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Measurement of the Single and Size-Classified Raindrops

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Abstract

To characterize the single raindrops as a function of their size we designed the sampling and handling apparatuses. Samplings of single and size - classified raindrops were performed at a height of 20 m above the ground level of a Kyoto University building located in Uji, Japan in rain events from middle of July to the end of August, 1999. And PIXE method was applied to the analysis of single raindrops sampled as a function of their size. Diameter change of frozen raindrops by liquid nitrogen did not affect the size segregation ability of our sampling apparatus. The number of raindrops increases with decreasing drop size. And it is found that the size distribution of raindrops verified depends on the rain events. Application of PIXE analysis to the measurement of single raindrops was very successful. Every element showed a continuous increase in concentration with decreasing raindrop diameter. It seems reasonable to say that our work should be helpful to obtain more detailed information on single raindrops and especially to study on the rainout and washout mechanisms.

Key words : single raindrop, PIXE analysis, wet precipitation, pollutants scavenging

1. INTRODUCTION

Removal of atmospheric aerosols and gaseous materials by precipitation is one of the most important natural cleaning mechanisms. Unfortunately, physical and chemical parameters, which influence wet deposition, are not well understood because wet deposition is a complex multiphase system. The analysis of single and size-classified raindrops is expected to give new and interesting information about anthropogenic air pollution and drop formation processes in the atmosphere.

At a specific relative humidity, which for most soluble components in the atmosphere is far below 100%, aerosol particles are not already liquid deliquesce into aqueous solution drops. As the relative humidity keeps increasing, the size of droplets increases in accordance with a water vapor equilibrium. If the relative humidity of the air parcel reaches a critical humidity, the value of which depends on the size and chemical composition of the aerosol present, the droplets become activated and grow freely by water vapor diffusion, and cloud and fog are formed. In addition to having solute concentration differences that arise from differences in the composition of the condensation nuclei, fog, and cloud droplets scavenge soluble gases like nitric acid and ammonia and act as a medium for various aqueous-phase reactions, including the oxidation of absorbed

SO_2 to sulfate (Spyros N. *et al.*, 1990).

Chemical processes in the atmosphere as the washout of particles and gases, the damage of the biological system and the change of the radiation balance are not sufficiently described by the usual determination of elemental concentration in rainwater (K. Bachmann *et al.*, 1993). Chemical content of raindrops is variable according to the mechanisms such as condensation nuclei inside clouds, pollutants scavenging, collision, coalescence and break up of falling raindrops, and evaporation.

In the past many investigations about the determination of the chemical components in rainwater have been made. The analysis was always carried out with bulk sample. But detail information cannot be obtained by this bulk sample analysis because the chemical content of raindrops depends on the drop size (B. Tenberken *et al.*, 1996).

It is strongly required that the sampling and analysis of single raindrops must be practiced. The analysis of single raindrops is expected to give new and great information about drop formation processes in the atmosphere and rainout and washout mechanisms. Although it is possible to obtain more detailed information by analysis of single and size-classified raindrops, there are some difficulties in sampling and analytical processes. Careful and skillful handling techniques are needed because it is possible to occur the evaporation and contamination.

In this study, to determine the characteristics of single raindrops as a function of their size we designed a sampling and handling apparatus. Added to this, an attempt was made to analyze the single raindrops by PIXE.

2. MATERIAL AND METHODS

2. 1 Sampling of single raindrops as a function of their size

For the sampling of single raindrops as a function of their size, we designed the sampling and handling apparatuses. Sampling apparatus is consist of a dewar vacuum flask filled with liquid nitrogen and 7-stage stainless steel sieves (Nonaka Rikaki Co.). A schematic

