A study of atmospheric ammonia by means of modeling analysis in the Kanto region of Japan

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1. Introduction

NH_3 plays an important role in the neutralization of acid and the formation of file aerosols. NH_4^+ particles have been important in considering transboundary air pollution because they have a relatively small dry deposition velocity. Moreover NH_4^+ contributes to the acidification of soil because NH_4^+ is converted to nitrate by the action of microorganisms and releases two H^+ in soil. Therefore, NH_3 and NH_4^+ particles have been important to the acidification of the environment, and studies of atmospheric ammonia have progressed well in the U.S. and Europe as well as the studies of SOx and NOx.

This study focuses on the atmospheric ammonia in the Kanto region of Japan. The Kanto region is the most densely populated area in Japan (40 million people), and consists of seven prefectures including the Tokyo metropolis. Since the area also has many livestock, the density of NH_3 emission derived from urban human activities and agriculture is the highest in Japan. Therefore, we have conducted observations of the concentrations for NH_3 and NH_4^+ particles and evaluated the budget analysis (Sakurai et al., 2002; Sakurai and Fujita, 2002). This study evaluates the behavior and budget for NH_3 and NH_4^+ particles by means of modeling analysis. The concentration and the deposition of NH_3 and NH_4^+ particles were simulated using Models-3/CMAQ (the Community Multi-scale Air Quality model, Environmental Protection Agency) with meteorological fields of MM5 (the fifth-generation Mesoscale Model of non-hydrostatic version, Pennsylvania State University / National Center for Atmospheric Research).

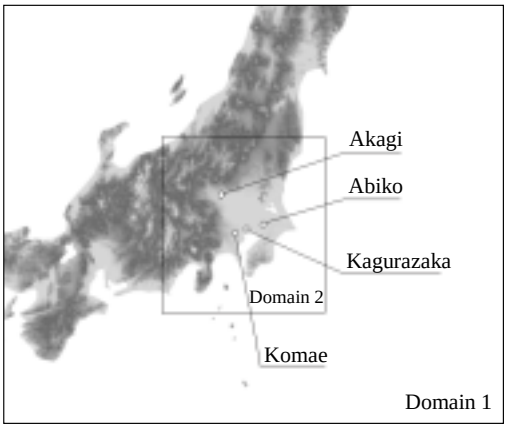

Figure 1. Modeling domain system, consisting of nested 15 km and 5 km grids.

2. Methodology

The observations of atmospheric ammonia were conducted at four stations (Kagurazaka, Akagi, Komae, and Abiko) in the Kanto region from January to December 2000, and at two stations (Kagurazaka and Akagi) from July to August 2002. Akagi is located in a rural area, and agricultural sources of NH_3 are relatively large. Kagurazaka is located in the Tokyo urban area, and anthropogenic sources of NH_3 are relatively large.

Figure 1 shows the modeling domain. The meteorological fields derived from MM5 had a nested system of 15 km (Domain-1) and 5 km (Domain-2) grids centered over the Kanto region. The domain sizes of Domain-1 and Domain-2 were 795×1125 km and 335×290 km, respectively. The vertical domain extended up to 100 hPa and was divided into 16 layers. The initial and boundary conditions of meteorological data were derived from MANAL (Meso-regional objective analysis

data) and RANAL (Regional objective analysis data) developed by JMA (Japan Meteorological Agency).

Models-3/CMAQ is a modeling system that calculates transport, deposition, gas-phase chemistry, aerosol dynamics and chemistry, and cloud processes, based on initial and boundary conditions, meteorological fields, and emission data. In the modeling system, the calculation was run by means of the chemical mechanism with CB4-AE2-AQ (Carbon Bond Mechanism version 4 with aerosols and aqueous chemistry), advection with piece-wise parabolic method, and diffusion with K-theory. The vertical domain extended up to 100 hPa and was divided into 8 layers

The input NH_3 emission was estimated separately from agricultural source, anthropogenic source, and natural source. Since detailed information on the NH_3 emission remains limited in Japan, the emission was estimated based on the emission factors developed in Europe and North America. The agricultural source consisted of livestock and fertilizer application. The anthropogenic source consisted of production plants, denitrification by combustion, vehicles, wastewater treatment, latrines, human perspiration and respiration, and pets. The natural source was the volatilization from soil surface. The emission amount of NH_3 in the Kanto region was estimated for 445 ton day^{-1} with an area of about 40,000 km^2; the amount of agricultural source was 60%, anthropogenic source was 37%, and natural source was 3% of the total.

