

## Trends of air pollution over the East China Sea analyzed with 20 years aerial observation data

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[Introduction] We have continued aerial observations over the seas between Asian continent and Japan for twenty years in order to analyze the transport and transformation of long-range transported atmospheric pollutants. Covered areas are the East China Sea, the Sea of Japan, and the Yellow Sea. During this period the economic growth in East Asia, particularly in China, was remarkable and as a result emission of atmospheric pollutants increased tremendously. In this report we will show the results of our analyses on the long term trend of gaseous pollutants such as SO<sub>2</sub> and ozone as well as ionic components of aerosols.

[Used data] The data used for the analyses were 11 data sets, i.e., those obtained in PEACAMPOT program in October, 1991, November, 1992, March and December, 1994, January and December, 1997, February, 1999, and March 2001 as well as those in LEXTRA program in March-April, 2008, and those in ASEPH project in October, 2009 and December, 2010.

Aircrafts employed were Cessna 404, and Fairchild Swearingen chartered from Showa Aviation Company, and Beachcraft Kingair 200T chartered from Diamond Air Service. The target of the observations was lower tropospheric (below 3000 m) atmosphere.

Items of observations common for 20 years experiments were gaseous species such as ozone, SO<sub>2</sub>, and NO<sub>x</sub> (NO<sub>y</sub>) as well as ionic species in aerosols collected with a high-volume tape sampler. Sulfate, nitrate, ammonium, and calcium were the main ionic species analyzed.

[Results and Discussion] Anthropogenic species in the gas phase (SO<sub>2</sub> and ozone) and aerosol phase (SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup>, and NH<sub>4</sub><sup>+</sup>) were analyzed in this presentation.

Sulfate and ammonium showed a quite similar trend. That means sulfate and ammonium existed in the form of (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> and/or NH<sub>4</sub>HSO<sub>4</sub>. It seems that sulfate is decreasing from 2001. Unfortunately, since the data between 2002 and 2007 are lacking (we carried out aerial observations over the main land China in that period), it is not obvious. However, it is reported that the emission of SO<sub>2</sub> in China is decreasing from 2006 (Lu et al., 2010). Our results seem in accordance with the report, although decreasing trend of SO<sub>2</sub> was not very clear in this work.

In contrast, was low before December, 1997 (maximum 1.5 ug/m<sup>3</sup>), but it is increasing after 1999 having maximum concentrations exceeding 7 ug/m<sup>3</sup>. It seems to show an increasing trend. Emission of NO<sub>x</sub>, which is a precursor of NO<sub>3</sub><sup>-</sup>, is increasing in China (Ohara et al., 2007), and that can cause increase of NO<sub>3</sub><sup>-</sup> over the East China Sea. More clearly, the ratio of NO<sub>3</sub><sup>-</sup>/SO<sub>4</sub><sup>2-</sup> increases reflecting the trends of sulfate and nitrate.

Ozone was analyzed using histograms for former period (1991-1999) and latter period (2001-2010). In 1990s ozone in the range of 40<O<sub>3</sub> conc.<45ppb appeared most frequently, whereas ozone in the range of 65<O<sub>3</sub> conc.<70ppb appeared most frequently in 2000s. This feature was more remarkable in the lower troposphere below 1500 m. Ozone in the boundary layer is increasing because of anthropogenic emission of NO<sub>x</sub> in China.

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Ohara, T., et al., (2007). *Atmospheric Chemistry and Physics Discussions*, 7(3), 6843-6902. doi:10.5194/acpd-7-6843-2007.

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