V. 3.   Microbeam Analysis of Individual Particles in Working Environment

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Introduction

Atmospheric aerosols affect large impact on environment and human health and many studies are carrying out[1,2].   Human beings living in developed countries spend from 80 to 90% of their time in buildings.   Therefore indoor aerosols at house and in working environment can have a large impact on human health and aerosol monitoring in working environment is very important for human health.   Especially, in machine workshops and laboratories, where specific materials and chemicals are processed, toxic aerosols may be released into air.   Therefore, an aerosol monitoring in working environments is one of the essential prerequisites for an efficient health protection system for the employees.

Aerosol particles are characterized by various physical and chemical factors.   The elemental analysis of atmospheric aerosols is very important too, since distribution of different elements reflects the sources and their generating processes.   Aerosol sampling on thin films and a subsequent PIXE elemental analysis is one of the most effective combinations for studying atmospheric aerosols[1].   Such an approach is advantageous for aerosol monitoring purposes because the elemental concentrations can be studied with a reasonable time resolution[3,4].   However, in bulk analysis, the elemental concentrations are averaged over many individual particles which have different origins.   Therefore, individual particle analysis is superior to bulk analysis for obtaining source-related information and for understanding the aerosol formation mechanisms[5,6].   In the previous study, the aerosols collected by a cascade impactor set in the machine workshop of Jožef Stefan Institute were characterized by a bulk PIXE analysis and a multimodal microanalysis with microscopic

scanning[7].   While we could provide an efficient aerosol monitoring capability, the source identification was not performed completely.   This study deals with in-doors aerosols that were collected in the machine workshop of Jožef Stefan Institute.   The samples were reanalyzed by a microbeam analysis system at Tohoku University with a superior resolution and source identification was carried out.

Sampling and analysis

Aerosol particles were collected inside the machine workshop of Jožef Stefan Institute, Slovenia using a nine-stage cascade impactor (Model 1-1L; PIXE international Co. LTD). The machine workshop has three rooms: workshop room, preparation room and carpentry room, as described in the previous paper[7].   A non-stop sampling in the workshop room with the cascade impactor was conducted during two time intervals corresponding to weekdays (working regime, February 13-15, 2008) and weekend (non-working regime, February 15-18).   The averaged air flow rate was 1.2±0.1 l/min and the aerosol particles were captured on a 2 μm thick Mylar film

The sample analysis was carried out with the microbeam analysis system at Tohoku University.   Technical details of the system were presented in previous papers[5,8,9].   For multimodal analysis, two X-ray detectors for PIXE analysis and two charged particle detectors for RBS and off-axis STIM were mounted to observe the sample simultaneously. A simultaneous set of PIXE / RBS / off-axis STIM spectra was accumulated upon irradiation of the sample by a 3 MeV proton beam with the 1×1 μm^2 beam spot size and 50-100 pA beam current.   The beam scanning area was set to between 40×40 to 100×100 μm^2 for individual particle analysis, Elemental maps were obtained by GeoPIXEII software[10]. After generating the elemental maps, individual particles were selected from these maps and corresponding PIXE, RBS, and off-axis STIM spectra were extracted and analyzed. Concentrations of carbon, oxygen, and hydrogen were derived from peak yields of extracted RBS and off-axis STIM spectra. Samples collected on stages 2 and 3 in non-working regime were analyzed.

Results

In the previous measurement, congregation of the particles, wider scanning area of 200×200 μm and low counting statistics restricted individual particle analysis[7].   By measuring the area where there are less particles accumulated and using a smaller scanning

area, the individual particles could be identified and the distribution of particle specific elements could be determined. In non-working regime, elemental concentrations of Zn, Ba, and Pb were more than ten times higher than in working regime, which was related the restoration of an old white painted wooden door[7]. In total, 49 particles were analyzed for the non-working regime. Carbon and oxygen are major components of these particles. Forty six particles out of 49 particles could be grouped in three main categories: Ba-rich, Pb-rich and Ca+Fe rich particles, as shown in Fig. 1. During particle collection by the cascade impactor, a time-resolved sampling using the 2D time step sampler was also carried out. The 2D time step sampler data displayed a strong correlation between Ba, Pb and Zn, meaning that these particles certainly have the same origin, and the corresponding analysis will be published in a separate paper. However, the individual particles show no correlation between these three elements. On the contrary, the sets of particles containing Ba and Pb are rather complimentary. Zinc is equally distributed between Ba-rich particles and Pb-rich particles. Figure 2 shows correlations among Ba, Zn and S. These elements are well correlated and it is estimated that the signal is produced by a mixture of $BaSO_4$ and ZnS. Amounts of S are more than those obtained from stoichiometric ratio, which is explained by the mixture. In fact, the combination of $BaSO_4$ and ZnS is used as white pigment for paints and is called lithopone. Apparently, these elements are related to restoration of the door.

