A Preliminary Study on a Method for the Morphological and Quantitative Analyses of Individual Snow Crystals and Its Application for Field Measurement

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ABSTRACT

The main aim of this study is to establish methods of morphological preservation and elemental quantification for individual snow crystals. Individual snow crystals were collected at a height of 20 m above ground level. To stabilize and preserve the original morphologies of the snow crystals, cyanoacrylate, which has been used to fix liquid droplets, was applied (Kasahara et al., 2000). Several different kinds of snow crystals (dendrite, sectored plate, quasi-sectored plate, and hexagonal plate) were successively stabilized using this method. The stabilized snow crystals were pretreated with acetone, and then the elemental components contained in a whole snow crystal were quantified with the Particle Induced X-ray Emission (PIXE) analytical technique. The snow crystal residual composition determined in the present study was dominated by sulfur and mineral components, and the elemental mass showed an apparent crystal size dependence, where the elemental mass gradually decreased as the crystal size increased.

Key words: Snow crystal, Snow scavenging, Stabilization, Elements, PIXE, Particle

1. INTRODUCTION

Air pollutants are ultimately removed from the atmosphere by the natural processes generally referred to as wet precipitation and dry deposition. Wet precipitation is the removal of air pollutant from the atmosphere in various types of precipitation such as rain, fog, snow, etc. Aerosols penetrate cloud droplets or ice crystals through the nucleation process, acting as cloud condensation or ice nuclei, and secondly through the process of impaction with cloud droplets or ice crystals (Pruppacher and Klett, 1997). A fraction of these droplets and crystals will then precipitate and fall from the atmo-

sphere (Croft et al., 2010).

Many previous studies have reported that snow is a more efficient particle scavenger than rain (Croft et al., 2010; Ma et al., 2003; Murakami et al., 1985; Magono et al., 1974). In addition to the nucleation particles, snow can capture and transfer large quantities of particulate matter from the atmosphere. Moreover, snow influences the earth radiation budget due to its optical reflectivity and absorptivity. Even though Bohren's critique (1986) concluded that there were inherent uncertainties in the effect of BC on snow albedo, Warren and Wiscombe (1985) stated a theory relating snow albedo to BC amount, which combines Mie scattering and a multiple scattering approximation, accounting for measured albedo as a function of ice crystal size at wavelengths where absorption by ice is dominant ($\lambda > 0.7\,\mu m$).

Even though ice crystal formation and snow precipitation are important environmental aerosol removal mechanisms, we still do not know the mechanisms underlying snow scavenging of aerosol particles, and we have not achieved sufficient experimental demonstration of these mechanisms, including field measurements.

Above all, the unsolved problems surrounding snow scavenging are due to the complicated shapes of snow crystals, making the experimental and theoretical analyses of snow crystal more difficult than those with raindrops (Hansen and Nazarenko, 2004; Warren and Wiscombe, 1985).

Since snow scavenging is also determined by ice nuclei and through the process of impaction with ice crystals at the individual particle level, an process that uses a bulk technique to describe these complex interactions will not allow the acquisition of the data necessary to improve our understanding of the processes underlying snow scavenging. A few previous studies have measured and characterized the chemical compositions of individual ice crystals (Targino et al., 2006; Ma et al., 2003; Seifert et al., 2002; Petzold et al.,

1998). However, a collaborative approach for the morphological preservation and secondary elemental quantification of individual snow crystals has not been reported.

The principal goal of this paper is to contribute to the methods of morphological preservation and elemental quantification of individual snow crystals collected on the ground. This study is also aimed at providing information that will improve our understanding of ice-nucleation and scavenging of pollutants by snow.

2. EXPERIMENTAL METHODS

2.1 Pre-Examination of the Solidification of Snow Crystals

It is possible to take pictures of natural snow crystals and snowflakes deposited on a sampling board. However, since snow crystals easily melt at room temperature, the stabilization of an original crystal is necessary for the morphological analysis of individual snow crystals. The stabilization of a snow crystal can help with the secondary quantificational analysis of the elements contained in a snow crystal. In order to preserve the original morphology of a snow crystal, stabilization is performed by applying cyanoacrylate, which has also been used to fix liquid droplets (Kasahara *et al.*, 2003, 2000).

Cyanoacrylate is the generic name for cyanoacrylate-based fast-acting adhesives. Cyanoacrylate is an acrylic resin that rapidly polymerizes in the presence of hydroxide ions. The details of this crystal solidification technique have previously been described (Carter and Hasegawa, 1975).

