論文の内容の要旨

Studies on Abundance and Emission Strengths of Anthropogenic Iron Oxide Aerosols Using Laser-Induced Incandescence Technique

（レーザー誘起白熱法を用いた人為起源酸化鉄エアロゾルの大気中の動態研究及び排出強度推定）

氏名　吉田　淳

Iron-containing aerosols in the atmosphere are considered to play important roles in the Earth’s climate. They absorb solar radiation as black carbon (BC) and brown carbon (BrC) do, resulting in a positive radiative forcing. Irons in aerosols are also an essential nutrient for phytoplankton and their deposition to the surface ocean is considered to affect the carbon cycle through the absorption of carbon dioxide into the ocean. Thus, iron aerosols affect the climate on both short time scales as light-absorbers and long time scales as nutrient sources for the ocean ecosystem.

It has been considered that mineral dust emitted from the arid area is a main component of atmospheric iron aerosols. Recently, the existence of anthropogenic iron aerosols such as fuel combustion has been reported. However, the abundance and behaviors of anthropogenic iron aerosols in the atmosphere, as well as their radiation effects have not been examined due to a lack of reliable observations. In most of the previous studies, iron aerosols were measured by the collection of particles on filers and subsequently analyzed in laboratories. Although these methods provide useful information on the chemical component and shape of iron aerosols, they cannot provide concentrations and size distributions with a sufficiently high time resolution that is required for statistical analyses. As a consequence, emission inventories of anthropogenic aerosols, that are required to evaluate their impacts on the climate, are compiled only by accumulating from individual emission sources (bottom-up method) and they are considered to have significant uncertainties. To improve the emission inventories, reliable and statistically enough observational data that can constrain the emission strength (top-down method) are needed.

In this study, I developed a new technique to measure light-absorbing iron oxide particles (FeO_x) including magnetite and hematite using a laser-induced incandescent (LII) technique. In this technique, a mass of individual FeO_x particles (170-2100 nm) can be quantified by measuring the incandescence light emitted from the particle in the laser beam. Furthermore, the scattering cross section and the position where the incandescence starts in the laser beam (incandescence onset position) are measured to evaluate the mixing state of FeO_x aerosols with other aerosol compounds (volatile or non-volatile) and to determine FeO_x chemical components (e.g., magnetite and hematite). From these information one can classify the detected FeO_x particles into anthropogenic (combustion) and natural origins (mineral dust). Thus, the developed LII technique has made it possible for the first time to estimate a mass (size), size distribution, and mixing state with a high temporal resolution (1–60 mins) under typical atmospheric conditions.

Using the developed LII technique, in situ measurements of FeO_x aerosols were made at Cape Hedo, Okinawa Island in April 2017. Air likely affected by sources over the Asian continent was frequently sampled due to prevailing northwesterlies. Based on the single-particles quantities measured by the LII, most of the observed FeO_x aerosols were found to be anthropogenic; a number fraction of mineral dust particles (known as Kosa) was below 10% during this experiment. Temporal variation of FeO_x aerosols was well captured and the statistically significant data enabled to obtain clear positive correlations between the mass concentrations of FeO_x and BC (R^2 = 0.717) and of FeO_x and CO (R^2 = 0.718) in air originating from China. These correlations indicate that the emission sources of FeO_x aerosols are spatially similar to those of BC and CO in China.

In this study, I used the observed slope of the correlation between FeO_x and BC (CO) as a scaling factor for global BC (CO) emission data in literature in order to estimate global anthropogenic iron emissions. As a result, the global emission strength of anthropogenic iron aerosols was estimated to be 1.4-3.4 FeTg/yr. This is the first estimate using the top-down method. Uncertainties in these estimates are discussed in this thesis.

In order to understand the abundance of FeO_x aerosols in wide-area, I also acquired and analyzed FeO_x data in East Asian (area influenced by air from the Asian continental), urban city in Japan, and the Arctic regions. Most of the FeO_x aerosols observed in these regions were anthropogenic based on the single-particles quantities measured by the LII. Among all observations, the shapes of FeO_x number size distributions are similar. Mass concentrations of FeO_x aerosols were 20-50% of those of BC. Furthermore, in all observations, it was found that the FeO_x aerosol concentration showed a positive correlation with BC and CO. These facts suggest that the source of anthropogenic FeO_x aerosols in the Northern Hemisphere likely spreads over a wide area as well as that of BC and CO.

The new method that I have developed was also applied to estimate FeO_x emission sources in urban areas. As a result, the vehicle is suggested to be an important source of anthropogenic FeO_x aerosols rather than steel and power plants.

The novel dataset of FeO_x aerosols that I obtained in East Asia was used to constrain global model simulation for evaluating the radiative forcing of anthropogenic FeOx aerosols as well as their possible impacts on the iron supply to the ocean. In this simulation, FeO_x/BC mass ratio and particle size distribution obtained in the dataset were used to constrain the anthropogenic iron emissions. As a result, an atmospheric burden of anthropogenic iron aerosols was estimated to be approximately 7.7 times that estimated in the previous study. The simulation showed that a level of the radiative forcing of anthropogenic FeO_x aerosols can be similar to that of BrC. In addition, the deposition flux of anthropogenic iron to the surface southern ocean was estimated to be 6.5 times of that estimated in the previous study.

To conclude, a series of reliable measurements in this study indicate that most of the light-absorbing FeO_x aerosols are anthropogenic origin and they likely play more important roles for the processes related to the climate than previously thought. Further improvement of FeO_x measurement techniques are important to reduce uncertainties in quantification of FeO_x concentration and size distribution. In addition, further observations including ground and aircraft measurements especially in southern hemisphere are also important to understand the abundance, transport and removal of iron aerosols and their impact on climate.