Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2018-475-RC1, 2018

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Is there an aerosol signature of cloud processing?" *by* Barbara Ervens et al.

Anonymous Referee #1

Received and published: 30 June 2018

The manuscript investigates the impact of sulfate and aqueous secondary organic aerosol (aqSOA) formation through cloud processing on relative aerosol mass increase, aerosol hygroscopicity, and organic aerosol oxygen content, focusing first on aerosol bulk properties and then on size segregated properties. Different air mass categories are studied, using measurements collected during the SEAC^{4}RS experiments on board the NASA DC-8, based out of-Houston. Simulations indicated that the impacts of cloud processing are more prominent on polluted air masses than clean background air masses, but less evident in heavily polluted conditions, such as in biomass burning plumes. One of the main implications of this work is that changes in aerosol mass and particle hygroscopicity are better indicator of cloud processing than chemical markers, due to their chemistry sinks. Nevertheless, depending on the initial proper-

Printer-friendly version

Discussion paper

ties of air masses, the impact of cloud-processing on the above-mentioned parameters might vary significantly, making sometimes difficult to detect aqueous phase processing, especially under clean conditions or in heavily polluted air masses.

Cloud processing is attracting a growing interest from both the observation and modeling research communities, due to its impacts on air quality and climate. Often field observations struggle to identify aqueous phase processing, which is observed instead during laboratory experiments, delaying its description in chemistry transport models. The present manuscript helps to explain some of the discrepancies among laboratory observations, field observations, and modeling results, and deserve publication in ACP with minor revisions.

General comments: The completeness of data collected during the NASA DC-8 experiments seems to be only partially deployed. For example, the HR-AMS data collected during the flights could be used to characterize the initial O/C and k parameters. For example, the model assumes that k_{org} is equal to 0.1, while Jimenez et al. (2009) shows that, depending on the organic oxygen content the k_{org} can vary from about 0 up to 0.2. Please explain if the use of specific k_{org} for different air mass types could have supported a more accurate analysis and justify why the authors decided to use a constant k_{org} for different masses.

Some authors observed that aqSOA both from dark-phase chemistry and OH reactions are characterized by optical properties typical of brown carbon (Laskin et al., 2015). Do the authors think that optical properties can offer further insights into cloud processing? Even though the chemistry model employed might not be able to simulate optical properties, it would be advisable to mention it, at least in the introduction, as a potentially additional tracer for aqueous phase processing.

Specific comments: Page 3: Do the authors think that back-trajectory analysis could complement the use of specific molecular tracers for the identification of specific air mass types? In addition, the backtrajetcory analysis could give an estimate of the age

of polluted air masses, to investigate the impact of fresh and aged emissions on aqSOA formation and their properties.

Table 1 could report the variability range of measurements to give an idea about the significance of differences among air mass types.

Page 10 line 337: The authors report that the size resolved composition measurements from the field experiments were noisy. Please specify if ere hygroscopicity and O/C ratios used as model input were assumed to be constant across the different size bins for the different air mass categories. In such a case, what can be the uncertainty associated with this assumption?

Figure 2: The relative mass increase calculated through equation 2 is derived for each single size bins? In such a case it is not clear why for a few size bins the dM/dlogD values are smaller than the unprocessed values, even if the relative percentage increase is significant (larger than 50

Technical comments:

Page 4 line 118: "4" in SEAC^{4}RS as apice Page 8 line 250: did the authors mean “the sharp peak when SO2 is added”? Page 8 line 276: remove B after period. Page 9 line 300: stopped Page 10 line 339: Figure 2 instead of figure 1 Page 11 line 381: Figure 2 instead of figure 32 Page 15 line 509: in addition to oxalate, authors could mention also hydroxyl methane sulfonate as a tracer of aqSOA with additional chemical sinks, like oxidation under high O3 concentration (Whiteaker and Prather 2003). These sinks set some limitation on its use as a proxy for aqueous phase processing.

References: Jimenez et al., Evolution of Organic Aerosols in the Atmosphere, Science 326, 1525 (2009). Laskin et al., Chemistry of Atmospheric Brown Carbon, Chemical Reviews, 115, 4335 (2015). Whiteaker and Prather, Hydroxymethanesulfonate as a tracer for fog processing of individual aerosol particles, Atmospheric Environment, 37, 1033 (2003).

Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-475, 2018.

Printer-friendly version

Discussion paper