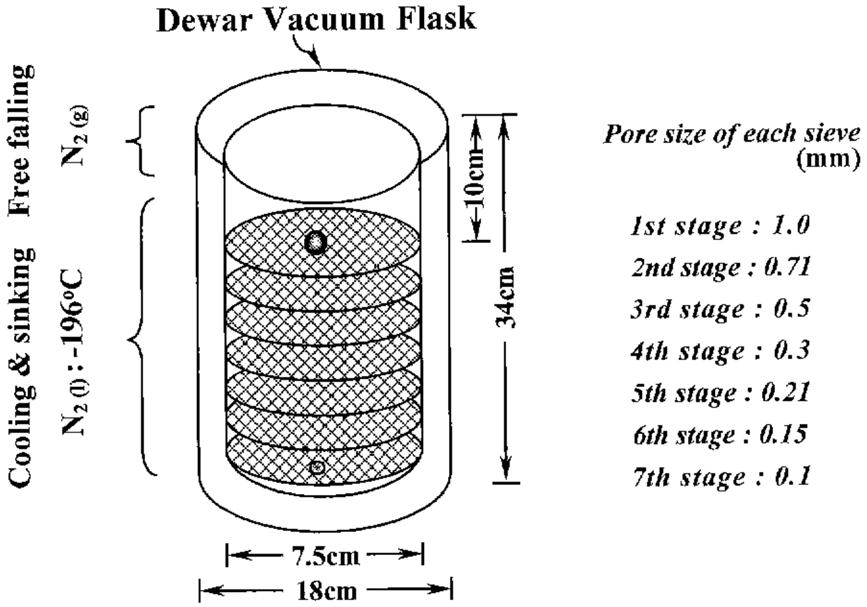


Fig. 1. Diagram of sampling apparatus for the collection of single raindrops as a function of their size.

diagram of the sampling apparatus is shown in the following Fig. 1. Sampling of single raindrops as a function of their size were performed at a height of 20 m above ground level of the Kyoto University building located in Uji, Japan in rain events from middle of July to end of August, 1999. Raindrops were collected for about 3 ~ 10 min according to the rate of rainfall.

Fallen raindrops into the liquid nitrogen are frozen and they sink to lower sieves owing to their higher density. The raindrops keep their spherical shape during the freezing process, and consequently by using stainless steel sieves of different mesh widths (0.106, 0.15, 0.212, 0.3, 0.5, 0.71, 1.0 mm) it is possible to separate the frozen raindrops according to their sizes.

2. 2 Handling of single raindrops

For handling of single raindrops without any evaporation and contamination, we designed a clean air chamber system. After sampling, the sieves were pulled out from the dewar vacuum flask and one of the frozen raindrops on each sieves was placed onto the non-hole nuclepore filter by using vacuum pipette (HAKO 392). Unfortunately, small single raindrops, which are smaller than 0.212 mm, can not be handled.

Every process was performed in the clean air system, which was filled with the cooling nitrogen gas. Handling process of single raindrops was shown in Fig. 2. A frozen raindrop on the filter was melted and dried under an infrared lamp. Fig. 3 shows the digital microscopic image of separated single raindrops collected on the sieve and a single raindrop (ϕ 0.71 mm) stain after dry by infrared lamp. Non-volatile compounds included in stain were analyzed by PIXE. The diameters of the every stain of single raindrops are in the range of 0.3 ~ 2.5 mm, therefore every portion of stain can be irradiated by 6 mm diameter proton beam of PIXE.

2. 3 Analysis of single raindrops

For analysis of single raindrops, Particle Induced X-ray Emission (PIXE) analysis was applied. PIXE analysis was performed with a proton beam of 6 mm diameter and 2.0 MeV energy from a Tandem Cockcroft accelerator. Beam intensities from 10 to 60 nA were employed and the total dose were about 20 μC. X-ray with an energy of 1.4 ~ 1.8 keV emitted from the target were detected by a Si (Li) detector which had a resolution of 152 eV at 5.9 keV. The target and detec-

