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

Lei Liu et al.,

We are grateful for referee#2's comments that are helpful for improving the quality of our paper. According to the suggestions of referee#2, the 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#2:

1. In this study cloud residual and cloud interstitial particles were collected at a site and analyzed to obtain insight into the aerosol-cloud interactions. Properties such as size, morphology, composition and the mixing state were studied to highlight the salient differences in clout RES and INT particles. The researchers also propose a model to further explain the cloud-aerosol interactions.
   Overall, the background, scientific rationale and the data presented in the study are sound. The presentation style is clear and easy to follow. However, there are a couple minor issues which need to be addressed.
   Response: We thank the referee#2' nice comments. All the comments and concerns raised by the referee have been explicitly considered and incorporated into the revised manuscript.
2. In Results, there is a section 3.2 and 3.4 but there is no section 3.3. It seems that section 3.4 needs to be changed to 3.3.
   Response: Corrected.
3. In section 3.2, the authors classify the particles as (1)S-rich, (2)OM, (3)Soot, (4)mineral and (5)fly-ash and (6)metal. Furthermore, they say (2)-(6) can be classified as refractory particles owing to their refractory behavior under electron beams. The authors then say that "Based on the mixing properties of individual particles, they can be further classified into four categories: S-mineral, S-soot, S-fly ash/metal, S-fly ash/metal-soot" together defined as S-refractory particles. While the classification seems reasonable, the use of the term "mixing properties" is a bit vague here unless more information is provided. What are the mixing

properties to which the authors are referring? Are S-refractory particles formed from refractory particles? If so, how? Is it a probabilistic phenomenon or is there a fundamental difference between the refractory particle in itself as opposed to a component of S-refractory particle?

Response: Thank you for your comments. We added more information about the mixing state in section 3.2. Please see 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)."

The explanation is helpful for the potential readers to understand how we classify the mixing state in this study.

To better answer the question, we give more explanations as below:

About the mixing state of individual particles, we have detailed discussion and clear definition in our previous paper (Li, W., et al., 2016a, 2016b). The mixing state of an aerosol particle can be classified as either internally mixed (whether distinct, homogeneous entities occur within the same particles) or externally mixed (whether they are separated entities in the air), please see the figure below.

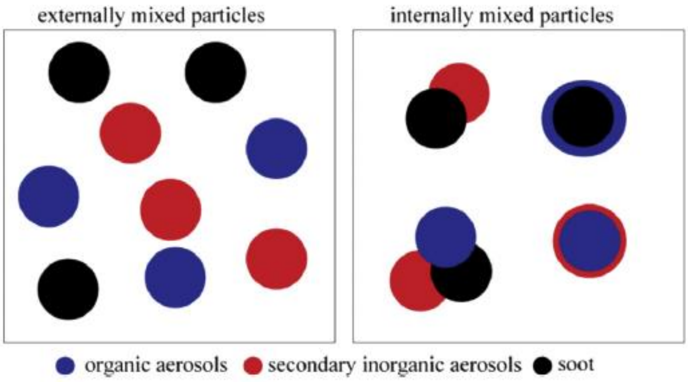

Fig. 14. Schemes of externally mixed and internally mixed particles.

The basic types of aerosol components (i.e., (1) S-rich, (2) OM, (3) soot, (4) mineral, (5) fly-ash and (6) metal) refer to the externally mixed individual particles. For the individual particles containing two or more basic types of aerosol components, they can be further classified into S-mineral, S-soot, S-fly ash/metal, and S-fly ash/metal-soot as internally mixed particles.

Refractory particles are directly emitted from natural or anthropogenic sources, such as mineral dust particles from deserts, soot and primary organic aerosol particles (POA) from incomplete combustion of fossil fuel and biomass, fly ash from coal combustion, and metals from heavy industries. The internally mixed particles such as S-mineral, S-soot, S-fly ash/metal, and S-fly ash/metal-soot are formed through condensation, coagulation, and cloud process of refractory particles with secondary particles (i.e., S-rich) during atmospheric transports. Therefore, in this study, we used S-refractory particle representing internally mixed particle between secondary sulfate and primary refractory particles (e.g., mineral, soot, POM, fly ash, and metal).

References:

Li, W., J. Sun, L. Xu, Z. Shi, N. Riemer, Y. Sun, P. Fu, J. Zhang, Y. Lin, and X. Wang (2016a), A conceptual framework for mixing structures in individual aerosol particles, *J. Geophys. Res.-Atmos*., 121(22), 13784-13798.

Li, W., L. Shao, D. Zhang, C.-U. Ro, M. Hu, X. Bi, H. Geng, A. Matsuki, H. Niu, and J. Chen (2016b), A review of single aerosol particle studies in the atmosphere of East Asia: morphology, mixing state, source, and heterogeneous reactions, *J. Clean Prod.*, 112, 1330-1349.