

Review of “In situ chemical composition measurement of individual cloud residue particles at a mountain site, South China (revised version)” by Lin et al.

### Summary and general comments

The authors present the results of online physico-chemical characterization of cloud residuals ( $> \sim 8 \mu\text{m}$  in diameter), cloud particles ( $< 2.5 \mu\text{m } d_a$ ) and ambient particles ( $< 2.5 \mu\text{m } d_a$ ) from the field study conducted in January, 2016 in a remote mountainous area of South China. Based on the dataset of the eight major particle types classified by using the SPAMS data obtained during several cloud events, the authors report some unique properties of cloud residuals, such as chemical composition, size and mixing state (that may be inherently related), presumably occurred during an aerosol's atmospheric aging and cloud processes.

The topic itself is an important addition to ACP. Overall, the authors conducted a careful study as well as rigorous data analyses to generate new results regarding cloud residuals that would be potentially valuable in the cloud microphysics research community. However, such care was unfortunately not taken in the preparation of the manuscript, with the manuscript containing a number of errors and typos. Although I do not have any major scientific concerns, I have numerous technical comments (including but not limited to) as listed below. I would urge the authors of the manuscript to thoroughly proof read their manuscript as this list gets too long.

### Specific comments

P4 L64-P5L78: This part is not well written and poorly structured. The authors need to logically address why it is particularly important to study the aerosol mixing state rather than just focus on other general properties, such as size (e.g., Dusek et al., 2006, Science; Sotiropoulou et al 2006, Aerosol Science & Technology) and bulk composition (e.g., Wiedensohler et al., 2009, JGR; Twohy and Anderson, 2008, Environmental Research Letters), to improve our understanding of CCN activation. Aerosol mixing state indeed influences the ability of aerosol to act as CCN (e.g., Wang et al., 2010, Aerosol Science & Technology). For instance, Medina et al. (2007, JGR) estimated that internal mixing assumption resulted in a 35% over prediction of CCN concentration in one study for semi-urban settings. However, the relative importance of the mixing state as compared to other properties appear to vary depending on the proximity to the pollution plume source and/or photochemical ageing activity as a function of oxidant concentrations (Ervens et al., 2010, ACP). Hence, additional detailed measurements to characterize the timescale and effect of the aerosol mixing state on CCN properties of particles are by all means needed to improve our theoretical understanding of CCN activation. Please do not copy and paste the reviewer's comments in the manuscript. The authors may want to do a careful and thorough literature review, digest the contents in a diplomatic manner and describe your thoughts to the reader along with your own story line. I also suggest the authors to address up-to-date information of lab studies regarding the effect of the aerosol mixing state on CCN ability or droplet activation (e.g., Wang et al., 2017, ACPD-2017-454 and references therein; Broekhuizen et al., 2004, GRL; Abbatt et al., 2005, Atmospheric Environment; Shilling et al., 2007, Journal of Physical Chemistry A). Explaining how your field study would potentially shed light on lab works, vice versa, may strengthen the paper.

P7L125-126: The authors state that the observed clouds contained only liquid droplets. This statement is speculative since the ambient temperature seemed to be below  $-7 \text{ }^\circ\text{C}$  at some point of the field campaign (P7L125), which could trigger heterogeneous ice nucleation of some materials contained in dust aerosols (e.g., Atkinson et al., 2013, Nature). Further scientific backing seems necessary for the reader to understand the cloud properties.

P7 L126-128: Was the ambient inlet coupled with a dryer downstream? Please clarify.

P7L130-131: “Therefore, it is reasonable to select...” - I disagree with this statement. Correctly, the authors presumed the droplet size to be larger than  $8 \mu\text{m}$ . This assumption should be stated in the manuscript.

P7L130-131: There have been observations of droplet size-dependant chemical composition in clouds (Moore et al., 2004, Atmospheric Environment). Please discuss it in the manuscript.

P7L131-134: What gas (i.e., dry synthetic air, nitrogen, etc.) was used to create the counterflow? What is the background aerosol concentration through GCVI (i.e., the measurement with the counterflow only) in this study? In addition, can the authors at least provide the estimate of the number fraction of residuals to total particles relevant to your study (i.e., CCN active fraction)? Did the author measure the total cloud particle concentration through GCVI without any counterflows at some point?