Although cyanoacrylate rapidly polymerizes in the presence of water (specifically hydroxide ions), it is debatable whether cyanoacrylate can react well with materials that have a high concentration of solutes and a low temperature. Hence, in order to examine the effects of concentration of inorganic, water-soluble compounds and solution temperature on solidification, a laboratory experiment was performed. Since the PIXE method applied in this study cannot identify the light-weight elements ($Z < 11$), KCl solutions were prepared by diluting a stock KCl standard solution (MonotaRO Co., Ltd.). According to the study of Ishii and Akitaya (1992), the solution concentration of bulk snow that falls in Sapporo, Japan varies between 0.84 and 34.46 mg L^{-1}. Thus, KCl standard solution was adjusted to include this concentration range, and the solution temperature varied from 0 to 44.5°C.

As mentioned earlier, cyanoacrylate forms a very strong adhesive whenever it is exposed to water. When a drop of cyanoacrylate falls onto the solution surface, it changes from a monomer to a polymer that binds to the surface water layer. The time required to complete solidification was defined as the time needed for the surface water to become white.

2.2 Collection and Stabilization of Individual Snow Crystals

Sampling of individual snow crystals was performed at a height of 20 m above ground level of the Fukuoka Women's University building (33.40°N; 130.26°E) on January 28, 2009. During the sampling period, the temperature was around −1.1°C and the average relative humidity was 64%.

For the collection of snow crystals, an acryl sampling plate (180 × 180 mm), consisting of four Petri dishes (47 mm diameter) and equipped with legs (1.2 m in height), was designed. The procedures used for snow crystal collection and stabilization were as follows:

(1) The sampling instrument was set up and pre-conditioned to the ambient outdoor temperature.
(2) Ice crystals were allowed to settle on the surface of Mylar® film (30 × 30 mm with 10 μm thickness) placed in each Petri dish.
(3) The sampling instrument was carefully moved to a laboratory clean chamber that could maintain an internal temperature of 0°C in order to prevent crystals from melting.
(4) Cyanoacrylate was warmed to 30°C to accelerate the generation of vapor.
(5) Fumes from the pads coated with a thin layer of cyanoacrylate were produced and exposed to snow crystals.

Even though snow falls as a snowflake (i.e., several agglomerated snow crystals), which is the typical shape of snowfall at ground level, about 150 individual snow crystals were collected before they melted.

Fig. 1 is a schematic illustration of the stabilization processes of individual snow crystals using α-cyanoacrylate vapor.

2.3 Pretreatment of the Solidified Snow Crystals

As a special-feature trial in this study, the quantitative analysis of elemental components contained in a whole snow crystal was performed using the PIXE analytical technique. Although the stabilization of snow crystals with cyanoacrylate has significant merit, it is questionable whether the solidified snow crystal can then be analyzed by PIXE. The self-absorption of X-rays should be considered because snow crystals can range in thickness from several tens to several hundreds of micrometers (Ji and Wang, 1999).

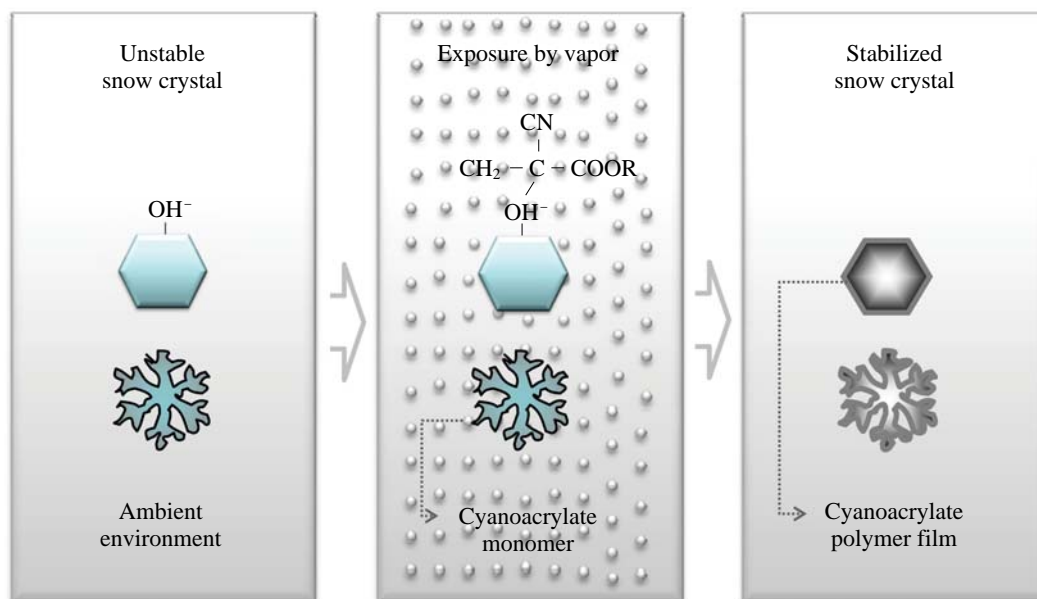Self-absorption of primary X-rays and emitted char-

