

Chapter 2

Aerosols

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Abstract Aerosols in East Asia, in particular the chemical constituents of PM_{2.5} (particulate matter 2.5 μm or less in diameter), are described in this section. Aerosols are colloidal systems in which small ($<100\ \mu\text{m}$ in diameter) solid or liquid particulate matter is suspended in air. These small particles are themselves also called aerosols. PM_{2.5} and *kosa* (Asian yellow sand dust) are important aerosol constituents, and their chemical components; namely, elemental and organic carbon, polycyclic aromatic hydrocarbons (PAHs), metallic elements, and inorganic ionic species, are discussed. The long-range transport of the aerosol components is the major topic in this section. The chemical transformation of aerosol components was evaluated and source apportionment was performed on the basis of model simulations and statistical analyses.

Keywords PM_{2.5} • *Kosa* • Elemental carbon (EC) • Organic carbon (OC) • Polycyclic aromatic hydrocarbon (PAH) • Metallic elements • Ionic species

2.1 Introduction

Because of rapid economic growth in Asia, particularly in East Asia, emissions of atmospheric pollutants are growing quite rapidly. Besides gaseous air pollutants such as O₃, nitrogen oxides (NO_x), SO₂, and volatile organic compounds (VOCs), many kinds of particulate air pollutants, consisting of sulfates, nitrates, and carbonaceous particles, as well as soil dust particles such as *kosa* (Asian yellow sand dust), are also emitted directly or produced secondarily by precursor gases during long-range transport. Such aerosols (see aerosol definition below) are transported downwind and they even cross the Pacific Ocean to North America, where they contribute to pollution. Clarification of the chemical change processes that occur during aerosol transport, and determination of the impacts of aerosols after

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transport on animals and plants, as well as on human health, are important environmental issues.

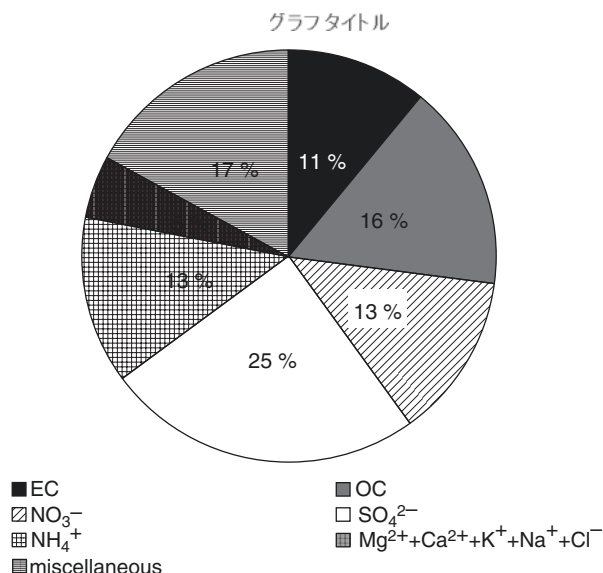
In January 2013, very high concentrations of PM_{2.5} (particulate matter with a diameter of less than 2.5 μm); described in detail in a later section) were observed in China, particularly in Beijing, where the mass concentrations of fine particles reported as PM_{2.5} were up to 1000 $\mu\text{g m}^{-3}$. The reported concentrations were far exceeded the daily air quality standards of not only Japan and USA (35 $\mu\text{g m}^{-3}$) but also even China (75 $\mu\text{g m}^{-3}$). The high levels of PM_{2.5} then became a topic of public concern in Japan and many people began to pay attention to the PM_{2.5} forecasts on television. The term “PM_{2.5}” was not so familiar at that time, yet people were concerned that something very dangerous was being transported from East Asia.

The International Agency for Research on Cancer (IARC, an agency of the World Health Organization) Working Group has unanimously classified particulate matter and outdoor air pollution as carcinogenic to humans (IARC Group 1), based on sufficient evidence of carcinogenicity in humans and experimental animals and strong mechanistic evidence (Loomisa et al. 2013). The IARC describes an agent in this group as follows: “The agent is carcinogenic to humans. This category is used when there is sufficient evidence of carcinogenicity in humans. Exceptionally, an agent may be placed in this category when evidence of carcinogenicity in humans is less than sufficient but there is sufficient evidence of carcinogenicity in experimental animals and strong evidence in exposed humans that the agent acts through a relevant mechanism of carcinogenicity.” At present, 117 agents are classified in this group (IARC 2015).