P7L134-135: The particle transmission efficiency of residual sampling instruments is size-dependant. Did the authors take it into account for your analyses or apply 50% loss throughout the analyses? Please clarify. It is not clear if the size-dependence is incorporated in your analyses (P11L225-228). Just citing a paper seems not enough to justify it.

P7L139: Why do the authors define what comes through an ambient inlet (2.5 micron 50% cut-off) as "non-activated" particles? Numerous lab and field studies show that submicron particles can be activated to <2.5 micron.

P9L165-166: What about the transmission efficiency? Was it accounted, too? Please clarify.

P10L197: cloud formation → high RH condition. Unless the authors provide the data of cloud properties, it is not fair to say cloud formation.

P12L244-246: The local biomass burning also contributes to the aerosol-cloud interaction in the North Slope of Alaska (Hiranuma et al., 2013, JGR), which seems more relevant to your study.

P13L257-259: So how did this 266 nm enrichment influence your own results? Please clarify.

P13L263-267: So what determines the ambient abundance of amines in this particular study? Please be conclusive.

P13L268-269: What is the implication of Bi et al. with respect to your study? Please be conclusive.

P14L286-287: The authors may state that a low fraction of dust is the limitation/artifact.

P14L288-296: Moteki et al. (2017, Nature Comm.) reports the aircraft observation of magnetite (up to ~1 cm<sup>-3</sup>) over the East Asia. The authors may read, digest and incorporate it in your manuscript.

P14L299-P15L310: Was a depletion of chloride (e.g., Laskin et al., 2012, JGR) dominant in this study?

P15L308-309: Can the authors quantitatively differentiate the source (industrial vs. maritime) by looking at other types of particles came along with the Na-rich particles?

P15L311-319: Just for curiosity, did the authors find any biological particles during the campaign? If so, how many of those are classified as Other?

P17L353-354: In P16L338-340, the authors state that the ammonium nitrate is not a dominant form of nitrate in this study, which seems contradicting to the statement given here...

P17L360-361: What is the measurement uncertainty regarding TMA counts?

P21L457-458: So size or mixing state – which factor was determinant to determine the cloud formation ability in this particular period?

### Technical comments

P3L45 & P4L57: Be consistent with ‘in situ’ or ‘in-situ’.

P4L56: → ...and, in turn, affect...

P4L59: Although → Despite or Even with

P4L64: The formation of → The ability of aerosols to act as

P4L71 Too many ‘however’s are bothering. There are a total of 10 however-sentences appearing in this manuscript.

P5L79: → an Aerosol Mass Spectrometer (AMS) or other online/offline single particle instruments is

P5L87: Oceans sound awkward.

P5L88: Start a new paragraph.

P5L90-93: → Although scientists have worked to...in China (Zhang et al., 2012b), only few studies have employed...

P5L94: → obtain the mixing state of individual ambient particles during

P5L95: → Their results showed...large particles

P6L98: → fog residual particles at ground level in an urban area of South China.

P6L99: → They found an abundance of anthropogenic particles, including...

P6L102: → a mountain site in South China

P6L113: → Our measurements were carried out during...

P6L115: → This station is located at 200 km...

P6L117: → ...(273 km<sup>2</sup>), where...

P7L124: → The ambient temperature

P7L126: → The measurements of the droplet size spectra in this region performed during the winter of...

P7L129: Some → Previous

P8L151: each single → single (or individual particles)

P9 170-171: Providing references for these two sentences would be nice.

P9L169-170: → Low levels of...exclude

P9L175: 73,996 – be consistent with the use of "," to describe numbers throughout the manuscript.

P9L180: → similar clusters, such as aged EC, ...

P9L184: → Assuming that the number of...

P10L198-199: I suggest deleting two ‘and’s

P10L200: → Note that, on...

P11L211: The word "big freeze" is a nomenclature that may refer to something else. I suggest rewording it.

P11L216: → the main six particle types

P11L221: → The strong K<sup>+</sup> ion...

P11L221-222: Awkward/incomplete sentence - I suggest rephrasing the sentence.

P11L227: → suffer from the bias related to...

P11L232: → ...(m/z -46NO<sub>2</sub><sup>-</sup>, -62NO<sub>3</sub><sup>-</sup>) and presumably derived from...

