Atmos. Chem. Phys. Discuss.,
doi:10.5194/acp-2017-23-RC1, 2017

Atmospheric Chemistry and Physics Discussions

Interactive comment on "In situ chemical measurement of individual cloud residue particles at a mountain site, South China" *by* Qinhao Lin et al.

Anonymous Referee #1

Received and published: 20 March 2017

Lin et al describe measurements of the chemistry of cloud droplet residues at a mountain-top site in South China in January 2016. Few measurements of cloud droplet residual chemistry exist, so these are important measurements to help improve our knowledge of cloud formation and properties, which are important for predicting weather and climate. Detailed comments follow.

It would be helpful for the authors to provide additional information about the cloud events. Please provide, at minimum, ambient temperature during the cloud events to justify the presence of cloud droplets only and no influence from ice crystals would be useful to discuss (related to Line 123); this is shown in Fig 1 and would be useful to refer to earlier to justify the presence of cloud droplets only. Is it correct that measurements

Printer-friendly version

Discussion paper

of cloud droplet size were not completed during this study? It would be helpful to know what fraction of the cloud droplet population was sampled, given the cut size of 8 um. It is stated on Line 135 that previous studies found an average cloud droplet size of ~10 um at this site, but the distribution is not discussed, nor is the time of year of the previous measurements. Since this work is published in Chinese, these information are not easily obtained by the reader. So, additional discussion would be helpful. For interpretation of the comparison between the cloud droplet residues and ambient particles, it is important to understand what fraction of the cloud droplets were measured. Previous studies (e.g. Bator & Collett 1997, J. Geophys. Res.) have found that cloud chemistry varies with droplet size. Sampling only the larger cloud droplets may also bias the cloud droplet residue size to larger particles, which is one of the observations. Since the cloud droplet activation process is also size-dependent, it is not possible for the reader to evaluate measurement vs droplet activation size dependencies currently.

Major Comments:

Lines 20-23, lines 220-221, Figure 4, & numerous other locations: Do these number fractions take into account the size bias in the instrument inlet transmission efficiency, which is clear in Figure S3? It is clear that there are particle size dependencies to the cloud residual chemical composition, particularly for the amine and aged EC particle types, that should be considered when reporting fractions. For example, even on lines 220-221, it is not clear if the authors are reporting 3.8% of the total cloud residues, or 3.8% of the particles measured from 0.7-1.9 um, or 3.8% across each of the size bins from 0.7-1.9 um.

Lines 64-68: Only two other cloud studies are mentioned, or referenced, here, which incorrectly suggests to the reader that the measurement of anthropogenic particles in clouds has only been measured twice. While not all papers need to necessarily be referenced here, it is important to provide a comprehensive view to the reader.

Lines 87-89: This list is not comprehensive and is missing many papers. The authors are strongly suggested to conduct a detailed literature search, as comparison of their results with these papers is important.

Line 205 & Figure 3: Provide possible ion marker formulae here. Provide a reference for the aged EC type based on comparison to other single-particle mass spectrometry studies (e.g., reference to Moffet and Prather 2009, PNAS). Given the strong K+ signal here, it is likely that both the “Aged EC” and “K-rich" are from biomass burning. Similarly, the OC particles are likely aged biomass burning particles as well, given the strong K+ signal.

Lines 209-210: The prevalence of wildfires, shown in Figure 2, suggests that these particles are primarily from biomass burning (see Pratt et al. 2011, ACP for a single-particle mass spectrum of fresh biomass burning particles). Pratt et al. (2010, J. Atmos. Sci.) and Hudson et al. (2004, J. Geophys. Res.) discuss the identification of aged biomass burning particles by single-particle mass spectrometry. It seems that the authors can say with greater certainty the source of these particles, and discussion of this would elevate the paper by providing another evaluation of the influence of biomass burning aerosols on cloud activation, which is important and interesting.

Lines 221-227: It is not clear if Roth et al. observed a decreased fraction of amines in the clouds compared to the ambient, or if amines just do not influence the site. The presence and behavior of amines would be location and season dependent, so this discussion is not clear and seems to be comparing studies without considering commonalities.

Lines 244-247: To further understand the source of the Na-rich particles, were they present for both coastal and non-coastal wind directions? Is there a difference in the average mass spectra for these wind directions considering the minor peaks? Are industrial sources located in both directions?

