Japan Geoscience Union Meeting 2014
(28 April - 02 May 2014 at Pacifico YOKOHAMA, Kanagawa, Japan)

AAS22-P02 会場:3 階ポスター会場 時間:5 月 1 日 18:15-19:30

エアロゾル中のイオウやカルシウムの化学種同定：地球冷却効果との関連や粒子表面での反応過程
Speciation of S and Ca species in aerosols with its relations to global cooling effects and processes of chemical reacti

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Speciation of particles in aerosols is necessary to interpret what effects each species in the aerosols can have on environment. For example, global cooling effect by aerosols influences earth's climatic change (IPCC, 2007). In particular, sulfate aerosols are known to cool the earth by forming cloud condensation nuclei (CCN) because of their high hygroscopicity, which induces indirect cooling effect. Because the hygroscopicity differs depending on the species, sulfate speciation in aerosols is important for the determination of the magnitude of the indirect cooling effect.

In this study, major ion concentrations in aerosol samples were measured by ion-chromatograpy. In addition, chemical species of calcium and sulfur in the each aerosol sample were determined using X-ray absorption near-edge structure (XANES) measured at BL-9A in Photon Factory, KEK. The speciation analyses can have some implications on the influence on the environment and the processes of chemical reaction of aerosols collected during several periods, such as (a) dust (Kosa) period (March 4-9, 2013), (b) the period with high PM2.5 concentration (Jan. 31-Feb. 1, 2013), and (c) the periods before and after (a) and (b).

Major ion concentration data showed that Ca^{2+} , which is originated from soil, and NO_3^- and SO_4^{2-}, which were from human activities, increased in the period (a) compared with those in the periods before and after the period (a). On the other hand, SO_4^{2-} and NH_4^+, which were emitted from human activities, increased in the period (b). In the period (a), it is considered that species originated from acids such as sulfate and nitrate which were incorporated into the particles increased in the samples whose aerodynamic diameter is over 1.0 um, because they have reacted with $CaCO_3$ which was increased by Kosa event. In addition, from the fitting of XANES spectra, it was found that gypsum with low hygroscopicity were the main sulfur species in the period (a), whereas NH_4HSO_4, $(NH_4)_2SO_4$, and hydrated sulfate with high hygroscopicity were main sulfur species in the period (b). Therefore, it is considered that when the concentration of PM2.5 increases, the indirect cooling effect can be large due to the large fraction of NH_4HSO_4, $(NH_4)_2SO_4$, and hydrated sulfat. On the other hand, the indirect cooling effect by sulfate aerosols can be smaller during the dust period due to the formation of non-hygroscopic gypsum by high amount of calcite in the atmosphere.

Using the results of calcium and sulfur speciation both in the bulk and at the surface by fluorescence and conversion-electron yield detection, respectively, in the XANES analyses, we can discuss how chemical reactions occur at the surface of aerosol particles in each period. The abundance ratios of gypsum, $CaCO_3$, and $Ca(NO_3)_2$ were different at the surface and the bulk. As a result, it was concluded that calcium species changes from gypsum, $Ca(NO_3)_2$, to $CaCO_3$ from the surface to the core of the calcite particle. This results showed that (i) sulfuric acid from the atmosphere forms insoluble gypsum at the surface of calcite, (ii) $Ca(NO_3)_2$, formed as a result of the reaction of nitric acid and calcite, exists in the middle part, and (iii) unreacted $CaCO_3$ remains in the core of the particle.

キーワード: エアロゾル, XANES, 硫酸塩化学種, 冷却効果
Keywords: aerosol, XANES, sulfate, grobal cooling effect