2.2 Aerosols and PM_{2.5}

Aerosols are colloidal systems in which small (<100 μm in diameter) solid or liquid particulate matter is suspended in air. These small particles are themselves also called aerosols. Atmospheric pollutants transported from Asia are not limited to fine particles classified as PM_{2.5}, but, as previously mentioned, PM_{2.5} aerosols have recently attracted much attention. Aerosols, including PM_{2.5}, are not a single compound, but are composed of many different chemical species. Aerosols include sulfuric acid mist, photochemical smog, asbestos particles, and diesel exhaust soot. Figure 2.1 shows the average chemical composition of PM_{2.5} aerosols in urban areas of Japan (Ministry of the Environment of Japan 2009). These aerosols affect not only the local environment but also contribute to global environmental issues, such as global warming, stratospheric ozone depletion, and acid rain. Sources of aerosols are not only anthropogenic but also natural. Major anthropogenic sources are photochemical smog, automobiles, and certain industries, while major natural sources are plants, soils, and seawater. Plant pollen (the pollen of Japanese cedar trees is a notorious allergen in Japan) and *kosa* transported from China are examples of natural-origin aerosols. Aerosol particles span a range of sizes from very fine (less than 1 μm in diameter) to coarse (up to 100 μm). In addition to the variety in

Fig. 2.1 Chemical composition of aerosols in urban areas of Japan (average of 2004–2008). *EC*, elemental carbon, *OC* organic carbon



size, they have a wide variety of chemical characteristics (different kinds of chemical compounds) and physical characteristics (liquid and solid; and a variety of forms, including both spherical and other symmetrical shapes [e.g., snow] and rugged, angular shapes). As we have been obtaining more information from recent progress in the chemical and physical analyses of aerosols, it has become clear that atmospheric aerosols play an important role in the environment.

Aerosols can be classified into four classes based on their origin. Particles that are particulate in origin are classified as primary and those that were originally gases but have been converted to particulate species by some chemical reactions in the air following emission are classified as secondary. Both primary and secondary particles have natural and anthropogenic sources. Some examples of each class are listed below.

- (i) Primary natural: Soil particles, such as *kosa*; sea salt; pollen; mold; and mushroom spores
- (ii) Primary anthropogenic: Soot and fly ash (a coal combustion product), dust from studded tires, and polycyclic aromatic hydrocarbons (PAHs) emitted during fuel combustion
- (iii) Secondary natural: Products of the atmospheric reaction of ozone and OH radicals with natural hydrocarbons emitted by plants (isoprene and terpenes) and stratospheric aerosols that make up the so-called Junge layer (fine sulfuric acid particles produced in the stratosphere by the oxidation of SO_2 that is emitted by volcanic activity)
- (iv) Secondary anthropogenic: H_2SO_4 (formed by the oxidation of SO_2 emitted from, for example, industrial sources, i.e., not from volcanoes) in air; ammonium salts (formed by the neutralization reactions of H_2SO_4 and HNO_3 produced by the oxidation of NO_x); and organic particles created by the

reaction of ozone and OH radicals in air with anthropogenic hydrocarbons, such as cyclic olefins and aromatic hydrocarbons

International Organization for Standardization (ISO) PM_{2.5} particles are defined as those that can pass through a size-selective inlet with a 50 % efficiency cut-off at 2.5 μm aerodynamic diameter (ISO 2012). PM_{2.5} is not a new species, but its levels have recently increased and it is believed to pose a great health risk. Because of their small size, such fine particles can lodge deeply in the lungs.

The term PM_{2.5} refers only to the size of the particles, not to their chemical constituents, which can include tobacco smoke and benzo[a]pyrene, a well known carcinogenic PAH. Therefore, we need to pay attention not only to the mass concentration of PM_{2.5} particles but also to their chemical composition. The main components of ordinary long-range-transported PM_{2.5} in East Asia are sulfate and nitrate salts, as well as organic compounds.

