egusphere-2023-290
Reply #1:

Insoluble aerosol particles are the main source of ice nucleating particles (INPs) in clouds, but their physical and chemical properties have still not been well determined. This manuscript presents analysis of insoluble particles in 12 hailstones collected from 8 hailstorms occurred in China between 2016 and 2021, by scanning electron microscopy (SEM) and energy dispersive X-ray spectrometry (EDX). The insoluble particles were grouped into three species by self-organized maps (SOM) and random forest method. The size distribution of the insoluble particles in embryos and different shells of sliced hailstones was analyzed and was fitted with logarithmic normal distributions. The subject is scientifically interesting and is well within the scope of the journal. But some parts of the manuscript are not well presented or not clear. I think the presenting quality should be substantially improved before it can be accepted for publication in ACP. Some more specific comments are as follows.

Thank you for your review. We have carefully considered every comment and incorporated all of the suggestions into the revised manuscript. The following texts are our point-to-point response.

Specific comments

1. Although a large number of insoluble particles were found in each of the hailstone samples, it is not sure whether and how many those insoluble particles have ever served as ice nuclei during the formation of the embryos and different shells of hailstones, since many of them might be captured during the formation, growth and falling out of the hailstones.

   Reply: Yes. We agree that we may not determine how many these insoluble particles have ever served as ice nuclei. This work is the very first step to answer this question. We collaborated with a PH. D student from another group, who found a positive correlation between insoluble particles we measured and immersion freezing nucleation particles in hailstones. The result is currently under review in Atmospheric Research. This finding is consistent with a parameterization for freezing nucleation that was developed based on the concentration of ice-nucleating particles and aerosols larger than 0.5 μm (DeMott et al., 2010).

   Our subsequent objective is to quantify the number concentration of immersion ice-nucleating particles and to determine the type of ice-nucleating particles in each shell in drop-freezing experiment. Then, we will establish a relationship between immersion ice-nucleating particles and size distribution of insoluble particles.

2. Some sentences are not clearly presented to the readers, such as "… simulation due to little regarding species and number concentration of heterogeneous ice-nucleating particles" in the abstract. I suggest the whole text be checked with the help of an English editor.

   Reply： We have made some changes to the manuscript in an effort to improve it. These changes do not affect the content or structure of the paper, and we have not listed all of them here. We would like to express our gratitude for the reviewers diligent work and h ope that these corrections will be satisfactory.

3. Line 18-19: “Further, classic size distribution modes of organics and dust were performed as logarithmic normal distributions”: not clear.

   Reply: The text has been revised as “classic size distribution of organics and dust followed logarithmic normal distributions”

4. Line 22-23: “Insoluble particles, acting as main heterogeneous ice-nucleating particles in the atmosphere, may indirectly impact precipitation formation”: why indirectly?

   Reply: “Indirectly” was removed.

5. Line 26-28: “Only few models calculate the number concentration of ice-nucleating particles in clouds, that leads to a misestimation about number concentration of ice particles and large errors in simulation”: not clear to me.

   Reply: The text has been revised as “Few models used the freezing parameterization, which establishes a connection between the number concentration of ice-nucleating particles and the number concentration of ice crystals explicitly. The absence of any description regarding the physical properties of ice-nucleating particles in models can result in an incorrect estimation of ice crystals and lead to significant bias in simulations”.

6. Line 31-32: “Measurements of the number concentration and species of ice-nucleating particles, mainly insoluble particles, were conducted by an airborne equipment or laboratory instrument with air parcels”: not clear to me.

   Reply: There are two situation for sampling ice-nucleating particles.

   The first involves airborne instruments, such as the continuous flow thermal gradient diffusion chamber (DeMott et al., 2010). This technique collects air during flight, activates ice-nucleating particles, and counts ice crystals. It can measure the number concentration of insoluble particles in several freezing modes (up to four modes) by changing the temperature and supersaturation in the chamber.

   The second is in laboratory where scientists measure the number concentration of ice-nucleating particles in air parcels. The air sample was sampled by aircraft in the air (e.g., Winter Icing in Storms Project 1994) or inlet at high altitude (e.g., Winter Icing in Storms Project 2000). Additionally, certain species of particles aerosols are tested in laboratories to determine their freezing efficiency.

   Lines 33-36 have been expanded to include additional details as: “There are two ways to sample ice-nucleating particles: The first involves an airborne instrument, named continuous flow thermal gradient diffusion chamber. The second is done in the laboratory, where scientists conduct freezing experiments".

7. Line 34-35: "Most field projects sampled air parcels in anvils of convective clouds, cirrus and winter mixed-phase stratiform clouds, keeping airborne equipment in good working condition": I am not sure what this sentence want to tell.

