The referee requests more information on the treatment of organic matter in the model. We have included this information now in section 3.

3b) Phinney et al. (DSR, 2410-2433, 2006) compare in their Figure 11 the size-resolved aerosol chemical composition measured near Ocean Station Papa with that obtained by O'Dowd (2004). The figure shows that their fine mode organic fraction is smaller than from O'Dowd (˜20% vs. 60%). This is partly due to the fact that their absolute organic mass concentration is smaller, i.e. 0.3 vs. 0.62 micrograms/m3, but also partly to the fact that their sulfate mass is much larger, i.e., 0.75 vs. ˜0.25 micrograms/m3. Phinney et al. note that the 0.3 micrograms/m3 fine mode organics are obtained after subtracting direct ship plume emissions from the measured organic mass. Even though part of the remaining organics (hydrocarbon contributions) may still be attributed to ship emissions and advection of pollution, the remainder (oxygenated carboxylic acids) may be associated with organics emitted from the ocean, also considering the relatively high amounts of MSA. The relative contribution was not specified

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quantitatively by Phinney et al., so it is not possible to provide a quantitative answer. Nevertheless, their paper strongly suggests that the contribution by ocean organics to the marine aerosol mass is significant, which contrasts with the comment of the referee.

REFEREE #2

1) The reviewer asks for more comparison with in situ measurements. We refer to our answer on comment 2) from referee #1. We have not investigated the impact on aerosol optical depth, but we plan to look into this aspect further when simulations are carried out on a higher horizontal resolution with more representative transport patterns (see also the following comment).

2) The reviewer comments that the model resolution used in our study, T21, may be too coarse to reproduce a realistic cloud climatology. In our previous studies on tropospheric ozone transports we have found that transport patterns associated with synoptic disturbances are simulated more realistic on higher resolutions (T42, T63). However, a higher resolution also involves longer simulation times: increasing the spatial and temporal resolution by a factor of two leads to an eightfold increase in computational time. This was not feasible in our study, where all in all fourteen 4-year simulations were performed, a total of 56 years. Although the agreement between simulation results and observations is relatively good in our study, we agree with the reviewer that the influence of model resolution on the results is to be investigated, as we plan to do in the near future.

Minor comments:

1) HAM considers mass units (kg, g) for each aerosol specie. For the calculation of the Raoult effect in the aerosol activation parameterization, mass is transformed to molecules. The molar weight of oxalic acid is taken to be representative for the organic matter, and this defines the relation between molecules and mass. We have added this information to the manuscript.

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2) We have changed the terminology to "initial conditions" or "emission scenarios" to avoid misunderstanding of the term "boundary conditions".

3) An explanation of the assumptions regarding the organic solubility is included in section 3.

4) We must emphasize that although the actual size range for each mode in the aerosol model is fixed, the average particle size for each mode is not fixed and can vary within the range. In our simulations, the organic particles from marine origin directly influence the modal size so that aerosol mass, aerosol volume and number concentration are always consistent. For example, in Ax the same organic mass is added to the accumulation mode as in Ae, but it is divided over more and smaller particles. The effect is discernible in, for example, the particle lifetimes and aerosol mass, as mentioned in section 4.1

5) Recent measurements in Mace Head (Ireland) showed the presence of relatively large quantities of aerosol organics, ˜ 620 ng/m3 in summer, in air masses advected from remote marine regions (O'Dowd et al., 2004). The nature of the organic species indicated a primary marine source involving bubble bursting processes during phytoplankton blooms (Cavalli et al., 2004). Secondary aerosol formation, possibly involving isoprene, also affects marine aerosol. Isoprene emissions from the ocean surface were reported by Yokouchi et al. (1999) and Sinha et al. (2007), with phytoplankton as a likely source (Shaw et al., 2003). Given the relation between phytoplankton and ocean surface emissions of primary organic particles, isoprene and DMS, the seasonality of the marine organic emissions is assumed to follow the emission of DMS. This information is added to Section 3.

6) The error in predicting CDNC from the cloud processing parameterization is generally below 30% when compared to a cloud parcel model (Haenel, 1987). Due to the cubic relation between CDNC and cloud drop radius the resulting error in the radius is considerably smaller, ˜ 10% which is in the order of 1.5 micron. For comparison, ac-

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counting for marine organics in our study changes the computed seasonally averaged CDNC by a factor 2 to 4, and the cloud drop effective radius by 6ś3 micron (Figs 5 and 6). Even when the amount and spatial distribution of organic emissions are the same, the computed CDNC can differ up to about 50% depending on the mixing state of the organics (internal or external) or the size and number concentration of freshly nucleated particles (Fig. 5). Therefore the possible effects of aerosol organics from marine origin may well surpass the effect of uncertainties or inaccuracies in the calculation of CDNC. We have added this discussion in sections 2 and 5.

Interactive comment on Atmos. Chem. Phys. Discuss., 7, 5675, 2007.