Why is the PM_{2.5} problem an important environmental issue? In Japan an environmental standard for particles smaller than 10 μm (SPM; suspended particulate matter) was established in 1973, and monitoring and regulation have been performed according to this standard. However, particles smaller than 2.5 μm can cause more serious damage to human health because they can penetrate deeply into the lungs and their concentrations show high correlation with human mortality (Dockery et al. 1993). In the United States, National Ambient Air Quality Standards for PM_{2.5} were established in 1997 and have been revised twice (2006 and 2013). The present standards are 35 $\mu\text{g m}^{-3}$ for the daily average and 12 $\mu\text{g m}^{-3}$ for the annual average. The World Health Organization (WHO 2005) suggested stricter limits for PM_{2.5} (25 and 10 $\mu\text{g m}^{-3}$ for daily and annual averages, respectively). A Japanese Air Quality Standard for PM_{2.5} was established in 2009 (35 and 15 $\mu\text{g m}^{-3}$ for daily and annual averages, respectively).

Another reason to regulate fine particles is that they consist mostly of anthropogenic species, whereas coarse particles are mainly of natural origin (Fig. 2.2). We cannot regulate the emissions of natural-origin aerosols, but we can and should

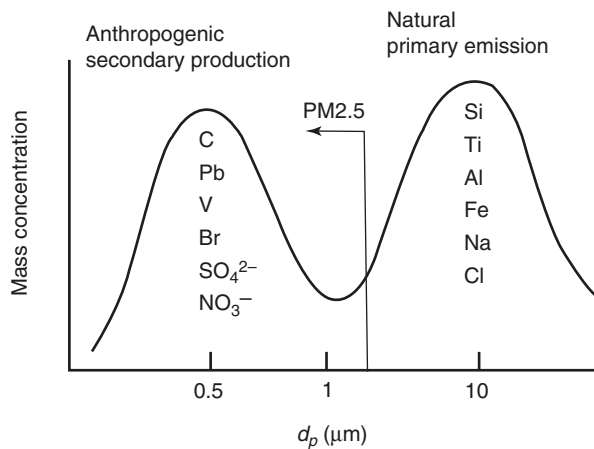


Fig. 2.2 General size distribution of aerosols and their main components

regulate anthropogenic emissions of aerosols and their precursors. We can expect the regulation of fine particles to have an effect. Moreover, the mass of a particle is proportional to the third power of its diameter, so one particle of 10- μm diameter corresponds to 1,000 particles of 1- μm diameter. If we regulate only aerosols containing coarse particles, it is difficult to control the concentration of fine particles.

2.3 *Kosa* and Chemical Species Transported with *kosa*

When very fine sand particles are blown up by strong winds in the Taklimakan and Gobi Deserts in inland China, the westerly wind transports them long distances. Figure 2.3 shows such a sandstorm, observed on 7 April 2001 (NASA 2008), that was caused by a very strong cyclone. In China, as well as in Korea, such a strong dust storm is regarded as a natural disaster.

Such strong dust storms can transport fine sand particles toward the Pacific Ocean, even reaching North America (NASA 2005). When an air mass containing dust particles passes over the coastal cities and industrial areas of China, anthropogenic air pollutants are adsorbed onto the surfaces of the aerosols and chemical transformation of the adsorbed compounds can take place. For example, SO_2 can be oxidized to sulfuric acid on the surface of such an aerosol. Such pollutants transported from Asia to America cause recognized global environmental problems. In contrast, alkaline compounds, such as CaCO_3 contained in dust particles, can contribute to the neutralization of sulfuric and nitric acids. Thus, we can say that dust particles have pluses and minuses that can offset each other.

Clear characteristics of the ionic composition of aerosols transported with *kosa* were identified from aerial observations of atmospheric pollutants over the East China Sea carried out by the author and colleagues (Hatakeyama et al. 2004). A dust storm broke out on 21 March 2001 in the Loess Plateau area in China and moved

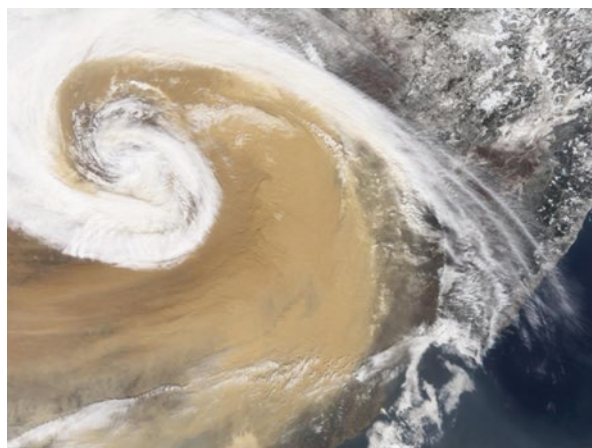


Fig. 2.3 A very strong sand storm over China observed from NASA satellite on 7 April 2001



Fig. 2.4 Flight track of aircraft, between points a and b, of aerial observations carried out on 20, 21, and 22 March 2001

eastward (Darmenova et al. 2005). Our aerial measurements of atmospheric pollutants were carried out on 20, 21, and 22 March 2001 over the East China Sea (Fig. 2.4), so we were able to detect *kosa* during our experiments. Figures 2.5, 2.6, and 2.7 show the variations in the aerosol mass concentrations and the concentrations of sulfate and calcium ions in the aerosols on 20, 21, and 22 March 2001, respectively.