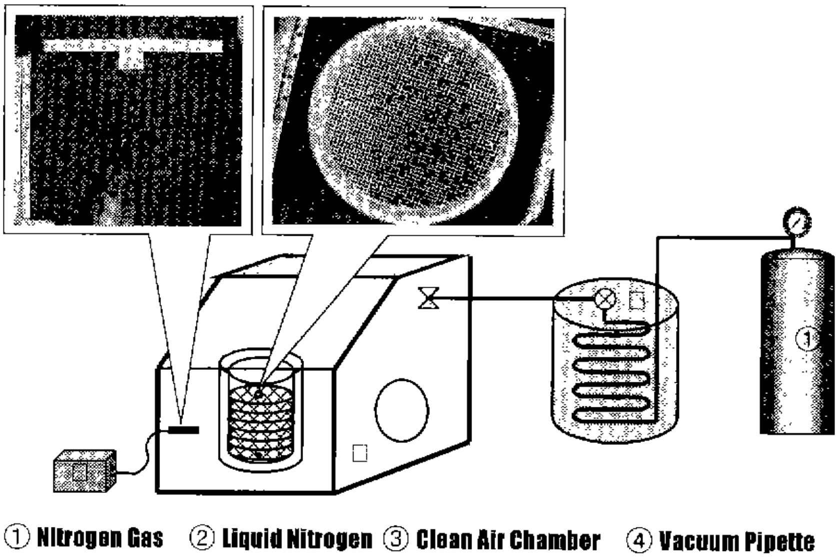
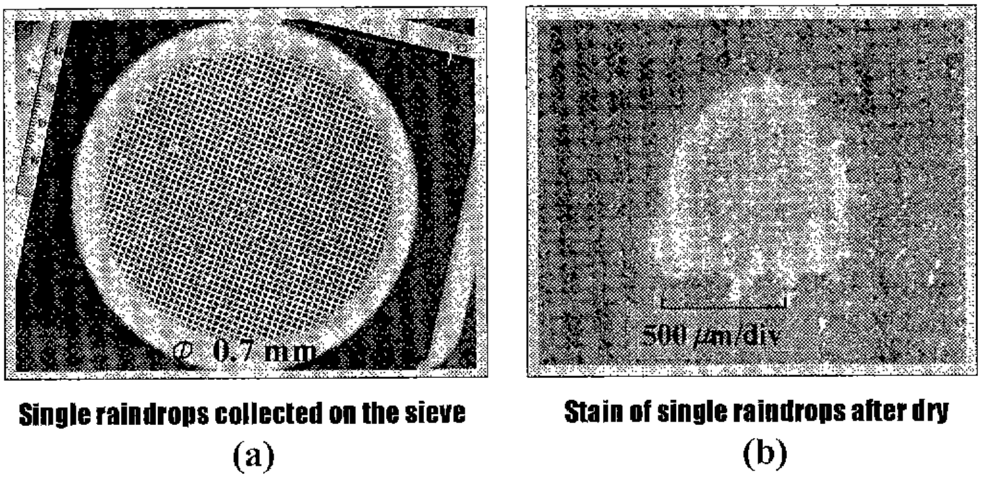


Fig. 2. Handling of single raindrops.

tor were set at 90° and 135° with respect to the direction of the ion beam, respectively. An absorber of 39.3 μm thick Mylar film was set between the target and detector to control counting efficiency of the lighter elements. The count rates for X-rays were kept below 1000 pulses per second. Fig. 4 shows the sche-matic diagram of PIXE. The more detailed analytical procedures and experimental set-ups used for PIXE was described elsewhere (M. Kasahara *et al.*, 1996).

The masses of 15 elements (Si, S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn. Br. and Pb) in single raindrops were determined. PIXE spectrum of a single raindrop



Single raindrops collected on the sieve (a)

Stain of single raindrops after dry (b)

Fig. 3. Digital microscopic image of separated single raindrops collected on the sieve (a) and a single raindrop (f 0.71 mm) stain after dry by infrared lamp (b).

