Atmospheric Chemistry and Physics Discussions
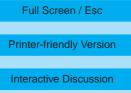
Interactive
Comment

Interactive comment on "Examining aerosol indirect effect under contrasting environments during the ACE-2 experiment" *by* H. Guo et al.

H. Guo et al.

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We thank anonymous referee #3 for many useful comments and suggestions. We will incorporate the corrections and modifications in the revised version.

My only real concern is with the incomplete aerosol observations near the cloud during ACE-2. This provides some wiggle-room for validating the cloud simulation, so I would like to see some discussion of how much the validation data was used to constrain the aerosol properties. In particular, to what extent was the composition and size distribution of the pre-existing aerosol adjusted to improve agreement with the measured droplet size distribution? Otherwise I congratulate the authors on a fine study and clear presentation.

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For the aerosol number concentration, Guibert et al. (2003) conducted intensive comparisons among surface measurements, and three airborne measurements (M-IV, Pelican, and C-130) below and within cloud layers. They showed that these measurements agreed to within reasonable uncertainties. For the clean case on June 26, 1997, the surface measurement of condensation nuclei (CN) was 226 mg^{-1} and the airborne measurement from M-IV was 200 mg^{-1}. For the polluted case on July 9, 1997, the surface and M-IV measurements were 608 mg^{-1} and 611 mg^{-1}, respectively. Moreover, the difference among the surface and different airborne measurements was generally $< 10\%$ for the above 2 cases.

For the aerosol chemical mass concentration, Putaud et al. (2000) showed that the sum of species concentration from chemical analyses was consistent with the aerosol mass concentrations obtained from the particle size distribution within the experimental uncertainties of 47% within the marine boundary layer.

We used these measurements to constrain our assumed aerosol number concentration and chemical mass concentration, and did not attempt any tuning for the particle size distribution.

We used a simple aerosol nucleation scheme following Chuang and Penner (1995), which assumed internally mixed aerosols. The aerosol size distribution was determined by the condensation of sulfuric acid vapor (H2SO4) on a prescribed pre-existing particle distribution and by aqueous-phase oxidation of SO2 followed by the evaporation of cloud drops. Furthermore, it was assumed that 75% of the mass fraction of sulfate (nss-sulfate) was produced by the aqueous phase pathway, which was consistent with the estimate by Langner and Rodhe (1991).

The prescribed pre-existing particles for the marine cases followed a three-mode log-normal distribution with the mean diameters at 0.03, 0.15, and 0.5 μm (Chuang and Penner, 1995). After the addition of sulphate, the sulphate-containing aerosol size distribution would grow

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to larger sizes. The resulting aerosol size distribution is close to the observed aerosol size distribution that had prominent modes with diameters at 0.05, 0.16, and 0.5 μm for both the clean and the polluted cases (Snider et al., 2003).

This discussion and references will be added to the paper on pages 11566-11567 (2.1 Case descriptions).

Reference:

Chuang, C., and J. Penner, Effect of anthropogenic sulfate on cloud drop nucleation and optical properties, Tellus, 47, 566-577, 1995.

Guibert, S., J.R. Snider and J.-L. Brenguier, Aerosol activation in marine stratocumulus clouds 1. Measurement validation for a closure study, J. Geophys. Res., 108(D15), 8628, doi:10.1029/2002JD002678, 2003.

Langner, J. and H. Rodhe, A global three-dimensional model of the tropospheric sulphur cycle, J. Atmos. Chem., 13, 225-263, 1991.

Putaud, J. P., R. Van Dingenen, M. Mangoni, A. Virkkula, H. Maring, J.M. Prospero, E. Swietlicki, and et al., Chemical mass closure and assessment of the origin of the submicron aerosol in the marine boundary layer and the free troposphere at Tenerife during ACE-2, TELLUS, 52B, 141-168, 2000.

Snider, J. R., S. Guibert, J.-L. Brenguier, J.-P. Putaud, Aerosol activation in marine stratocumulus clouds: 2. Kohler and parcel theory closure studies, J. Geophys. Res., 108(D15), 8629, doi:10.1029/2002JD002692, 2003.

Minor comments.
1. Page 11563, line 8. Change "values" to "value".

This will be fixed.

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2. page 1156, lines 18-20. What were the aerosol and sulfate concentrations above the boundary layer for these cases?

There were sub-micron aerosol chemical composition measurements at the Izana Observatory (IZO) at 2370 a.s.l. (geometric altitude above sea level). For the clean (June 26) and polluted (July 9) cases the average nss-sulphate concentrations were 0.15 ± 0.02 and 0.61 ± 0.03 $\mu g/m^3$ (Putaud et al., 2000). However, it should be noted that these samples were only collected during night time only (20:00-8:00 UTC), because during the daytime surface thermal winds brought the polluted low-altitude air up.

The aerosol concentrations above the boundary layer for both the clean (June 26) and polluted (July 9) cases from the M-IV airborne measurement were about 400 cm^{-3} or 400 mg^{-3} (Guibert et al., 2003).

Also, an analysis of the entire campaign showed that the condensation nuclei (CN) mixing ratio values above the boundary layer were relatively constant for most cases, although the values measured in the boundary layer changed significantly depending on the origin of the boundary layer air mass (Guibert et al., 2003). This implies that the free troposphere was decoupled from the boundary layer, which is consistent with the results of Verver et al., (2000) who analyzed back trajectories.

This discussion will be added to the manuscript.

Reference:
Guibert, S., J.R. Snider and J.-L. Brenguier, Aerosol activation in marine stratocumulus clouds 1. Measurement validation for a closure study, J. Geophys. Res., 108(D15), 8628, doi:10.1029/2002JD002678, 2003.
Putaud, J. P., R. Van Dingenen, M. Mangoni, A. Virkkula, H. Maring, J.M. Prospero, E.

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Swietlicki, and et al., Chemical mass closure and assessment of the origin of the submicron aerosol in the marine boundary layer and the free troposphere at Tenerife during ACE-2, TELLUS, 52B, 141-168, 2000.

Verver, G., F. Raes, D. Vogelezang and D. Johnson, The 2nd aerosol characterization experiment (ACE-2) meteorological and chemical context, TELLUS, 52B, 126-140, 2000.

3. page 11566, lines 23-26. What was the assumed size distribution of the 25% of the sulfate that was no produced by aqueous chemistry? What was the assumed composition of the pre-existing aerosol? You might mention the lack of aerosol composition measurements to better constrain this assumption.

The 25% of the sulfate that was no produced by aqueous chemistry was assumed to be from the condensation of sulfuric acid vapor (H2SO4) on a prescribed pre-existing particle distribution. The prescribed pre-existing particles for the marine cases followed a three-mode log-normal distribution with diameters at 0.03, 0.15, and 0.5 m (Chuang and Penner, 1995).

This discussion will be added to the manuscript.

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