P12L234: → An aged time of 81-88 min...showed an increase...

P12L246: → the majority of aged EC

P12L246: → Asian

P12L249-250: → ...particles were only...droplets, and the aged EC residuals were...

P12L251-252: → The Jungfraujoch station is predominantly within the free tropospheric condition, such that the biomass...

P13L262: → the size range

P13L263: → Aqueous reactions improving ... have been observed...

P13L267: → ...amine within the cloud.

P13L273: → Previous studies showed that dust particles that are internally mixed with sulfate and nitrate promote CCN activities...

P13L275: partly → partial

P13L276: A slightly increase → A slight increase

P13L273: Internal mixing ... is expect to act as CCN... → Dust particles that are internally mixed with sulfate and nitrate are expected to act as CCN...

P13L279: → during the spring season

P14L281: → Asian dust storms that occurred in March-May

P14L283: → Local dust emissions

P14L289 → and nitrate, making up 4.1%

P14L291: contributes → contribute

P14L293-294: → ...Fe-containing residuals have presumably come from...

P14L285: may occupied → may have occupied

P14L293: → Fe-containing residuals were

P14L299-300: → ...Na-rich particles are formed from varied sources...

P15L305: → The continental air masses

P15L306: → Industrial emissions were

P15L308: → This might suggest that the Na-rich particles were contributed from both industrial emissions and sea salts.

P15L318: → iron and steel products manufacturing facilities

P15L322: → Organic carbon tends to be...

P15L323: wealth → worth (I am not a big fan of too many "note" phrases. There are 13 notes in this manuscript, which seems a lot. Consider making smoother transitions and better flows between sentences/paragraphs without using too many notes).

P16L327: secondary inorganic species?

P16L336: → to be in the form of

P16L338: Low as compared to what?

P16L342: → in mass spectra (Figure 3)...

P16L343: → Thus, our data suggest that...

P16L340: → in two cloud residual types

P16L346: → In this study, we found that...

P16L346: → The data indicate that nitrate-containing particles account for...

P17L366: → Relatively high portions...

P17L368: → in the form of...

P17L370: → ...(Pratt et al., 2009). It may...

P18L373: → ...is mainly based on...

P18L374: → particles were internally...

P18L375: → ...to the K-rich type and probably...

P18L378: sensitive → sensitivity

P18L381: → Figure 6 displays the hourly... and Nf values of the nine types of...

P18L382-383: Awkward sentence – please rephrase.

P18L386: changed → shifted

P19L398-400: Incomplete sentence.

P19L400: → The southwesterly...

P19L401: → from the northerly

P19L402: → North China or northern China (the authors use the South China word consistently in the manuscript... why not for North???)

P19L403: → These changes might have led...

P19L411-412: → we selected to analyze cloud residuals that...on 18-19 Jan as compared to cloud residuals that...

P20L420: → for both the...

P20L431: → This data implies...

P21L453: → ...with one hour intervals. The ambient...

P21L456: → Thus, the data suggest that the initial...

P21L457: → cloudy air occurred around

P21L458: → in the size range of 200 nm up to 500 nm

P21L458-461: Awkward/incomplete sentences.

P22L465: → showed that there were no significant changes

P22L468: → as discussed in Sect. 3.3.

P22L473-474: do not active as CCN → are not active as CCN

P22L478: In comparing → When comparing

P22L478-479: has been observed to account for → accounted for

P22L484: → the differences

P23L487: → when compared to

P23L490: → nitrate-containing

P23L491: is most likely to be → possibly reflect the

P23L493: → confirmed that the update of gaseous  $\text{HNO}_3$  is an...

P23L494: → the increased nitrate level

P23L495: → (Figure 9), and...

P23L495: → in the cloud residuals

P23L496: → when compared with

P24L515: We did find → There was a high fraction... amine cloud residuals found when the...

P24L527: → The change observed in  $N_f$

P24L530: induce the activation of the nuclei particles to become cloud droplets → act as CCN

Figure 2: yellow dots and fire dots sound awkward...

Figure 3: 'of the sampled particles during the whole sampling period' – not necessary.

Figure 7: → with an interval of one hour

Figure 9: → ; wherears,