Fig. 1. Schematic illustration of stabilization processes of individual snow crystals by the Cyanoacrylate vapor.

acteristic X-rays may be caused by the relative thickness of a sample (Liang *et al*., 2008). Hence, in order to clearly elucidate the chemical properties of a snow crystal, the individual snow crystals stabilized by cyanoacrylate vapor were melted with acetone after microscopic examination. Acetone is a widely available solvent capable of softening cured cyanoacrylate (Moschos and Droutsas, 1997).

2.4 Elemental Analysis of Individual Snow Crystals

After evaporation with acetone, the pretreated samples were placed on the PIXE sample holder and were irradiated with the proton beam of PIXE. PIXE is a powerful non-destructive elemental analysis technique used routinely by geologists, archaeologists, art conservators, environmental researchers, etc.

In this study, the elemental concentrations of the individual snow crystals were determined by a PIXE instrument installed at the Cyclotron Research Center of Iwate Medical University. This PIXE analytical system has great advantages such as excellent sensitivity, a nondestructive nature, the analysis of a wide range of elements ($Z > 10$), and a short measuring time (3-10 minutes for typical environmental samples). The beam spot size (about 3 mm) of PIXE is suitable for the detection of the elemental composition of a whole snow crystal. Furthermore, the PIXE used in the current study can provide absolute mass concentrations for multiple elements.

The more detailed analytical procedures and experi-

mental set-up used for the PIXE analytical system have been previously described (Sera *et al*., 1999).

3. RESULTS AND DISCUSSION

3.1 Stabilizibility of Cyanoacrylate Vapor

The stabilization process depends not only on the concentration and temperature of the objective materials, but also the temperature of the cyanoacrylate. High temperatures may result in overexposure to cyanoacrylate fumes and polymerization failure on the surface of the snow crystal. Low temperatures may result in failure to quickly polymerize, requiring a longer processing time.

The time required for complete solidification as a function of KCl solution and cyanoacrylate properties is shown in Fig. 2. As shown in the figure, the solidification time for surface water was dependent on the solution temperature; the solidification time increased as the solution temperature decreased. The solidification time of surface water varied between 3 and 12 seconds depending on the solution temperature (0.1 mol L^{-1} of KCl solution). The time to complete solidification was relatively short (12 seconds) even when the solution was at a temperature of 0°C. There was also slight variation in solidification time (from 12 to 15 seconds) with solution concentration (fixed at a 0°C solution temperature). Meanwhile, there was a critical effect of cyanoacrylate temperature (fixed at 0.1 mol L^{-1} of KCl and 0°C solution temperature) on the time
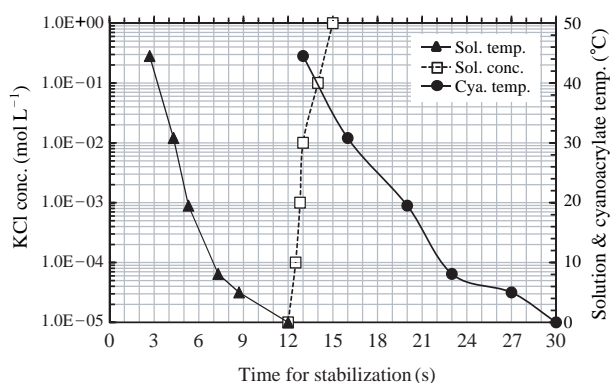
Fig. 2. The time required to complete solidification as the functions of KCl solution and cyanoacrylate properties.

for crystal stabilization. With no acceleration of vapor generation by heating, the successful stabilization of crystal takes at least 30 seconds. The warming of cyanoacrylate can therefore result in sufficient vapor generation and rapid stabilization of snow crystals.