   Reply: As mentioned in response to point 6, airborne continuous flow thermal gradient diffusion chamber is used to count ice-nucleating particles in sampled air while the aircraft is flying. These detections take place in cloud-free regions (Winter Icing in Storms Project 1994 and Mixed-Phase Arctic Cloud Experiment), inside plumes (PACDEX), within the base of stratiform clouds (PACDEX), and through cirrus clouds (PACDEX).

   Bad weather events can hinder flights during field projects. For example, turbulence disrupted communication systems during the 6th research flight of Alliance Icing Research Study-2. Pilots on the 11th research flight were worried about that cloud top transit could create problems if long-term supercooled conditions were experienced.

   These weather-related events were recorded in flight logs, but not all field studies share their flight reports with open access. Reports and articles indicate that no flight can sample air parcels through cores in convection, especially deep convection in severe storms.

   We have removed "keeping airborne equipment in good working condition" and the word "most". The Lines 33-41 has been revised for clarity as "T There are two ways to sample ice-nucleating particles: The first involves an airborne instrument, named continuous flow thermal gradient diffusion chamber(DeMott et al., 2010; Prenni et al., 2009; Rogers et al., 2001). The second is done in the laboratory, where scientists conduct freezing experiments (Hoose and Möhler, 2012). In most cases, it is necessary for aircraft to collect air parcels for measurement of the physical properties of ice-nucleating particles in the air. However, former field projects sampled air parcels in anvils of convective clouds, cirrus and winter mixed-phase stratiform clouds. No flight report or article has reported that they sampled air parcels through cores in deep convection. This phenomenon is consistent with consideration for flight security. Thus, current observation is insufficient for describing the whole convective cloud, especially the deep convection in severe storms."

8. Line 39-41: The logics of paragraph is incorrect.

   Reply: Thank you. We found the logical mistake and modified the sentence. The text has been revised as "Hailstones, as a product of deep convective clouds, serves as a carrier of information within these clouds. Recently, analysis revealed large diversity in number concentration of soluble ions among hailstones from different hailstorms (Li et al., 2018). Further, the detection of soluble ions along with isotopic analysis of a huge hailstone revealed an up-and-down hailstone growth trajectory, which demonstrated that the different shells were formed at different heights (Li et al., 2020)."

9. In Table 1, why only one value is provided for the two samples from Guyuan City?

Reply: We have listed the particles number of each hailstone as your suggestion and revised footnote of Table 1.

10. In Line 169, Formula (2): why $N_{used} = N_{filter}$?

Reply: It is an assumption. The number of insoluble particles present in the consumed diluting solution (N_{used}) range from 10^5 to 10^6. It is expected as a true value. As mentioned in lines 100-101, we repeated rinsing for 5 times to ensure particles adhered to the membrane as much as possible, so that N_{used} was assumed to be equal to the number of insoluble particles found on the filter membrane (N_{filter}).

Additional details regarding this process have been included in lines 226-228 of our manuscript as "Assuming the rinsing operation ensures all insoluble particles in the shell were on the membrane, the number of insoluble particles in the consumed solution (N_{used}) is equal to the number of insoluble particles counted on the membrane (N_{filter})."

11. Line 173, formula (3): The inversion form of this formula might be easier to be understood.

Reply: Thank you for your comment. The formula has been revised to:

$$\frac{S_{images}}{S_{filter}} = \frac{N_{count}}{N_{filter}} \qquad (R1-1)$$

12. Line 182: How formula (5) is derived?

Reply: The Formula (4) is:

$$n_{liquid} = \frac{1}{V_{liquid}} \cdot \frac{S_{filter}}{S_{images}} \cdot \frac{V_{diluted}}{V_{used}} \cdot N_{count} \qquad (R1-2)$$

Take the logarithm on both sides:

$$\ln n_{liquid} = -\ln V_{liquid} + \ln S_{filter} - \ln S_{images} + \ln V_{diluted} - \ln V_{used} + \ln N_{count}$$

Next, differentiate the equation:

$$\frac{dn_{liquid}}{n_{liquid}} = -\frac{dV_{liquid}}{V_{liquid}} + \frac{dS_{filter}}{S_{filter}} - \frac{dS_{images}}{S_{images}} + \frac{dV_{diluted}}{V_{diluted}} - \frac{dV_{used}}{V_{used}} + \frac{dN_{count}}{N_{count}}$$

As,

$$dS_{filter} = dS_{images} = 0$$

Now, we get the formula (5):

$$dn_{liquid} = n_{liquid} \cdot \left(\frac{dV_{liquid}}{V_{liquid}} + \frac{dV_{diluted}}{V_{diluted}} + \frac{dV_{used}}{V_{used}} + \frac{dN_{count}}{N_{count}}\right) \qquad (R1-3)$$

We also add more description into the manuscript.

