Short Communication

Chemical composition of a hailstone: evidence for tracking hailstone trajectory in deep convection

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Hail is produced by all types of deep convective storms and occurs commonly for short periods in most parts of the world [1]. The growth path of hailstones in hailstorms remains largely uncertain [2–4]. Due to the limitations hampering direct observations of severe hailstorms, it is difficult to identify definitively the in-cloud conditions and hailstone growth trajectories that lead to hailstone formation [2,3]. However, natural hailstones can provide insights into hydrometeors and aerosols in deep convective storms. Hailstones have been collected and examined, and various techniques have been applied to measure representative atmospheric chemical isotopes [2,5], which is of great help when investigating hailstones’ origin and structure, as well as for understanding the microphysical mechanisms and aerosol-collection processes in hailstorms [2,5,6]. Hailstone layers are a record of the vertical profile of the storm [5]. During their growth, hailstones collect cloud droplets and rain drops in a specific way as they travel inside the hailstorm, following predictable individual paths [2,7]. Trace substances, such as water-soluble ions originating from the Earth’s surface moving vertically from the atmospheric boundary layer to the free atmosphere, can be ingested into cloud droplets and rain drops [2,5], in which case, aerosols may be removed at any point during the life cycle of a hailstone. Since hailstone trajectories in storms have received much attention, hailstone embryo and outer growth layers have been tested in the laboratory for water stable isotopes [8]. Based on the decreasing temperature profile with increasing altitude in the troposphere and equilibrium isotopic fractionation during condensation [9], a wide range of isotopic depletion of water vapor in the tropical and subtropical upper troposphere has been observed in both convective dehydration and gradual dehydration mechanisms [9], suggesting that water isotope fractionation is, in principle, a sensitive tracer for diagnosing transport and dehydration mechanisms of hydrometeors in cloud, including hailstones. Hence, a clear distribution of water-soluble ions and water stable isotopes in a hailstone may potentially act as a firm bridge between the vertical distribution of aerosols and the growth trajectory of hailstones in convective storms.

In 2016, hailstones from 15 hailstorms were collected [2], including one hailstone sample with a diameter over 50 mm from Daxin County, Guangxi, southern China, providing us with a valuable opportunity to detect layered chemical distribution. The present study analyzed layered concentrations of eight detected water-soluble ions (Na+, K+, Ca2+, Cl−, NO3−, SO42−, HCOO− and CH3COO−) and contents of water stable isotopes (δD and δ18O) in five layers, from the hailstone outer surface to the inner core (Tables S1 and S2 online). These eight ions, which may be used as markers to deduce the growth mechanisms of hailstones as well as the distribution of aerosols in storms, varied in concentrations in different layers of the hailstone sample, with “V”-shaped distributions (Fig. 1a and Table S1 online). All eight ions had higher concentrations near the innermost layer (d5) but decreased in the intermediate layers before increasing again towards the outer layers. The concentrations of each of the eight ions were lower in the intermediate layers (d2, d3, and d4) than in the innermost layer (d1) and the outermost layer (d5). Similarly, the concentrations of water-soluble ions with respect to radius and water stable isotopes (δ18O and δD) also exhibited “V”-shaped distributions (Figs. 1a and S3 online), and the concentrations of δ18O and δD both reached their minimum at layer (d2), with values of −8.38 and −53.70, respectively, followed by the contents at the innermost layer (d1) and the outermost layer (d5).

This “V”-shaped distribution of water-soluble ions contrasts with the monotonically increasing distribution of microorganisms...
with radius from the inner to outer layers \([10]\), suggesting that different tracers may be used to represent the different stages of the lifecycle of a hailstone, likely due to differences between the absorption of water-soluble ions and microorganisms. Microorganisms tend to behave more like insoluble giant aerosol particles in larger rain drops, and contribute most during the final stage of the hailstone growth \([5,10]\), whereas water-soluble ions may act as CCN from the activation of cloud droplets at the cloud base and then throughout the entire life cycle of the hailstone. While another possible reason may come from the different atmospheric environment, structure and evolution of the two hailstorms leading different distribution of tracer and different trajectory of hailstones in two cases. These results suggest a connection between the course of dissolution of ions into hailstones during their growth process and the trajectory of hailstones in convective storms, but more evidence based on other techniques and adequate explanations from numerical simulations are needed.