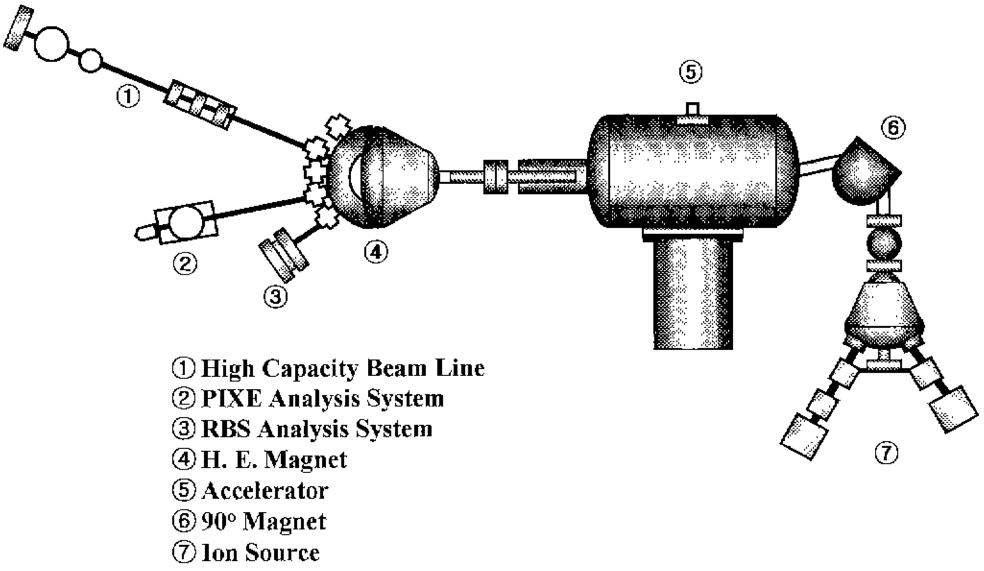


Fig. 4. Schematic diagram of PIXE.

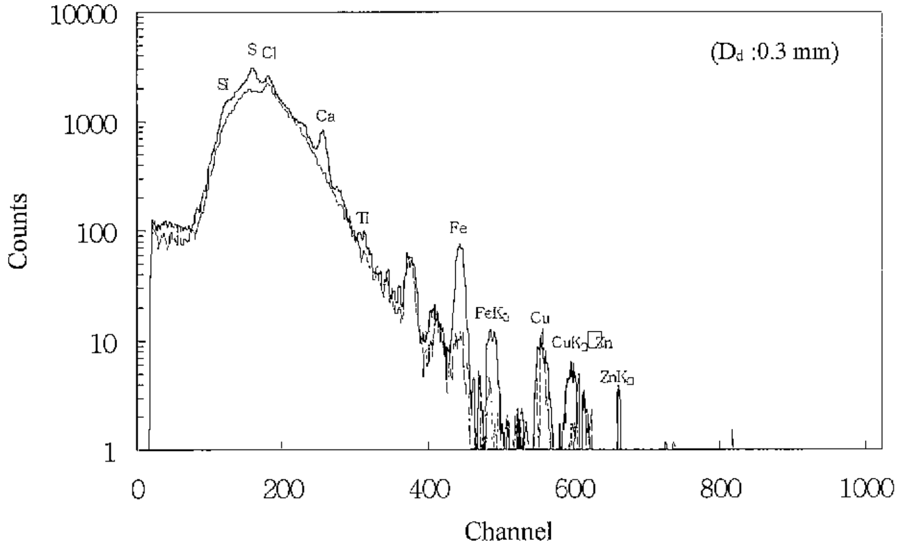


Fig. 5. PIXE spectrum of a single raindrop (ϕ 0.3 mm).

is shown in the Fig. 5.

3. RESULTS AND DISCUSSIONS

It is debatable point that raindrops can be segregated accurately by sampling apparatus designed by our own self. To examine the diameter change of droplets after freezing, each calculated amount (8.81 μl, 14.121 μl, 22.44 μl) of bulk rainwater which simultaneously sampled was frozen by liquid nitrogen. And then diameters of more than 60 single frozen droplets were measured at least 2 times by Digital microscope (KEYENCE, VH-7000). Fig. 6 shows the diameter change of droplets after freezing by liquid nitrogen. Even though, 30~35% of droplets show lager 0.1 mm than calculated diameter. A large percentage of the droplets shows good agreement with the calculated diameter. Consequently, it can be said that diameter change of frozen raindrops by liquid nitrogen did not affect the size segregation of our sampling apparatus.

To investigate the size distribution of single raindrops, samplings were performed in short time (2 min.). After sampling, we took a picture of each sieve by using Digital microscope which has data saving and print out functions. It was possible to count the separated raindrops on the each sieve.

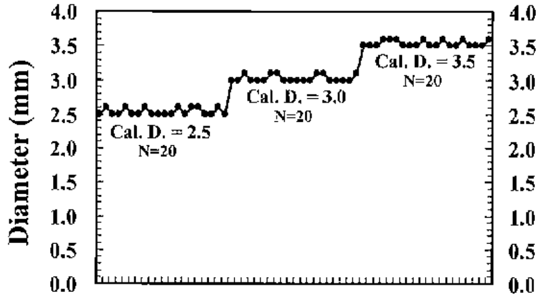


Fig. 6. Diameter change of droplets after freezing by liquid nitrogen.