The concentrations of particulate matter were generally low on 20 March (Fig. 2.5); PM₁₀ was less than 0.5 mg m^{-3} and PM_{2.5} was less than $12 \text{ } \mu\text{g m}^{-3}$. The sulfate and calcium ion concentrations were less than 10 and $1 \text{ } \mu\text{g m}^{-3}$, respectively. In contrast, following the arrival of *kosa*, a very high mass concentration peak of PM₁₀ ($\sim 2.5 \text{ mg m}^{-3}$) and a broad peak of PM_{2.5} ($60 \text{ } \mu\text{g m}^{-3}$) were observed on 21 March (Fig. 2.6). It is interesting to note that PM₁₀ showed a sharp peak south of Jeju Island, Korea, whereas PM_{2.5} showed a broad peak over the East China Sea between Jeju Island and Fukue Island, Japan, at low altitude ($\sim 500 \text{ m}$). The calcium ion concentration showed a maximum ($\sim 11.5 \text{ } \mu\text{g m}^{-3}$) close to the PM₁₀ peak (point a in (Fig. 2.6)). In contrast, the distribution of SO_4^{2-} ions was similar to the distribution of PM_{2.5} (point b in (Fig. 2.6)). On 22 March, the peak PM₁₀ concentration ($\sim 3.4 \text{ mg m}^{-3}$) was recorded off Kyushu Island, Japan (Fig. 2.7). The peak PM_{2.5} concentration ($\sim 60 \text{ } \mu\text{g m}^{-3}$) also reached Kyushu Island.

On 20 March, when the concentrations of both PM_{2.5} and PM₁₀ were low, the $\text{SO}_4^{2-}/\text{Ca}^{2+}$ ratios of total suspended particles (TSP) were very large at both high and low altitudes (average, 27.8 for altitudes higher than 2,500 m and 15.3 for those lower than 1,000 m). This means that the contribution of anthropogenic sulfate was high on 20 March. On this day, a cold front passed quickly over the Korean Peninsula

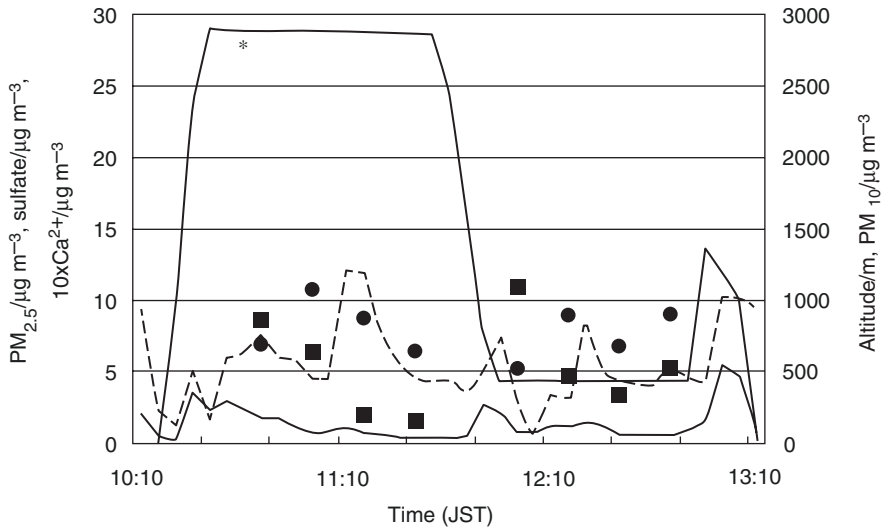


Fig. 2.5 Variations in aerosol mass concentrations (black solid line PM_{10} , dashed line $PM_{2.5}$; black solid line marked with * shows altitude) and concentrations of sulfate (circles) and calcium (squares) ions in the aerosols on 20 March 2001