3.2 Originally Stabilized Individual Snow Crystals

As mentioned earlier, though snow crystals were usually collected as snowflakes during ground-level measurement, non-agglomerated individual crystals were also observed. These individual snow crystals with various shapes were successfully stabilized in their original sizes and shapes. Fig. 3 illustrates several different kinds of white polymerized (polycyanoacrylate) snow crystals (left) and their referential morphologies (Libbrecht, 2007) (right).

The improved stabilization method (i.e., handling of snow crystals at 0°C and accelerating cyanoacrylate vapors by warming) successfully stabilized snow crystals with the original morphology.

Individual snow crystals collected in this study ranged from 0.5 mm to 1.5 mm in diameter. Hexagonal plates occurred as interconnected aggregations but were more often observed as single crystals. The dendritic form, which is the other common shape of snow crystals, occurred in numerous configurations. Dendrites normally had six arms that were often branched. Each arm of a dendritic crystal exhibited bilateral symmetry, which was accentuated by the presence of one or more pairs of branches that emanated from the main axis of the arm. The shapes of the ice crystals collected from different snow events probably vary depending on the temperature and moisture conditions that prevail during the formation and descent of the snow crystals (Rango *et al.*, 1996). Unfortunately, the detailed examination of minute surface structures such as rime
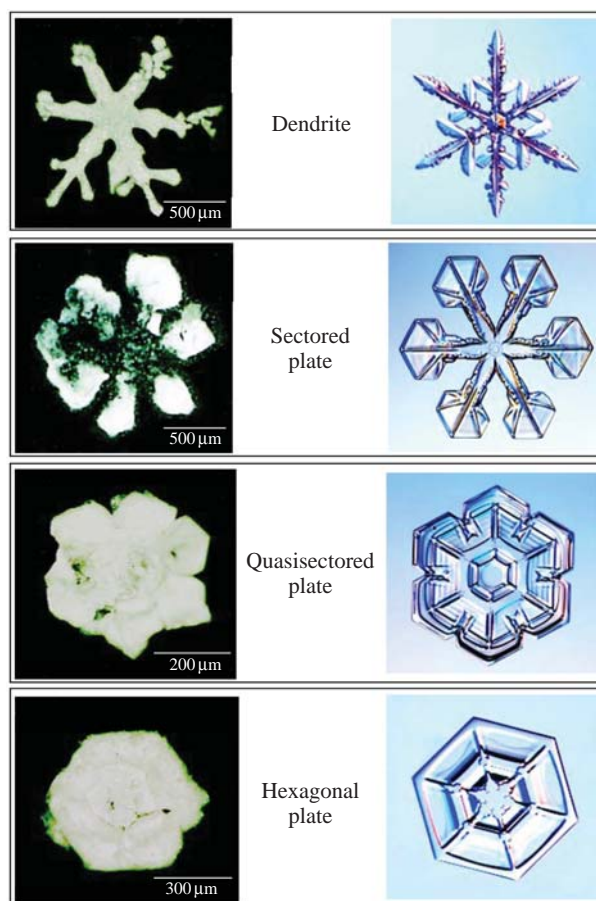

Fig. 3. Matching of several different kinds of fixed (left column) to natural (right column) snow crystals. The morphology of natural snow crystals is quoted from a reference (Libbrecht, 2007).

(supercooled cloud drops), microdroplets, short prismatic columns, and amorphous films were hampered in this study by the limited resolution of the digital microscope.

3.3 Quantification of Chemical Components in a Single Snow Crystal

Fig. 4 shows the image of a single solidified hexagonal crystal after melting with acetone. The sample pretreated with acetone was quite thin compared to the stabilized snow crystal, and thus the elemental quantification was less affected by the self-absorption of X-rays. Many particle clusters or their residues that were attached or incorporated into the falling/developing snow crystal, including nuclei, were found at the boundary and in the center portions of the melted snow crystal. These materials were the target of the PIXE analysis and were irradiated by X-rays.