3. Results and discussions

Figure 2 and Figure 3 show the seasonal variations for observed and calculated concentrations of NH_3 and NH_4^+ particles at four stations in 2000. Although the

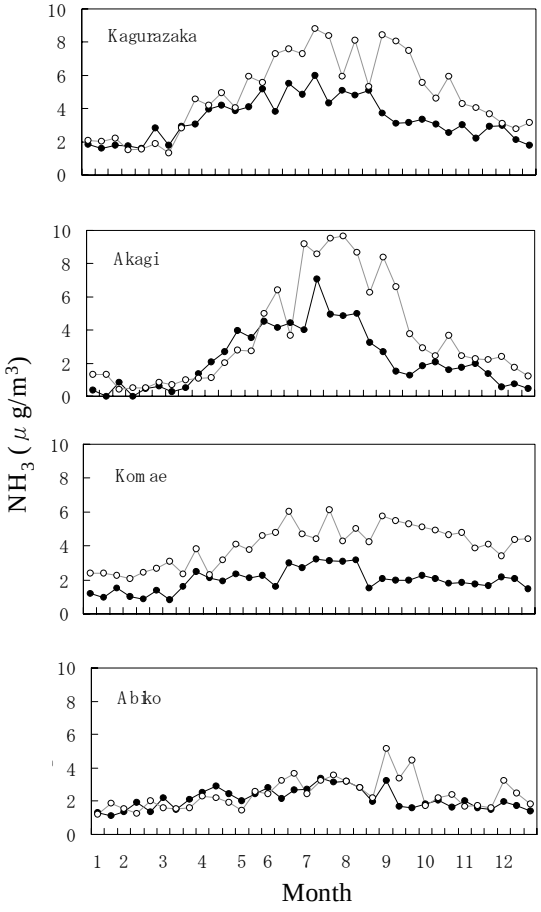

Figure 2. Seasonal variations for observed and calculated concentrations of NH_3 at four stations in 2000. (○:calculation, ●:observation)

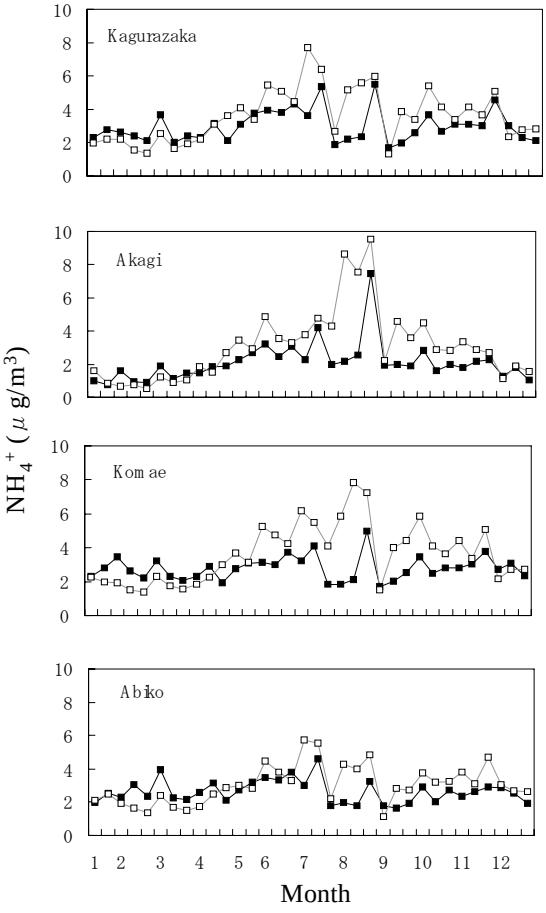

Figure 3. Seasonal variations for observed and calculated concentrations of NH_4^+ particles at four stations in 2000. (□:calculation, ■:observation)

