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## COMPONENT ANALYSIS AND CLASSIFICATION OF AEROSOL USING TOF-SIMS

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Very small particles called aerosol are suspended in air. In general, bulk analysis is performed on lumped aerosols with various sources after the filter collection. The average composition of the collected particles can be obtained, but it cannot be obtained for each particles. On the other hand, surface analysis techniques such as TOF-SIMS has opposite performance. In this study, we proposed a method to perform the lumped aerosol analysis included the individual particle information from SEM images. However, in this method the accuracy of individual information is not sufficient compared with TOF-SIMS. In this study, shape cluster analysis using SEM images was compared with component cluster analysis using TOF-SIMS. In order to confirm the correlation between the shape and the components, the particles which were recognized as the same types were selected, and both cluster analyses were performed using a total of 298 particles. Therefore, we found particles that cannot be separated by the current method using shape clusters, and proposed a solution. With this method, if there is a correlation between the shape and type from detailed investigation by TOF-SIMS, it can be used as a novel monitoring technique.

#### I. Introduction

There are many particles of invisible size in the atmosphere where we live in. Those fine solid or liquid particles suspending in air are called an aerosol. Aerosols come from generation source of natural origin, such as pollen and yellow-sand, and from urban areas, such as from railways and cars. In recent years, by the economic development of Asia, aerosols emitted from industrial areas have become a large factor in air pollution. As for aerosols having various generation source as described above, the components are analyzed by a filter and then subjected to a bulk analysis. In bulk analysis, quantitative

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data can be obtained with high accuracy and can be performed relatively quickly. However, since only the average composition of the collected particles is known, it is impossible to identify the source focusing on individual particles or analyze the floating history in principle <sup>1)</sup>.

In our research group, we developed an FIB-TOF-SIMS (focused ion beam time-offlight secondary ion mass spectrometry) apparatus that can analyze individual particles of aerosol, the component distribution of the surface structure and internal structure of individual particles has been analyzed <sup>2)</sup>. On the other hand, in order to ensure reliability of statistical information, aerosol analysis requires a large number of individual particle data. However, TOF-SIMS has a problem that it takes an enormous amount of time and labor to perform a large number of aerosol analyzes. Therefore, with TOF-SIMS developed by our laboratory, field of view can be changed. Hence, though the amount of data for each particle decreases, securing throughput without losing individual particle information, we thought that widening the field of view and observing a large number of particles at once would secure statistical reliability. In addition, previous studies by TOF-SIMS have revealed a high correlation between the particle component (origin) and the particle shape. Therefore, a method was devised to acquire SEM (scanning electron microscope) images of many particles and determine the type of the particles from the shape information of each particle. According to this method, the classification and the number information of the particles can be obtained quickly and easily.

Although the method of analyzing individual particles from SEM images is quick and easy, it is not sufficient in terms of accuracy when compared with component analysis using TOF-SIMS. Therefore, in this study, we performed cluster analysis by component imaging using TOF-SIMS, and compared it with cluster analysis using SEM images. In this study, we thought if we could improve the accuracy while keeping it to a certain extent quick and easy.

#### II. Experimental

In this study, we collected aerosols using a movable stage type impactor developed in our laboratory as shown in Fig. 1. In this sampler, the sample holder is raster-movable, and aerosol particles are evenly collected in a 2 mm square, so sampling suitable for individual particle analysis is possible <sup>3)</sup>.

For component cluster analysis, component analysis was performed using FIB-TOF-SIMS developed in our laboratory <sup>2)</sup>. For shape cluster analysis, SEM images for shape information was performed using a field emission scanning electron microscope (FE-SEM, JSM-6700F, JEOL Ltd.), and information of many particles was analyzed by image analysis software (ImageJ) <sup>4)</sup>.

As a sample, the aerosol collected in Kumamoto on October 29, 2016 from 15:00 to 15:40 was used for the experiment. In this experiment, in order to confirm the correlation between the shape and the components, both cluster analyzes were performed using the same location and the same particle. Cluster analysis was performed on a total of 298 particles by selecting only those recognized as the same particles.