Fig. 5 shows an example of the PIXE spectra of a

single hexagonal form snow crystal sized with a 0.7 mm circumscribed circle and a Mylar® film that was applied as backing material. It was possible to resolve several significant elemental peaks (i.e., sulfur, potassium, calcium, and iron) corresponding to the channel numbers of the PIXE spectrum. Details about these elemental compositions are discussed in Fig. 6. Relatively distinct peaks of calcium and iron were also found in the blank spectrum. These background peaks were presumably derived from impurities in the cyano-
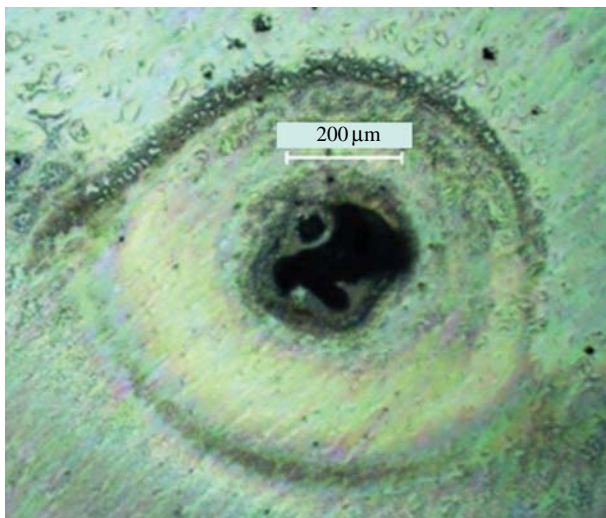
acrylate and the Mylar® polycarbonate film and from the metallic materials in the PIXE sample chamber. However, these unwanted background levels were corrected using the ratio of the integrated level of background counts to that of signal.

A laboratory study by Mitra *et al*. (1990) suggested that columnar crystals generally have the lowest concentrations of scavenged materials, while dendritic sectored crystals have the highest. This indicates that the efficiency with which snow scavenges pollutants is dependent on crystal-shape.

In the present study, in order to estimate the crystal size dependence of scavenging efficiency, a thin hexagonal plate, which is the most basic snow crystal geometry, was selected as the target of PIXE analysis. Fig. 6 shows the mass variation of four elements as a function of snow crystal size. The size of a snow crystal corresponds to the diameter of the circle circumscribed onto a planar hexagonal crystal. The mass data were based on the average values of 10-15 snow crystals. Sulfur amounts ranged from 20.7-32.5 ng, making it the most abundant element in the individual crystals. The crust origin components such as calcium, potassium, and iron were also commonly detected in mass levels varying from 14.2 to 28.4 ng, 6.5 to 19.8 ng, and 4.8 to 22.8 ng, respectively.

Using optical and electron microscopes, Magono *et al*. (1979) observed thousands of aerosol particles, including soot, in a typical snow crystal in Sapporo. Although the light-weight elements (Z < 11), including carbon, could not analyzed in the present study, PIXE
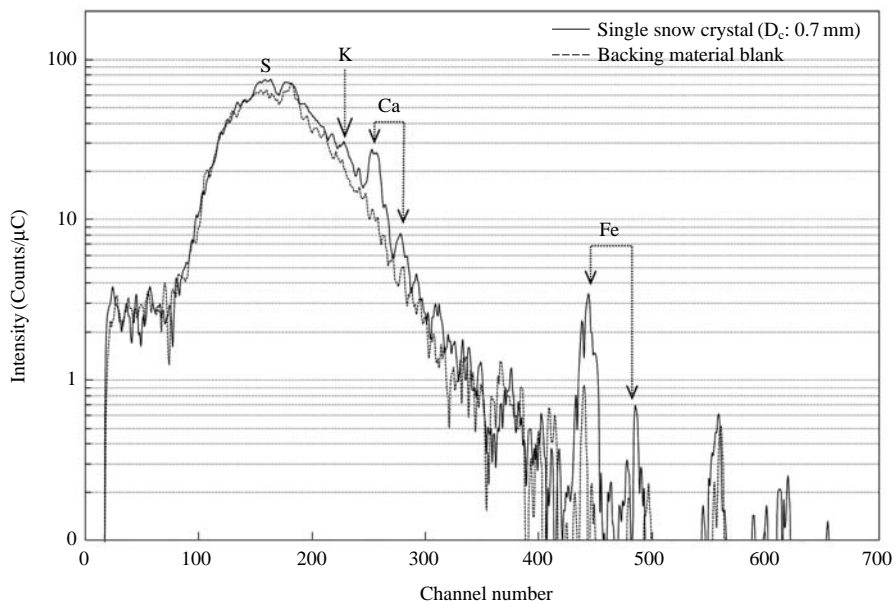

Fig. 4. Image of a single solidified snow crystal after melting by acetone.

