Atmos. Chem. Phys. Discuss., 1, S40–S43, 2001
www.atmos-chem-phys.org/acpd/1/S40/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Modelling the contribution of sea salt and dimethyl sulfide derived aerosol to marine CCN" *by* "Y. J. Yoon and P. Brimblecombe"

Anonymous Referee #2

Received and Published: 28 September 2001

General comments:

The strength and relative importance of various natural CCN sources in a marine environment has been investigated using a model approach. While this is by no means a new research issue, the authors bring up some new important aspects that can be considered worth publishing. The manuscript is well written and easy to follow, but not entirely scientifically sound. My major criticism concerning this latter point is presented below in more detail.

Specific comments:

(1). As shown in several recent publications (e.g. Katoshevski et al., 1999; Pirjola et al., 2000; Capaldo et al. 1998, JGR104, 3483-3500), marine CCN production depends

Print Version
Interactive Discussion
Original Paper

strongly on the dynamics of particles smaller than about 100 nm in diameter. The aerosol dynamical model used in this manuscript relies on that developed by Pandis et al. (1994) and Russell et al. (1994), having only one size section in the particle size range $<$100 nm. Because of this, important information concerning the dynamics of nucleated (or entrained) particles is lost. First,all nucleated particles are immediately inserted into the first size bin (covering the range 30-100 nm) of the model. In reality, the growth from 1 nm (a typical size for newly-formed particles) to 30 nm may take more than a day in a marine boundary layer, during which time these particle could be scavenged away by pre-existing larger particles. Second, condensation of sulfuric acid (and other condensable material) from the gas phase to the particles in the first size bin leads some transfer of particles from this size bin to the second size bin ($>$100 nm), thus producing new CCN. In reality particles in the first size bin may be quite too small to become new CCN by condensation. The apparent uncertainties arising from the structure of used model should be brought up much more clearly in the manuscript.

(2). The authors try to separate three natural sources for CCN in the marine boundary layer: (A) sea-salt emissions, (B) MBL nucleation associated with DMS emissions, and (C) FT entrainment. The natural component of the third source (which actually is investigated here) is also associated with DMS emissions (although not necessary with as regional emissions as in the case of the second source). In my opinion, sources (B) and (C) cannot be treated independently of each other. Based on the simulations made in the above-mentioned papers, the most plausible scenario for non-sea-salt CCN in the MBL appears to be the following: most of these particles come from the FT, in which they have been formed as a result of DMS oxidation, nucleation, and subsequent growth. However, when entraining to the MBL, most of these particle are too small to act as CCN. The sub-CCN-size particles accumulate sulfate (plus additional material) in the MBL by condensation and cloud processing, becoming eventually CCN unless scavenged by precipitation. In this scenario, FT entrainment and MBL chemistry together act as a source of new CCN, since without the other very few new CCN would be produced. Based on this and the very simplified structure of the model applied in

ACPD

1, S40–S43, 2001

Print Version

Interactive Discussion

Original Paper

this work, it is clear that this manuscript cannot give any specific answer on the relative importance of MBL nucleation and FT transport as sources for CCN. I suggest that the authors concentrate on distinguishing between sea-salt and non-sea-salt CCN, and are very careful about what they say on the relative importance of MBL nucleation and FT transport.

(3). In the sensitivity tests, only two things has been taken into account: the sulfuric acid mass accommodation coefficient and the entrainment processes. Several other parameters should be investigated, including (A) the DMS emission flux (the uncertainty in current DMS flux parameterizations is a factor of about two, as demonstrated by Kettle and Andreae (2000), JGR 105, 26793-26808), (B) the yield of SO2 in the atmospheric DMS oxidation (the exact value of this yield has not been established), and (C) the uncertainty in the magnitude of various SO2 sinks (deposition to sea surface and heterogeneous oxidation in sea-salt particles, both of which reduce the amount of gaseous sulfuric acid that can be produced from SO2). The authors might construct a simple table in which the results of various sensitivity tests have been summarized.

(4). It is not clear to me why the authors have used the value of 0.02 when testing the sensitivity of the system to the sulfuric acid accommodation coefficient. First, values below 0.1 seem unrealistic based on the few direct measurements reported recently (Jefferson et al. (1997); Pöschl et al. (1998) J. Phys. Chem. 102, 10082-10089). Second, as seen by the model results (Figure 5b), the value of 0.02 leads to gaseous sulfuric acid concentrations which are too large compared with direct measurements in marine boundary layers (see various papers by Weber and co-workers).

(5). The authors give practically no information on how they have specified the FT aerosol and trace gas characteristics when investigating the influence of entrainment processes. More detailed information in this regard should be provided.

Technical comments:

Page 9, first paragraph in section 2.3. The paper by Turner (1996) mentioned here cannot be found from the reference list.

Calling all particles $<$100 nm in diameter as a "nucleation mode" is somewhat old-

ACPD

1, S40–S43, 2001

Print Version

Interactive Discussion

Original Paper

fashioned. A term "Aitken mode" or, in this case, an "Aggregate of the nucleation and the Aitken mode" should be used instead.

Interactive comment on Atmos. Chem. Phys. Discuss., 1, 93, 2001.