

## **Cloud scavenging of anthropogenic refractory particles at a mountain site in North China**

Lei Liu et al.,

We are grateful for referee#1's comments that are helpful for improving the quality of our paper. According to the suggestions of referee#1, these comments have been addressed and the paper is carefully revised. The corrections and the responses are as following:

In the revised paper, the red color was marked as the revised places.

### **Reply to Referee#1:**

1. Overall, the article investigated cloud residual and interstitial particles and addressed the potential impact of aerosol particles on environment (including human health) and climate over one of China's heavily polluted regions. The study (or campaign) was carried out in a mountain site 1500 m a.g.l. by in-situ sampling followed by lab analysis employing TEM-EDS. The research gained some valuable data to unveil some of the chemical properties and mixing states of cloud residual and interstitial particles in a uniquely polluted area. The underlying logic is sound and the structure of the article is appropriate. The figures are clear and appropriate. However, there is room for grammar improvement and concept clarification.

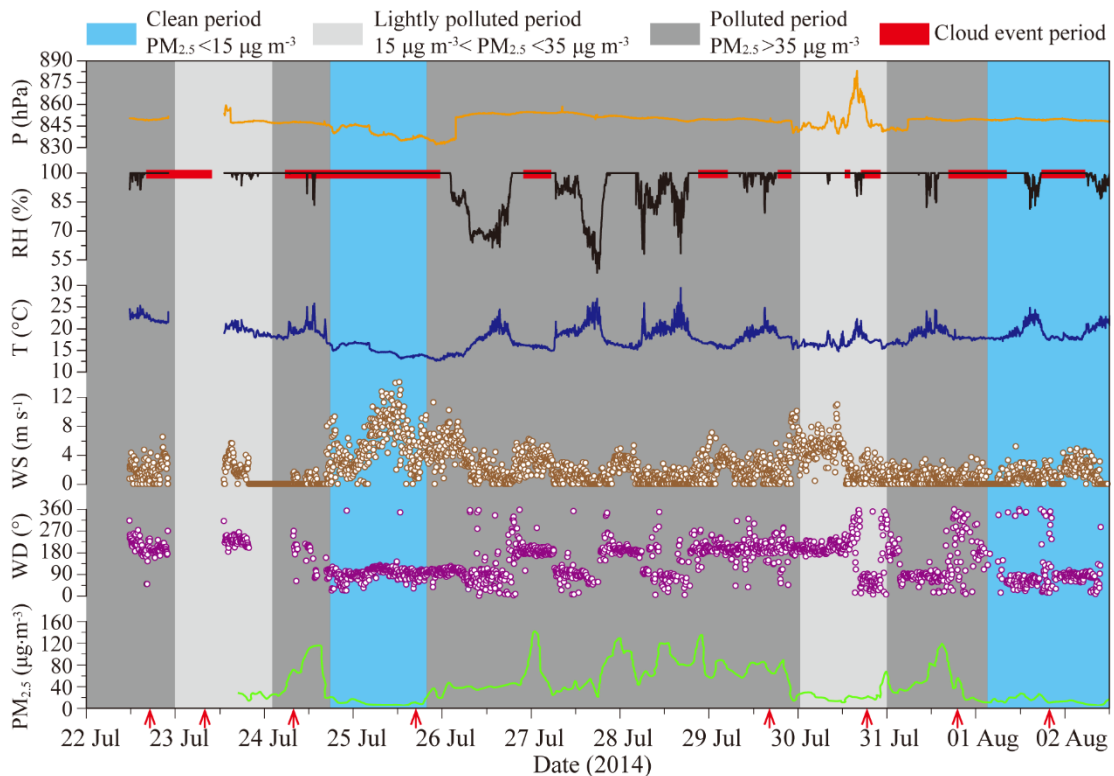
**Response:** We thank the referee#1' critical comments. All the comments and concerns raised by the referee have been explicitly considered and incorporated into the revised manuscript.

2. Table 1: There is a gap in individual particle sampling between July 25 and July 29. Is this because of instrument down time or other reasons? If it is the former, it could be appropriately addressed in the manuscript. If it is due to other reasons, they need to be justified.

**Response:** All the individual particle samples in this study were collected during cloud events. As shown in the Figure below, we missed the collection in the late midnight of 26 July and no cloud event occurred between 27 July and 28 July. To avoid this puzzled question, we add one sentence in section 2.2 Line 126-127:

“When cloud events occurred at the summit of Mt. Tai, individual particle

samples were collected during the cloud events except one cloud event in the late midnight of 26 July”



- Line 16: The statement of “our knowledge about aerosol-cloud interaction in heavily polluted conditions is weak” is not convincing. It might be difficult to accurately describe or explain the aerosol-cloud interaction in polluted regions, as the pollution type could vary from region to region. However, scientific communities are aware of the possible pollution sources and how these sources could influence aerosol-cloud interaction. It’s just a matter of how complicated the particles could be in terms of its chemical composition and how active it could serve as CCN. Nevertheless, the NCP could be a unique area for particle pollution and well deserve sufficient scientific investigations.