13. Line 227-228: "Each number concentration at diameter D total number concentration of insoluble particles with diameter ranging from D−1 μm to D+0.1 μm", not clear to me;

Reply: The bin width is 0.2 μm in Fig. 7 and Fig. 10, and 2 μm in Fig. 8 and Fig. 9.

14. Line 230-231: "Blue and gray bars show the standard deviation of insoluble particles from seven hailstorms and one hailstorm, respectively": not clear.

Reply: Blue bars show the standard deviation of the number concentration of insoluble particles in hailstones (BJ1, BJ2, BS, FS,GY1, GY2, YT and GYA) from eight different hailstorms. On the other hand, gray bars represent the standard deviation of the number concentration of insoluble particles in five hailstones (BJ2 to BJ6) collected from the same hailstorm. The caption has been revised.

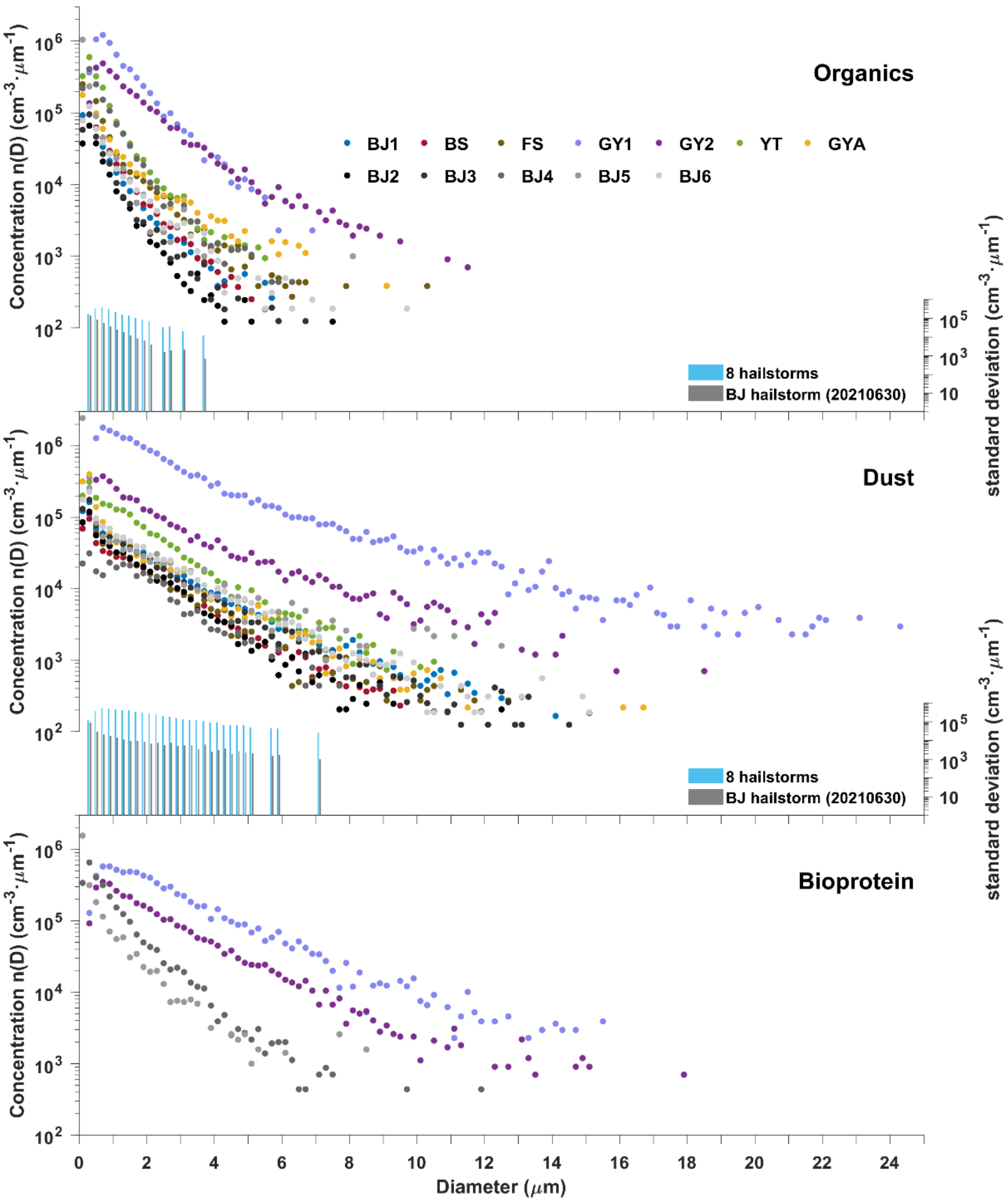

Fig. 7: Size distribution of organics, dust, and bioprotein aerosols of insoluble particles in 12 hailstones. The colored dots represent data from 7 hailstones BJ1, BS, FS,GY1, GY2, YT and GYA which were from different hailstorms. The black and gray dots correspond to data from hailstones (BJ2 to BJ6) that were from the same hailstorm occurring in Beijing on June 30, 2021. The blue and gray bars indicate the standard deviation of number concentration of insoluble particles from 8 hailstones (BJ1, BJ2, BS, FS,GY1, GY2, YT and GYA) from 8 cases and 5 hailstones (BJ2 to BJ6) from one case, respectively. Abbreviations (corresponding to Table 1): BJ - BeiJing; BS - BaiSe; FS - FuShun; GY - GuYuan; GYA - GuiYAng; YT - YanTai.

15. Line 236-237: "These initial ice particles are likely formed by insoluble particles where heterogeneous nucleation processes": incomplete sentence.

    Reply: We completed this sentence. Text has been revised as "These initial ice particles are formed through nucleation of insoluble particles where heterogeneous nucleation take place."

16. Line 294 and other places: There should be no unit for logarithmic function.

    Reply: Thank you. We deleted the units of logarithmic terms in the Lines 359-360 and Lines 369-370.

Technical corrections

1. Change "for" to "of" after "description" in line 29;
   Text revised.