Testing water stable isotopes using other techniques will provide robust results regarding the possible hailstone trajectories, which differ from water-soluble ions, as the growth temperatures of hailstones are correlated well with isotopic features \([6]\). As gradual dehydration is expected to follow Rayleigh distillation, in which all condensate is removed during adiabatic cooling, the vertical profiles of \(^{18}\)O and \(\delta D\) for atmospheric water vapor is expected to decrease monotonically from the ground surface to the coldest tropopause \([9]\). As a result, layer \(d_2\) may form at the highest layer, whereas layers \(d_1\) and \(d_5\) form at lower layers, providing corroborative evidence for the one-way hailstone trajectories suggested by the results of water-soluble ions.

The possible absorption processes and growth mechanisms of hailstones have been investigated using simulations with WRF-Chem (Fig. S4 online) and a 1-D hail growth model (Fig. S5 online), in which hailstones were initialized as a liquid embryo in collision coalescence with a radius ranging from 1.5 to 3.0 mm at intervals of 0.5 mm, which is a typical diameter for a large super-cooled raindrop to form a hailstone, and were seeded in the cloud base until it fell below a height of 2 km (Fig. 1b). Hailstones with initial radii of 2.0 and 2.5 mm can ascend as high as 6 or 8 km in the given sounding (Fig. S5 online), extracted from the simulation of WRF-Chem, and hail-producing regions with reflectivity of 60 dBZ have been observed at similar heights by radar (Figs. S1 and S2 online). The hailstones all followed a similar one-way trajectory with fixed background conditions (Fig. 1b), consistent with simulated trajectories reported in previous studies \([4]\). A similar "V"-shaped distribution of ion concentrations, with higher concentrations during the initial and final stages, was also observed, in agreement with laboratory findings (Fig. 1a). The experimental results captured the general distribution of the measured ions' primary mode of variability from the inner core to the outer surface of the hailstone. Note that the size of the hailstone may have been overestimated due to the simplified approximation of the radius growth, which did not take into account the melting process. Nevertheless, the results obtained from the 1-D model serve to establish a potential bridge between hail growth trajectory and the distribution of water-soluble ions in a hailstone.

This is the first study to document a "V"-shaped layered distribution of water-soluble ions and isotopic compositions in a huge hailstone, suggesting an up-and-down hailstone growth trajectory, along which both nucleation scavenging and impaction scavenging of aerosols take place. The "V"-shaped distributions were verified using a simulation. However, questions remain regarding whether the instantaneous concentrations of ions and isotopic composition fluctuate and whether the resolution of the described vertical growth trajectory of the hailstones can be improved. Further investigations are required to obtain accurate measurements of all types of aerosol particles, acting as CCN and ice nuclei, and to obtain detailed atmospheric environmental profiles and better knowledge of hail growth by analyzing a variety of hailstones from different hailstorms with different properties, such as shape, radius, transparency, etc. Nevertheless, our study shows more than the hailstone trajectory, as we relate the vertical distribution of aerosols in hailstorms to the growth trajectories of hailstones based on a layered analysis of water-soluble ions and water stable isotopes.

Conflict of interest

The authors declare that they have no conflict of interest.
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Author contributions

Xiaofei Li wrote the original draft under the concept presented by Qinghong Zhang. Xiaofei Li performed the experiments on detecting water soluble ions. Yongrui An performed the experiments on detecting water stable isotopes directed by Liping Zhou. All authors discussed and contributed to the final manuscript. Qinghong Zhang directed this project.

Appendix A. Supplementary materials

Supplementary materials to this article can be found online at https://doi.org/10.1016/j.scib.2020.04.034.

References

[1] Allen JT, Giammanco IM, Kumjian MR, et al. Understanding hail in the earth system. Rev Geophys 2020;58. e2019RG000665.
[2] Li X, Zhang Q, Zhu T, et al. Water-soluble ions in hailstones in northern and southwestern China. Sci Bull 2018;63:1177–9.
[3] Intergovernmental Panel on Climate Change. Climate change 2013: the physical science basis. In: Stocker TF, Qin D, Plattner GK, et al., editors.