**Response:** We appreciated that you can point it out. We carefully revised this sentence. Please see Line 17-18:

“Few studies have been conducted to characterize the aerosol-cloud interactions in heavily polluted conditions worldwide”

- Line 20: Mixing state should only refer to aerosol or particle as an ensemble. When it comes to an individual particle, you can only describe it as a pure material or a mixture based on chemical composition. You can definitely describe the mixing state after investigating chemical composition of all the individual

particles sampled. Therefore, I would express Line 20 as...used to investigate size and chemical composition of individual cloud RES and INT particles, and study the mixing states of these particles.

For definition of aerosol/particles mixing state, please refer to:

N. Riemer and M. West [2013], Quantifying aerosol mixing state with entropy and diversity measures, *Atmos. Chem. Phys.*, 13, 11423-11439, DOI: 10.5194/acp-13-11423-2013

For how to use individual particle chemical composition to describe mixing state, please refer to:

Deng, C., Brooks, S. D., Vidaurre, G., and Thornton, D. C. O.: Using Raman Microspectroscopy to Determine Chemical Composition and Mixing State of Airborne Marine Aerosols over the Pacific Ocean, *Aerosol Science and Technology*, 48, 193-206, 10.1080/02786826.2013.867297, 2014.

**Response:** Thank you for your comments. Definitely, the difference about definition of mixing state of aerosol particles should be clarified in this context. We revised the sentence in Line 20 according to the suggestion and added more content in section 3.2 Line 184-191 to explain it. As the referee's suggestion, we cited these two important papers. Please see:

**Line 20-22:** "Transmission electron microscopy (TEM) was used to investigate size and chemical composition of individual cloud RES and INT particles, and study the mixing states of these particles."

**Section 3.2 Line 184-191:** "Mixing state of aerosol particles is currently classified into population mixing state (Riemer and West, 2013) and single particle mixing state (Deng et al., 2014; Li et al., 2016b). Riemer and West (2013) defined the population mixing state as the distribution of the aerosol chemical species among the particles in a given population. However, based on the single particle mixing state of an individual particle acquired by TEM (Li et al., 2016b), this study emphasizes the distribution of different types of aerosol components within and on particle surface. Furthermore, single particle mixing state can be further divided into externally mixed particle and internally mixed particle (i.e., individual particles containing two or more types of aerosol components) (Li et al., 2016b)."

5. Line 30 to 32, The readers have to understand the relationships built on three "from"s, there must be a better grammar expression for this segment. Additionally,

the article needs to discuss more about biogeochemical cycle if it is to be emphasized in abstract.

Response: We revised the sentence and deleted biogeochemical cycle here because the issue seems far away from the interactions of aerosol-cloud. Please see Line 31-33:

“Our findings provide an insight into the potential impacts on cloud radiative forcing from black carbon and metal catalyzed reactions of SO<sub>2</sub> in micro-cloud droplets containing soluble metals released from fly ash and metals over polluted air.”

6. The article didn't measure black carbon but discussed its potential impacts. The researches of this study need to define the relationship of black carbon and the TEM measured species, e.g., soot. The authors should justify the interchangeable use of black carbon and soot identified by TEM if this is the case.

Response: Thank you for your comments. We added more explanation to show that soot and black carbon can be interchangeable in this study in Line 204-208:

“In much of the climate-change and environmental literature, “soot” and “black carbon” are commonly used interchangeably, and “black carbon” is the most commonly used term in the climate-science community (Andreae and Gelencsér, 2006; Buseck et al., 2014). In the following sections, we use the term “soot” for the classification of particle types and the term “black carbon” for the discussion of the climate issues.”

Andreae and Gelencsér [2006] have given a clear definition about soot and black carbon in their paper. “Soot” is formed by combustion process and presents in the atmosphere as fine particles. Soot particles usually have chain-like structure composed of aggregates of spherules made of graphene layers, consisting almost purely of carbon. “Black carbon” generally implied to have optical properties and composition similar to soot carbon. In the literature, “black carbon” or “soot carbon” are often used synonymously for the major light-absorbing component of combustion aerosols. Buseck et al. [2014] also mentioned that black carbon and soot are commonly used interchangeably in much of the climate-change and environmental literature and no consensus exists in the atmospheric science community about whether soot is a part of black carbon, black carbon is a part of soot, or black carbon is equivalent to soot.