Fig. 5. PIXE spectra of a single hexagonal form snow crystal and a Mylar® film which was applied as backing material. D_c means the diameter measured by the circumscribed circle.

analytical results suggested that sulfur had a strong impact on ice formation through heterogeneous nucleation in clouds as well as below-cloud snow scavenging. Sulfur showed a significantly high mass level regardless of raindrop size. It is difficult to establish the
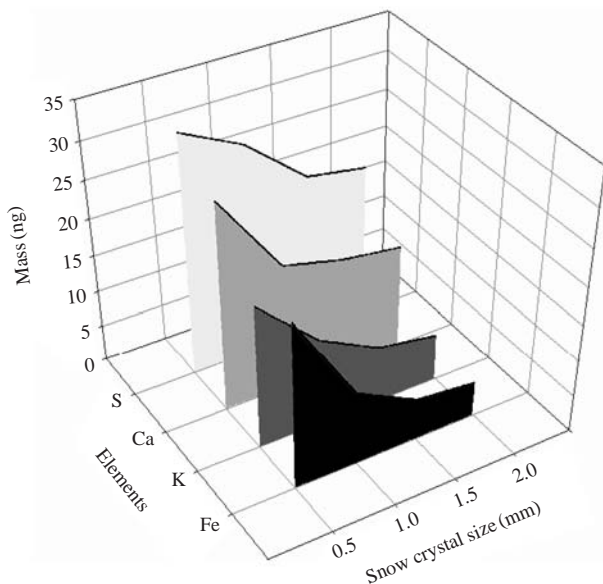

Fig. 6. Crystal size dependence of major elemental masses in hexagonal crystals.

sources of sulfur, but one possibility is that a falling snow crystal provides a suitable surface for the absorption of gaseous SO_2 as well as for collision with submicrometer sulfates. The results of the current study also indicate that a large number of soil origin particles were incorporated into the ice crystals, and these mineral particles were also effectively absorbed by the snow crystals during a snowfall event.

Every elemental mass determined in the current study showed an apparent crystal size dependence; elemental mass decreased gradually as crystal size increased. In a previous theoretical study (Flossmann *et al.*, 1985) and another field measurement (Munger *et al.*, 1989), this dependence of chemical components on crystal size in a cloud droplet was not observed. It can be therefore said that the snow crystal size-dependence of elemental concentration is caused by the pollutant washout of snow crystals below cloud level.

Slinn (1974) suggested that the rate at which ice crystals scavenge particles can be determined by the following equation:

$$\Psi_s(\gamma, t, \alpha) \cong \frac{p(\gamma, t)}{v_t^2} \varepsilon(\alpha, l),$$

where $p(\gamma, t)$ is the precipitation rate in rainwater equivalents (γ) per time (t), v_t is the average settling speed of the snowflakes, and $\varepsilon(\alpha, l)$ is the particle (α)/ice crystal (l) collection efficiency, in which l is a charac-
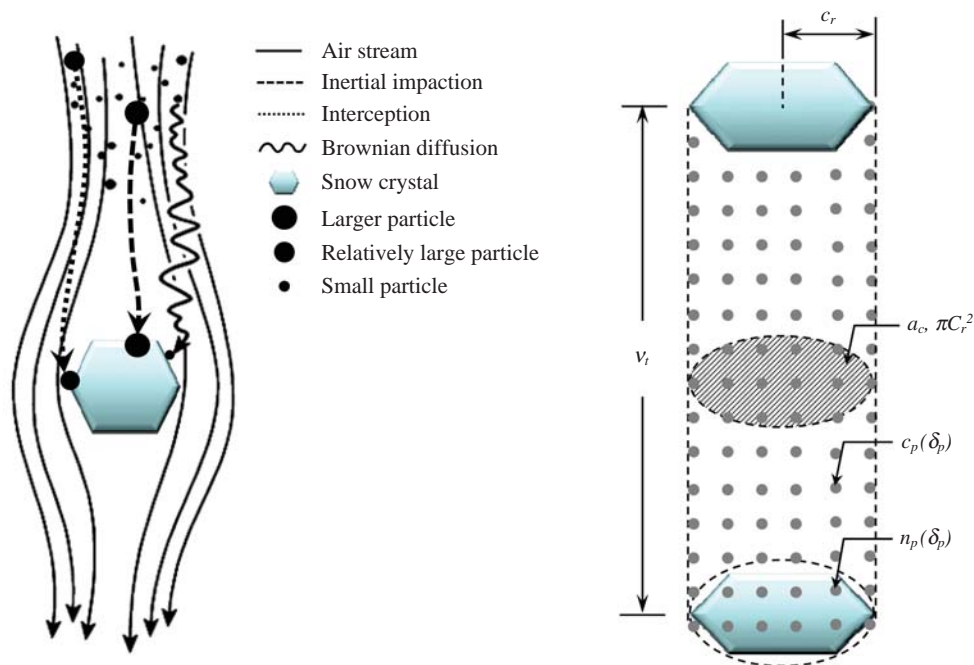

