Oxidation and removal of nitrogen oxides in Tokyo

Masato Fukuda[1]; Yutaka Kondo[2]; Yuzo Miyazaki[3]; Yu Morino[4]; Nobuyuki Takegawa[5]; Takuma Miyakawa[1]; Yugo Kanaya[6]; Hiroshi Tanimoto[7]

[1] Earth and Planetary Sci., Univ. of Tokyo; [2] RCAST, Univ. of Tokyo; [3] Earth and Planetary Physics, Univ. of Tokyo; [4] RCAST, Univ. Tokyo; [5] RCAST, Univ of Tokyo; [6] FRCGC/JAMSTEC; [7] NIES/AED

http://noysun1.atmos.rcast.u-tokyo.ac.jp/

Reactive nitrogen (NOx = NO + NO2) play an important role in the formation and destruction of tropospheric ozone (O3). The primary loss of NOx occurs mainly through oxidation of NO2 followed by formation of HNO3. Peroxyacetyl nitrate (PAN) is also one of the major oxidized products of NOx. The final loss of NOy occurs through wet and dry deposition of HNO3, where NOy is defined as the sum of the gaseous nitrogen oxides and fine particulate nitrate (NOy = NOx + NO3 + 2(N2O5) + HNO3 + Peroxyacyl nitrates (PANs) + Organic nitrates + Particulate nitrate (NO3-)). Removal of HNO3 implies not only loss of NOy but also acid deposition onto surface, therefore studying these processes is important. Some studies have been made to understand oxidation of NOx and removal of NOy in urban and sub-urban air through field observations at various locations. However, observational studies on the behaviors of nitrogen oxides in urban areas in Japan are very limited.

Intensive measurements of nitrogen oxides, together with other trace gases were made for the first time near the urban center of Tokyo during the periods of July-August, 2003-2004, and January-February, 2004. The controlling factors of the NOy partitioning were investigated using observational data. The NOx/NOy ratios were found to decrease during the daytime due to increasing photochemical activity. However, NOx was the dominant species of NOy throughout the observational periods with the NOx/NOy ratios of 0.63-0.87 in summer and 0.83-0.95 in winter, because measurements of these species were made near emission sources of NOy. The partitioning of oxidized products of NOx (NOz) was different between summer and winter. The dominant species of NOz was HNO3 (HNO3/NOz ˜0.35) in summer and NO3- (NO3-/NOz ˜0.42) in winter. This seasonal change of the dominant species of NOz in winter from summer is due to the shit of the HNO3-NO3- partitioning to NO3- at low temperatures.

In order to quantify loss of NOy, the loss fractions of NOy were estimated using CO as a tracer of anthropogenic emissions. As a result, the estimated loss fraction of NOy was largest of 24% during the daytime in summer, while it was smallest of 4% in winter. It is suggested that the seasonal variations of loss fractions of NOy in winter from summer were caused by both decrease of production of HNO3 with reduction of OH concentrations and change of the HNO3-NO3- partitioning.