Fig. 7 illustrates size distribution of raindrops for different rate of rainfall. The number of raindrops increases with decreasing drop size in both rain events. Especially, sudden change of raindrop number was found in the range of small raindrops. Consequently, it is found that the size distribution of raindrops verified depends on the rain intensity. Unfortunately, the number of 0.21 mm raindrops in second rain event could not be counted because they were piled up.

So far, in the study of simulation model for the scav-

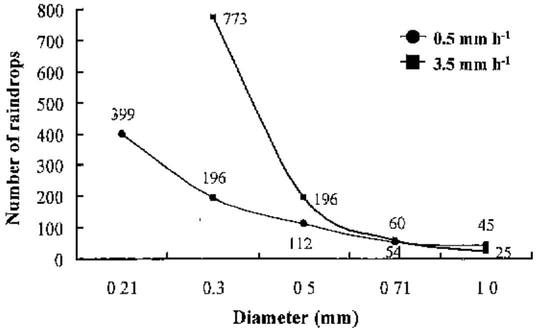


Fig. 7. Size distribution of raindrops for different rain intensity.

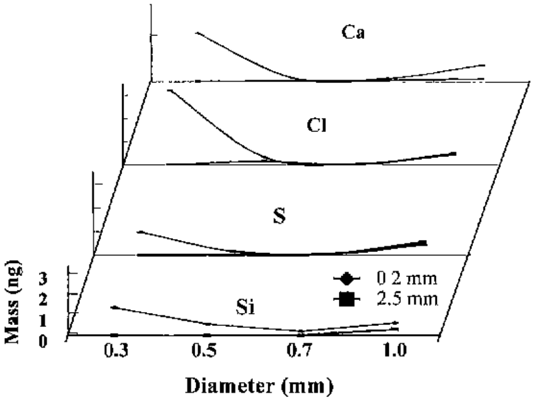


Fig. 8. Variation of major elements masses as a function of raindrop diameter.

enging of atmospheric aerosols by raindrops, the size distribution of raindrops was assumed to be normal distribution and the radius of raindrops was presumed from the rain intensity or other methods.

Consequently, we can reasonably say that the results of our work should be helpful to establish the more precise simulation model for scavenging atmospheric pollutants by raindrops.

Fig. 8. shows each mass of major elements as a function of raindrop diameter. Every element showed a continuous increase in concentration with decreasing diameter. Especially, there was a marked increase in the range of between 0.5 mm and 0.3 mm diameter. On the other hand, slight increase was found between 0.7 mm and 1.0 mm diameter.

Even though little is known at present on the reasons for the elemental mass variation with the drop sizes, it is expected that several mechanisms are responsible for the variation of the elemental mass as a function of drop size. During rainfall the single raindrops can take up gases such as SO_2, HNO_3, NH_3, and aerosols. Aerosol origin components such as Ca, Cl, Na, K, Na and Mg are taken up by aerosol scavenging. Also, the chemical reactions of the scavenged compounds must be considered.

It can be said that smaller raindrops should have higher elemental concentration because they have lower velocities and consequently longer lifetimes than larger ones. And the reason of high concentration in small raindrops might be caused by the effect of evaporation, i.e. small raindrops show a much higher degree of evaporation than larger ones which lead to an increase of the elemental concentration.

4. CONCLUSION

To investigate the characteristics of single raindrops as a function of their size we designed the sampling and handling apparatuses. Diameter change of frozen raindrops by liquid nitrogen did not affect the size segregation ability of our sampling apparatus. The number of raindrops increases with decreasing drop size. And it is found that the size distribution of raindrops verified depends on the rain intensity. We can reasonably say that the data about size distribution and diameter of raindrops should be helpful to establish the more precise simulation model for the scavenge of atmospheric pollutants by raindrops. PIXE method is applied to the analysis of single raindrops sampled as a function of their size. Every element showed a continuous increase in concentration with decreasing diameter. Although the experimental data set was not sufficient to discuss results quantitatively, some trends were available. Further study such as measurement and microanalysis of single raindrops as a function of rain rate, samplings at below the cloud raindrops are in progress.

Consequently, it is concluded that our work should be helpful to get more detailed information on single raindrops and especially to study the rainout and washout mechanisms.

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