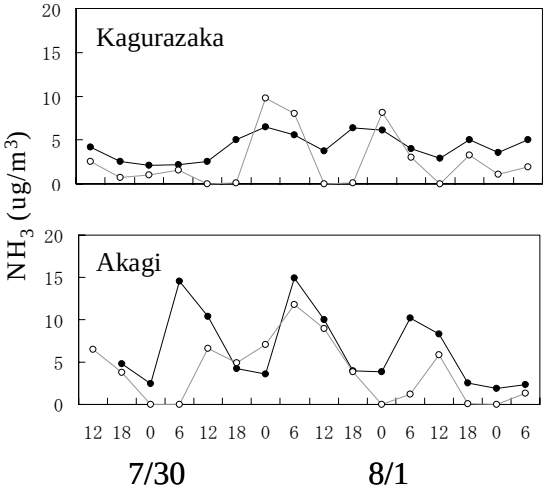

Figure 4. Time variations for observed and calculated concentrations of NH_3 at Kagurazaka and Akagi in summer 2002. The numbers on the horizontal axis indicate the start time of each sampling. (○:calculation, ●:observation)

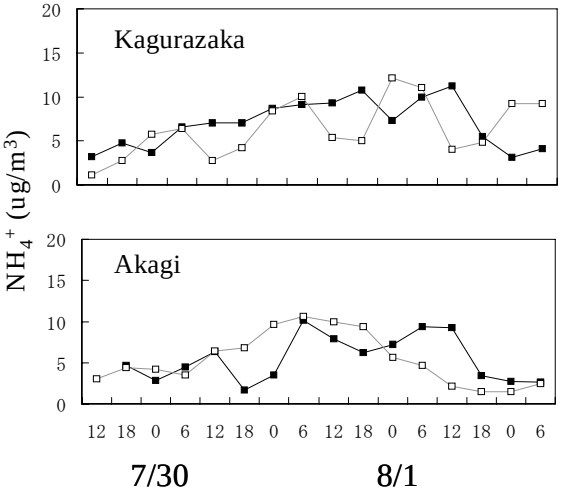

Figure 5. Time variations for observed and calculated concentrations of NH_4^+ particles at Kagurazaka and Akagi in summer 2002. The numbers on the horizontal axis indicate the start time of each sampling. (□:calculation, ■:observation)

calculated concentrations of NH_3 and NH_4^+ particles during the summer season were somewhat higher than the observed concentrations at every station, the calculation results showed good agreement with the seasonal variations of observed concentrations. Most fertilizer is applied to fields in the summer season. Moreover, the emission strength of NH_3 based on the volatilization generally increases as the temperature rises. Thus, it was suggested that the high concentrations of NH_3 and NH_4^+ particles in the summer season reflected the increase of emission strength.

Figure 4 and Figure 5 show the time variations for observed and calculated concentrations of NH_3 and NH_4^+ particles at Akagi and Kagurazaka in summer 2002. The calculation results showed good agreement with the time variations of observed concentrations. The high concentrations of NH_3 were appeared in the daytime at Akagi, and in the nighttime at Kagurazaka. Since the emission strength of ammonia based on the volatilization generally increases as the temperature rises, the peaks in the daytime at Akagi reflected the influence of the emission sources that exist around the sampling station. On the other hand, it was confirmed that the peaks in the nighttime at Kagurazaka were caused by transport of NH_3 from the northern Kanto region. In addition, it was suggested that the high concentration of NH_3 in the nighttime at Kagurazaka occurred due to the reduced vertical dilution and high emission rate from the anthropogenic source in the nighttime (Sakurai et al., 2003).

Reference

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