2. Change "in" to "of" after "suppression" in line 54;
   Text revised.

3. Table captions should be provided on top of the tables;
   Table revised. Please see Table 1 (line 81).

4. Line 116: Change "Ault et al. in 2012 and Kirpes et al. in 2018" to "Ault et al. (2012) and Kirpes et al. (2018)";
   Text revised.

5. Line 117: Remove "Ault et al. 2012; Kirpes et al. 2018";
   Text revised.

6. Line 122: Change "Species of aerosol particles vary regionally" to "Species of aerosol particles vary with sampling location";
   Text revised.

7. Line 166: Formula (1): Change N_{dilute} to $N_{diluted}$;
   Text revised.

8. Line 236: Change "droplet" to "droplets".
   Text revised.

References:

DeMott, P. J., Prenni, A. J., Liu, X., Kreidenweis, S. M., Petters, M. D., Twohy, C. H., Richardson, M. S., Eidhammer, T., and Rogers, D. C.: Predicting global atmospheric ice nuclei distributions and their impacts on climate, Proc. Natl. Acad. Sci., 107, 11217–11222, https://doi.org/10.1073/pnas.0910818107, 2010.

Hoose, C., Kristjánsson, J. E., Chen, J.-P., and Hazra, A.: A Classical-Theory-Based Parameterization of Heterogeneous Ice Nucleation by Mineral Dust, Soot, and Biological Particles in a Global Climate Model, J. Atmos. Sci., 67, 2483–2503, https://doi.org/10.1175/2010JAS3425.1, 2010.

Li, X., Zhang, Q., Zhu, T., Li, Z., Lin, J., and Zou, T.: Water-soluble ions in hailstones in northern and southwestern China, Sci. Bull., 63, 1177–1179, https://doi.org/10.1016/j.scib.2018.07.021, 2018.

Li, X., Zhang, Q., Zhou, L., and An, Y.: Chemical composition of a hailstone: evidence for tracking hailstone trajectory in deep convection, Sci. Bull., 65, 1337–1339, https://doi.org/10.1016/j.scib.2020.04.034, 2020.

Prenni, A. J., Demott, P. J., Rogers, D. C., Kreidenweis, S. M., Mcfarquhar, G. M., Zhang, G., and Poellot, M. R.: Ice nuclei characteristics from M-PACE and their relation to ice formation in clouds, Tellus B, 61, 436–448, https://doi.org/10.1111/j.1600-0889.2009.00415.x, 2009.

Rogers, D. C., DeMott, P. J., Kreidenweis, S. M., and Chen, Y.: A Continuous-Flow Diffusion Chamber for Airborne Measurements of Ice Nuclei, J. Atmos. Ocean. Technol., 18, 725–741, https://doi.org/10.1175/1520-0426(2001)018<0725:ACFDCF>2.0.CO;2, 2001.