In the current knowledge, “Black carbon” is the most commonly used term in

the climate-science community. So for aerosol particle classification, we used the term “soot”, when we discussed the climate issues we used the term “black carbon”.

**References:**

Andreae, M. O., and A. Gelencsér (2006), Black carbon or brown carbon? The nature of light-absorbing carbonaceous aerosols, *Atmos. Chem. Phys.*, 6(10), 3131-3148, doi:10.5194/acp-6-3131-2006.

Buseck, P. R., Adachi, K., Gelencsér, A., Tompa, É., and Pósfai, M. (2014), Ns-Soot: A material-based term for strongly light-absorbing carbonaceous particles, *Aerosol Sci. Tech.*, 48, 777-788, doi:10.1080/02786826.2014.919374.

7. Line 48: Is chemical composition of clouds important? Isn't cloud droplet mainly composed of water in terms of mass? It is understandable that chemical composition of aerosol is important for cloud nucleating, not as important as particle size though according to Dusek et al., 2006. It seems that the authors of this manuscript mean chemical composition of cloud RES and INT particles here.

**Response:** We agreed with the referee#1. We revised this sentence according to the suggestion. Please see Line 47-50:

“Owing to the rapid industrialization and urbanization in Asia, large quantities of aerosol particles from anthropogenic sources are released into the atmosphere, which can dramatically affect the chemical composition of CCN, and furthermore change the properties of clouds such as radiative forcing, lifetime, and precipitation patterns”

8. Line 58-59: The impact of Fe-bearing particles on oceans is undeniable but seems to be beyond the discussion of this research. There is no measurement about how much Fe-particles are transported to ocean. Therefore, it seems to be not very relevant and insignificant to be mentioned.

**Response:** We deleted this statement according to the suggestion.

**Technical corrections:**

1. Line 74: “Field observations are requested to...”, so, who has requested field observations? I think it would be more appropriate to say “Field observations are

needed to...”

**Response: Changed.**

2. Line 79: I would recommend changing “Many studies...” to “Numerous studies...” or “Several studies...” “Many” sounds just vague and exaggerating.

**Response: Changed.**

3. Line 79 to Line 83: From “Many studies...” on, it would be better if these can be incorporated into one sentence.

4. Line 92: “Capture interactions” is questionable and sounds exaggerated.

**Response: We used “reveal” to replace “capture”.**

5. Line 101: Similar to a previous comment about Line 79, there are two occurrence of “many”. First, it would be better to have some vocabulary variations. Second, “many” doesn’t sound academically accurate.

**Response: We replaced the first “many” by “several”, and the second “many” by “a number of”.**

6. Line 124-125: Recommend changing to “More detailed information about the setup of a modified sampler can be found in Li et al., 2011a.”

**Response: We changed the sentence according to the suggestion.**

7. Line 142: Recommend changing to “...interstitial particles mostly distributed on the peripheral areas of TEM grid...”

**Response: Changed.**

8. Line 145: Recommend changing “separate” to “distinguish between”.

**Response: Changed.**

9. Line 146: Recommend changing “In a word, many previous” to “Generally, a number of previous”

**Response: Changed.**

10. Line 154: “(see the supplement)” to “(refer to the supplement)”

**Response: Changed.**

11. Line 191: Recommend changing from “such as steel mills and smelters.” to “such as production activities in steel mills and smelters.”

**Response: Changed.**

12. Line 233: “we can still identify them...” to “they can still be identified...”

**Response: Changed.**

13. Line 243 to Line 244: It can be challenged mathematically that 76% is 3.5 times higher 22%. Generally, it is preferred to express it as “76% is 2.5 times higher

than 22%” or “76% 3.5 times of 22%”

**Response: We rewrote the sentence. Please see Line 260:**

“we found that 76% of cloud RES were a mixture of sulfates and refractory particles, which is 3.5 times of 22% in cloud INT”

14. Line 259: “reveals” should be changed to “reveal”

**Response: Corrected.**

15. Line 270: “the particle number” to “particle number”

**Response: Corrected.**

16. Line 280 and Line 293: Better to rephrase the expression of “We believe”. It is too subjective to be overused in a scientific journal paper.

**Response: Line 280 “We believe” was replaced by “The present study reveals”.  
Line 293 “We believe” was replaced by “It is highly probable”.**

17. Line 285: It would be better to just use “black carbon” instead of “BC”, even though it has been defined earlier. “BC” has only been discussed in introduction section and hasn’t been discussed ever since. It won’t take much more space though.

**Response: Changed.**

18. Line 309: “If they are” to “If they were”. The verb is in the subjunctive.

**Response: Changed.**

19. Line 321: “is still a mystery in polluted air” to “is still unresolved”

**Response: Changed.**

20. Line 322: “should be further considered” to “should be further studied”

**Response: Changed.**