Fig. 7. The conceptional illustrations of particle collision mechanisms (left) and the collided particle number (right) by a falling snow crystal.

teristic dimension of the collecting element in the ice crystal and is not necessarily related to the overall size of the ice crystal.

According to this equation, if the snow crystal collects more ambient particles, it may simply be that the snow crystal has traveled a longer distance and hence has had more opportunity to collect pollutants. Another possibility is that the efficiency with which snow crystals scavenge ambient particles is dependent on crystal size.

The collection efficiency of snow crystals for aerosol particles can be defined by the ratio of the number of aerosol particles captured by a snow crystal to the number of aerosol particles existing in a volume swept by falling snow crystals. According to this definition and a conceptional illustration of particle collision mechanisms for a falling snow crystal (Fig. 7), the aerosol particle collection efficiency of snow crystals, E_p, can be described by the following equation (Mitra *et al.*, 1990):

$$E_p(\delta_p) = \frac{n_p(\delta_p)}{c_p(\delta_p) \cdot a_c \cdot d_f},$$

where δ_p is the diameter of aerosol particles, $n_p(\delta_p)$ is the number of aerosol particles with size δ_p captured by a snow crystal, $c_p(\delta_p)$ is the concentration of aerosol particles with size δ_p in the atmosphere, a_c is the area of the circumscribed circle of snow crystal, and d_f is the distance from the bottom of the cloud to the ground integrated by v_t, as shown in Fig. 7.

According to this equation, the collection efficiency of aerosol particles by snow crystals is inversely proportional to the area of the snow crystal. Sauter and Wang (1989) reported that this collection efficiency could be explained by the relative strengths of the inertial forces of the particles and the hydrodynamic drag force created by the descent of the snow crystal. Large crystals create greater drag forces during the fall and force the aerosol particles to more closely follow streamlines, hence reducing the collision efficiency. Their experimental study of natural snow crystals suggests that particles are less likely to collide with a larger crystal, regardless of crystal shape. This may explain why the elemental masses determined in the present study decreased with increasing snow crystal size.

Meanwhile, some researchers (Ehrman *et al.*, 1998; Mosimann *et al.*, 1994) have suggested that the compositions of snow crystals also depend on cloud microphysics such as extent of collision-coalescence, riming, etc. Although the details are still under discussion, it is also known that aerosol chemical composition is a factor in ice crystal nucleation and is also very important during below-cloud scavenging, which occurs after nucleation.

4. CONCLUSIONS

Since snow crystals are susceptible to sublimation and melting, it is difficult to study the sizes and shapes of snow crystals using a frozen specimen. Furthermore, it is difficult, if not impossible, to conduct a proper chemical analysis after a morphological study. However, quantifying the residual composition of a snow crystal (or a whole snow crystal) is an important task for understanding crystal formation processes and heating/cooling rates. From this practical standpoint, a tentative attempt at the morphological and quantitical analyses of individual snow crystals was made. Using a crystal stabilization technique, the original shape of the unagglomerated individual snow crystal can be preserved. As a special feature of this study, the stabilized individual snow crystals were successfully analyzed by PIXE. However, the optical determination of minute surface structures of stabilized crystals and the quantification of light elements including carbon could not be accomplished in the current study. While some problems remain unsolved, the collaborative approach of a stabilization technique with PIXE analysis is a suitable method for the study of the physical characteristics as well as for the subsequent chemical analysis of individual snow crystals. Since cyanoacrylate contains carbon, and it is not easy to discriminate carbon from the residues of a snow crystal, more research is necessary to develop a better stabilization method. Further study is being planned to quantify other light elements using an element analyzer such as SEM-EDX.

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