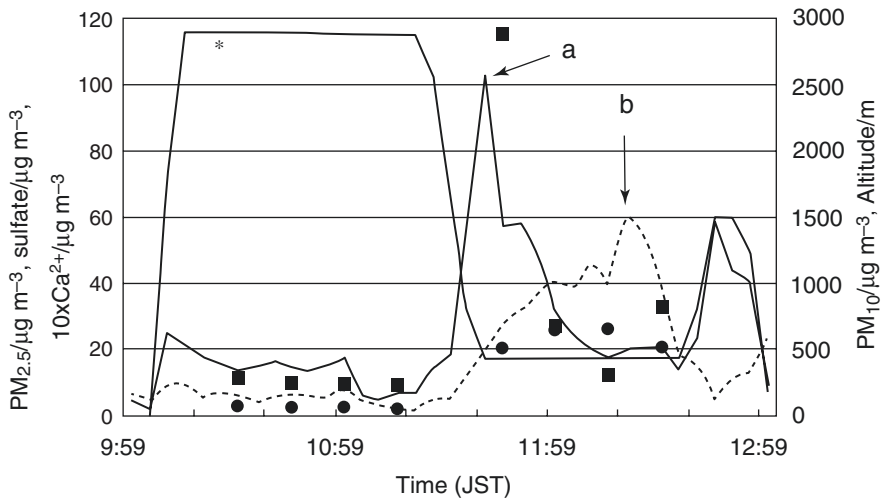


Fig. 2.6 Same items as those shown in Fig. 2.5, but on 21 March 2001

and the observation area, after which a high-pressure system moved into the observation area from central east China. The ratio of $\Sigma(\text{anionic species})/\Sigma(\text{cationic species})$ (taking nitrate, sulfate, and chloride as representative anionic species; and ammonium, calcium, and sodium as representative cationic species) was between 95 and 121%. $\Sigma(\text{anionic species})$ was always greater than $\Sigma(\text{cationic species})$.

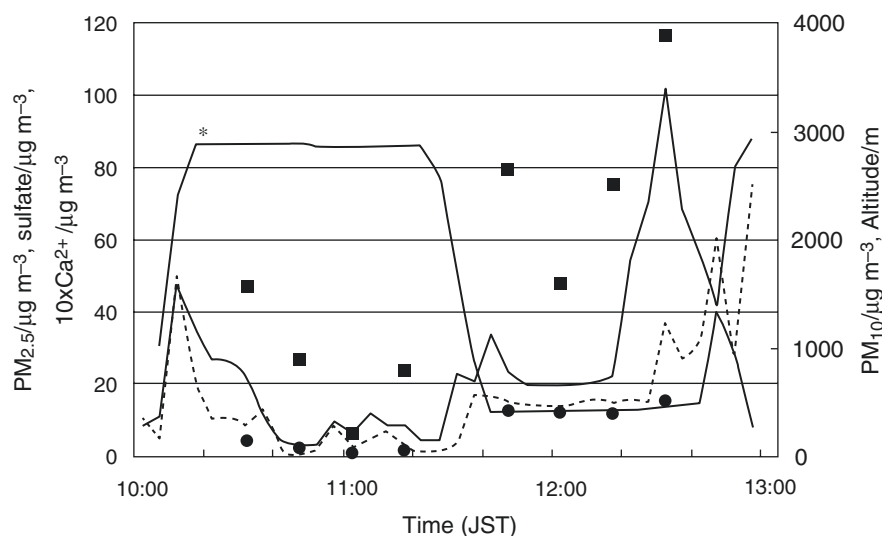


Fig. 2.7 Same items as those shown in Fig. 2.5, but on 22 March 2001

Therefore, the anthropogenic contribution to the chemical composition of the aerosols was very large on 20 March.

In contrast, on 22 March, when the concentration of PM10 was very high, the $\text{SO}_4^{2-}/\text{Ca}^{2+}$ ratios were very small (average, 1.0 for altitudes higher than 2,500 m and 1.8 for those lower than 1,000 m). It should be noted that the entire observation area was covered by the dust storm. The satellite image taken on 22 March 2001 (NASA 2001) shows the dust continuing to blow across the Korean Peninsula and Japan. This is the reason that the $\text{SO}_4^{2-}/\text{Ca}^{2+}$ ratios were so small. Moreover, $\Sigma(\text{cationic species})$ was greater than $\Sigma(\text{anionic species})$ on this day. This result also suggests that soil dust particles existed in excess on 22 March.