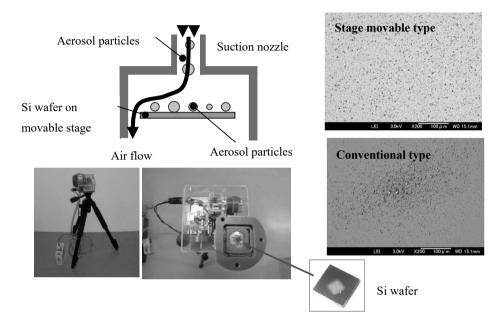


Fig. 1 Movable stage impactor

#### III. Result and Discussion

In this experiment, clusters were generated at the height of 40 in the generated dendrogram as shown in Fig. 2. In the shape cluster analysis, it was classified into 4 clusters. In the component cluster analysis, it was classified into 5 clusters. Although the number of clusters in the shape cluster analysis was one less than the component, it is considered that nitrate and sulfate were contained in the same shape cluster. In Table 1, shape and component clusters were summarized.

In this cluster analysis, particles such as dissolved yellow-sand and needle-like particles containing chlorine, which could be cluster analyzed by component, could not be cluster analyzed by shape. For example, the particles shown in Fig. 3 could be separated by component, but could not be separated by shape, despite the appearance being very different. It is considered that this can be divided into shapes by using a brightness profile as shown in the same Figure. In addition, particles with an orange frame among the particles divided into shape 4, component 2 and shape 4, component 5 in

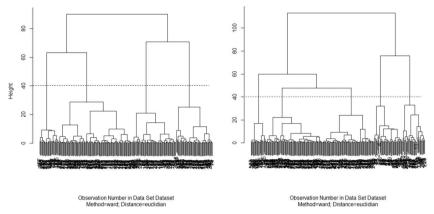


Fig. 2 Dendrogram of cluster analysis (left: shape, right: component)

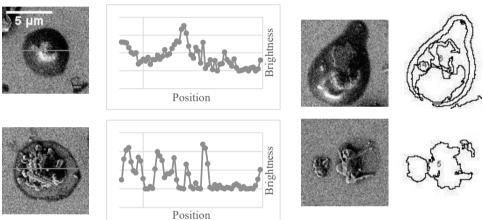


Fig. 3 Particles and brightness profile of the same cluster

Fig. 4 Particles that could not be separated by shape analysis and outlines taken by ImageJ

Table 1, as shown in Fig. 4, the outline of the particles is not well taken, Particles are sometimes stuck together and determined as the same particle. A possible solution is to avoid using particles that are not outlined as shown in (a), separately from the data set before cluster analysis. It is considered that the particles in which the particles adhere to each other as in (b) can be reduced by adjusting the sampling time.

#### **IV. Conclusion**

In the component cluster analysis using the previous method, clusters were divided according to intensity, but this time the analysis was quasi-normalized with particles having the highest Ion count, so cluster analysis was performed for each component. We also found particles that could not be separated by the current method, and proposed a solution. Examples of the application of this method, component cluster analysis can be

used statistical support for detailed investigations by TOF-SIMS for components, and shape cluster analysis can be used monitoring when there is a correlation between shape and type after investigation.

Table 1 Cluster analysis results (horizontal rows indicate shape cluster, vertical columns indicate component cluster)

	Composition 1	Composition 2	Composition 3	Composition 4	Composition 5
Shape 1	22 pieces  ε <sub>μm</sub>	4 pieces	18 pieces	22 pieces	10 pieces
Shape 2	24 pieces	10 pieces (a)	5 pieces	46 pieces	42 pieces
Shape 3	2 pieces	20 pieces	1 piece	17 pieces	23 pieces
Shape 4	2 pieces	4 pieces  (b)	3 pieces	10 pieces (c)	13 pieces (d)

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