Figure 3 shows the correlation among Pb, Cl and O. High concentrations in Cl and its correlation to lead indicate that lead is also present under the form of Patteson's white lead $PbCl_2$. No clear correlation between Pb and O can be drawn from the graph of Pb in function of O, as organic matter is present in significantly large quantities. Whereas Pb, Zn and Ba are usually found in smaller particles, the removal of the old paint layer also explains that these elements are contained in larger particles, as they were here generated by grinding, and not by nucleation.

Although the results are not shown due to the lack of space, Ca and P are well correlated for the particles that contain high weight percentages in phosphor. Atomic ratio of Ca:P is 1.7. While the high amount of organic matter makes it impossible to determine the compounds, the compound might be $Ca_3(PO_4)_2$.

Conclusions

In-doors aerosols collected in the machine workshop of Jožef Stefan Institute were reanalyzed by a microbeam analysis system a Tohoku University. By measuring smaller sample area with low particle density, the individual particles could be identified. In total, 49 particles were analyzed on sample collected during the non-working regime. Looking at the correlation pattern of different elements in individual particles, the source of aerosol particles could be identified. The detected Ba-Pb-Zn correlation is related to a removal of an old paint layer by grinding, contained under the chemical state of Patteson's white lead $PbCl_2$, and the mixture of $BaSO_4$ and ZnS, known as the pigment called Lithopone. We demonstrated the capability of individual particle analysis to identify the source of these particles. Thus individual particle analysis is applicable to find the aerosol source.

Acknowledgments

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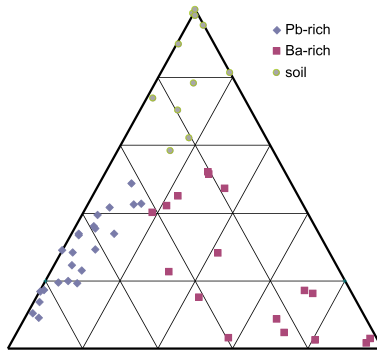
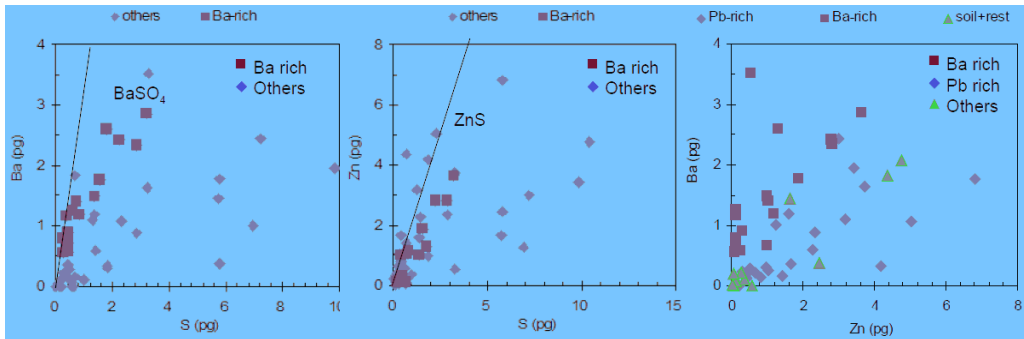Figure 1.　Ternary diagram of the three main groups.
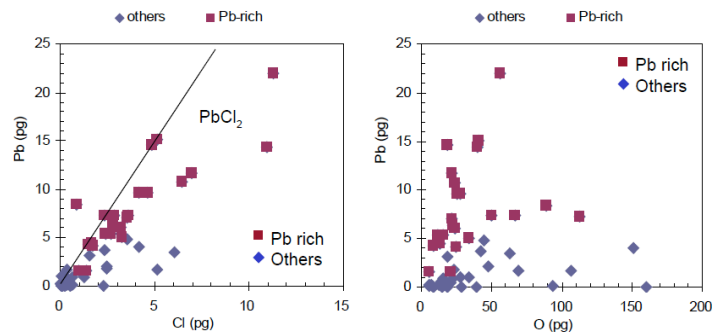

Figure 2.　Correlation among Ba, Zn and S concentrations in individual particles.

Figure 3.　Correlation among Pb, Cl and O.