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Interactive Comment

Interactive comment on "Comprehensive airborne characterization of aerosol from a major bovine source" *by* A. Sorooshian et al.

A. Sorooshian et al.

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We appreciate the feedback in this short comment and as a result we have made appropriate additions in the manuscript. We provide more text in the experimental description of the CCN instrument and have also added an appendix with four figures to provide additional details regarding calibration, operation, and data analysis. Our response to the comment follows the original comment below.

Original comment: Comprehensive airborne characterization of aerosol from a major bovine source” by Sorooshian et al. is a very interesting study, and we would like to compliment the authors on their achievements. The paper presents a comprehensive set of aircraft measurements, including quantitative data on aerosol particle hygroscopicity and cloud condensation nucleus (CCN) activity. With regard to the collection and presentation of CCN data, however, we would like to ask for clarification of the measurement procedures and results along the following lines. Water vapor supersaturation is the primary variable inducing and determining the CCN activation of aerosol particles in the atmosphere as well as in CCN field measurements and laboratory experiments. With regard to measuring CCN at certain water vapor supersaturations, Rose et al. (2008) have recently demonstrated that: (a) The effective water vapor supersaturation in a continuous-flow CCN counter (DMTCCNC) as applied in this study depends strongly on the operating conditions (pressure,temperature, flow rate) and needs to be carefully calibrated with reference aerosols and Köhler model calculations. (This point was clear before Rose et al. 2008, but it seems to be frequently overlooked and may not be known to all scientists interested in CCN data.) (b) The applicability of flow models for calculating the water vapor supersaturation in such instruments also depends on experimental calibration (non-ideal temperature offset, etc.), and the model results can deviate substantially from measurement results, especially at low supersaturation (up to 40% and more). (c) The water vapor supersaturation values inferred by Köhler model calculations depend strongly on the type of Köhler model and on the water activity parameterization and other parameters applied in these calculations. Specifically, the critical water vapor supersaturation values calculated for ammonium sulfate particles, which are usually used for reference and calibration, can deviate by up to 20% or more from the most accurate models available. Therefore, the CCN measurement results presented in Sorooshian et al. (2008, Tab. 3, Figs. 16 and 17) depend strongly on the operating conditions and calibration of the CCN counter. Presumably, the normalized data presented in Fig. 17 depend also on the Köhler model used for the reference substance ammonium sulfate. The manuscript, however, provides practically no information about these aspects. It is not clear how the pressure, temperature and flow rate were adjusted and kept constant during the flights, and no uncertainty estimates are given for the nominal supersaturation values. Without such information it is hard to appreciate the reliability of the presented results and to compare them with other data (e.g., with regard to the influence of flow rate on CCN activation and droplet growth kinetics). Thus, we suggest to include more information about the determination, regulation, and uncertainty of water vapor supersaturation in the applied CCN counter and to specify which Köhler models have been applied. From the manuscript we understand that the authors intend to prepare and publish a more detailed paper on the CCN measurements. Nevertheless, we think that a minimum of technical information and/or uncertainty indicators should be supplied whenever new data are presented and discussed. U. Pöschl, D. Rose, S. Gunthe and M. O. Andreae Reference: Rose, D., Gunthe, S. S., Mikhailov, E., Frank, G. P., Dusek, U., Andreae, M. O., and Pöschl, U.: Calibration and measurement uncertainties of a continuous-flow cloud condensation nuclei counter (DMT-CCNC): CCN activation of ammonium sulfate and sodium chloride aerosol particles in theory and experiment, Atmos. Chem. Phys., 8, 1153-1179, 2008.

Response to Short Comment:

The Continuous-flow Streamwise Thermal Gradient Chamber (CFSTGC) used in this study was calibrated using ammonium sulfate particles classified with a Differential Mobility Analyzer (DMA) as follows. An aqueous solution of ammonium sulfate is atomized via a collision-type atomizer operated at 4 psig. The droplet stream was passed through multiple diffusional driers (operating at 5-10% RH), resulting in a polydisperse dry aerosol (with modal diameter ˜ 50 nm), which was sent through a DMA (TSI 3081L) operated with a sheath-to-aerosol flow ratio of 10:1. The classified aerosol was then sent to a Condensation Particle Counter (TSI 3010 CPC) and the CFSTGC (operated at constant flow rate, temperature gradient, and pressure) to measure the total aerosol concentration and the total activated particles, respectively. The level of supersaturation in the instrument is determined from the minimum dry particle diameter, Dp50, that activates in the instrument, found by plotting the ratio of CCN to CN concentration as a function of mobility particle diameter. A sigmoidal fit to the data (neglecting the effects of multiple particle charging, which is minor for the aerosol used) then determines Dp50 as the dry diameter for which the CCN/CN=0.50. The instrument supersaturation is related to Dp50 by applying Köhler theory assuming that ammonium sulfate has a density of 1760 kg m-3, surface tension of water, molar mass of 0.132 kg/mol and an effective van't Hoff factor of 2.5, since studies have found the value to lie between 2 and 2.5 (Gerber et al., 1977; Brechtel and Kreidenweis, 2000; Pradeep Kumar et al., 2003; Rose et al., 2008). The standard deviation in the supersaturation was determined from the standard deviation observed in the Dp50. The error bars in the temperature difference are 1 sigma of the observed variation during the calibrations. Droplet diameter of activated CCN at the exit of the growth column is also measured; in this study, the growth rate of ammonium sulfate particles is characterized by the average size of droplets formed from particles with dry diameter equal to Dp50.

The instrument was calibrated numerous times throughout the study for supersaturations ranging from 0.1 to 1.4% SS, for pressures between 700 and 1000 mb, and for flow rates ranging between 500 and 1000 cm3/min. The resulting calibration curves are presented in new figures placed in Appendix A (Fig. A-1 and A-2) (the average size of activated droplets, together with 1 sigma error bars, are shown in Figures A-3 and A-4).

The instrument was not operated at constant pressure; instead, the pressure was allowed to vary with altitude. As a result, some time is required for the instrument supersaturation to adjust every time altitude changes; the data used in the closure study was filtered for transients resulting from changes in instrument pressure and temperature gradient. The calibration curves are then interpolated with respect to temperature gradient and pressure to determine the in-flight supersaturation.

References:

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Interactive comment on Atmos. Chem. Phys. Discuss., 8, 10415, 2008.