Lines 263-267: While ammonium nitrate does not contribute to the dust nitrate ob-

served here, it is not appropriate to generalize this statement to suggest that previous studies in other very different locations did not measure ammonium nitrate.

Lines 319-329: Was there a mass spectral difference in the K-rich particle type between the N and SW wind directions that would aid in source identification for the two air masses?

Section 3.6: This authors should consider incorporating this discussion into the other sections of the manuscript so that comparisons are made when results are discussed. In addition, the authors should consider differences in atmospheric composition at the various sites when discussing specific comparisons (i.e. are the contributing sources and magnitudes the same, or may this be a reason for differences? Or, are the seasons the same?). Currently only a general statement on lines 431-433 states that differences are specific to geographic location. As noted above, the literature cited is also not comprehensive, and this section would benefit from additional literature searching.

Figure 4: It would be useful to add comparisons to the ambient and interstitial particles here. In addition, since the cloud residue types changed significantly based on air mass origin (N vs SW), as stated in Section 3.4, it would be useful to separate out these wind directions and show the fractions of cloud residue, interstitial, and ambient, separated for the two wind directions.

Technical Comments:

Lines 24-26, 45, & other locations (search document): Please clarify what is meant by "intensity" in these statements.

Lines 29-30: The phrase "To estimate how atmospheric aerosol particles respond to chemical properties of cloud droplets" is not clear since aerosols determine cloud droplet chemistry, outside of aqueous processing from dissolved trace gases.

Lines 41-42: For readers not familiar with the region, it would be helpful to know the suggested source of the amine particles, which the authors are presumably referring

to with respect to the wind direction change.

Line 44: What does “highly associated” mean in this context? This phrase occurs in other locations in the manuscript as well.

Lines 76-80: This sentence incorrectly cites Pratt et al 2010a for cloud droplet residues. The authors should also consider Drewnick et al 2007, J. Atmos. Chem., who did observe lower sulfate mass fractions for droplet residues compared to ambient aerosol. Pratt et al 2010 (J. Geophs. Res.) shows increased mixing with sulfate/nitrate for liquid clouds, compared to ice clouds, as another example.

Line 161: It should be clarified that “cloud droplet residue concentrations”, not “cloud droplet concentrations” were measured by the SMPS. This is an important distinction.

Lines 163-165: Wouldn’t hazy days with low visibility be characterized by high, rather than low, PM2.5 concentrations? This is confusing.

Line 165: Change “rainy” to “rain”.

Line 216: It is important to consider the relative enhancement in amine peaks when using a 266 nm laser and that the amines themselves may not comprise the majority of the particle mass. See Pratt et al. 2009, Environ. Sci. Technol.

Lines 230-232: It should be clarified when discussing previous vs the current study, and if previous studies are being discussed, the season should be noted if there are seasonal variations.

Lines 231-237: Some grammar fixes are needed here.

Line 279: Please be more specific with the statement “plays a key role in cloud processes”. Do you mean that these particles were preferentially activated? Is there evidence of this?

Lines 358-396: The phrasing in these paragraphs should be improved for greater clarity and correct grammar.

Line 390: This is an important finding, yet the phrasing “sulfate was observed to diminish” is not clear, particularly when considering the following sentence. Please clarify.

Lines 395-396: What discrepancy? This is not clear.

Line 397: Quantitatively, what is “no remarkable change”? The phrase “remarkable change” is used elsewhere in the manuscript as well, but it isn’t defined.

Table 2: Not sure what the authors mean by “way” here.

Figure 2: The lines and numbers on this map are difficult to read. It would be useful to make the text bold perhaps and increase the width of the lines.

Figure 3: This plot is difficult to read. The authors should consider showing only the most abundant (and discussed) particle types in the main text figure and moving the others (including “Other”, which is somewhat meaningless as an average mass spectrum if it is made up of a diverse population of particles) to the supplemental information.

Figures 7 and 8: Please indicate whether positive peak areas indicate preferentially in the cloud residues and negative indicate preferentially in the ambient/interstitial particles. This is currently not clear in the figure captions.

Figure S4: It is not clear in the maps where RH < and > 90% are located.

Interactive comment on Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-23, 2017.

Printer-friendly version

Discussion paper