The mass concentration of coarse particles (PM10) on 21 March was high near Jeju Island (Fig. 2.6, point a), whereas that of fine particles (PM2.5) was high near Kyushu Island (point b). The concentration of Ca^{2+} showed a sharp peak following the distribution of PM10, whereas the concentration of sulfate showed a broad peak following the distribution of PM2.5. This means that the coarse particles were mainly *kosa* and that the fine particles contained a large amount of anthropogenic pollutants. Judging from the high concentration of PM2.5, we conclude that the lower atmosphere over the western Pacific was probably polluted most of the time in spring owing to continental outflow. On 21 March the $\text{SO}_4^{2-}/\text{Ca}^{2+}$ ratios were small at high altitude ($\sim 2,800$ m) and near Jeju Island at low altitude (average 2.1), whereas the ratios near the fine-particle peak depicted in Fig. 2.6 were large (average 12.3). The $\Sigma(\text{anionic species})/\Sigma(\text{cationic species})$ ratio was small (63–69 %) for the samples with small $\text{SO}_4^{2-}/\text{Ca}^{2+}$ ratios, whereas it was large (106–129 %) for the samples at lower altitude. As mentioned above, the air mass containing *kosa* arrived near Jeju Island on 21 March, when it had not yet reached the Kyushu area.

Nishikawa et al. (1991) reported that the $\text{SO}_4^{2-}/\text{Ca}^{2+}$ ratio in soil from the Gobi Desert was 0.004. They also reported that the $\text{SO}_4^{2-}/\text{Ca}^{2+}$ ratios of aerosols collected on Yakushima Island, Japan, during *kosa* and non-*kosa* periods were distinctly different. During the *kosa* period in April 1988, the ratios varied from 0.7 to 0.9, whereas in the corresponding non-*kosa* period the ratios ranged from 13.5 to 22.5. On the basis of these data, we can conclude that the air mass observed on 20 March was affected by anthropogenic pollutants. In contrast, the air mass observed on 22 March was affected by the Asian dust storm. Although the atmosphere near Jeju Island on 21 March was already affected by the Asian dust storm, the atmosphere near Fukue Island had not yet been affected by *kosa* particles.

Although sandstorms appear to be natural phenomena, increases in the frequency of sandstorms and subsequent deterioration of the environment arise from human activity. It has been reported that dust storms struck northwestern China on average once every 31 years from 300 CE to 1949, whereas since 1990 there has been one almost every year (Liu and Diamond 2005). This increased frequency is believed to be the result of overgrazing, deforestation, soil degradation, and desertification in Central Asia and across desert areas of China.

2.4 Elemental Carbon (EC) and Organic Carbon (OC)

Carbonaceous material is a major component of $\text{PM}_{2.5}$ in the megacities of China. Many studies have reported the chemical composition of $\text{PM}_{2.5}$ in Beijing; Chan and Yao (2008) reported that carbonaceous species (EC + OC) accounted for 27–42 % of the $\text{PM}_{2.5}$ mass in that city, and concentrations of EC + OC varied from 10.8 to 51.9 $\mu\text{g m}^{-3}$. In Shanghai, carbonaceous material accounted for 41.4 % of the $\text{PM}_{2.5}$ mass (Ye et al. 2003) and OC represented 73 % of the carbonaceous material. The annual average concentrations of EC and OC were 6.77 and 15.43 $\mu\text{g m}^{-3}$, respectively, at a monitoring site close to downtown Shanghai.

Elemental carbon (EC), also known as black carbon (BC), originates from the incomplete combustion of fuel during anthropogenic activities such as fossil fuel combustion, biomass burning, and biofuel burning for cooking and heating. EC is considered to be one of the most important aerosol components, because it is likely the second largest contributor to global warming. As one of the short-lived climate pollutants (SLCPs; which also include methane and tropospheric ozone), EC has become a target in the effort to lower global warming (Climate and Clean Air Coalition 2015). These SLCPs are also dangerous air pollutants, with various detrimental impacts on human health, agriculture, and ecosystems. It is easier to obtain a consensus for their reduction, compared with reaching a consensus for CO_2 reduction; demands for CO_2 reduction often meet opposition because this requires the reduction of fossil fuel energy use, which is widely considered to lead to the lowering of living standards.

The author's group has utilized the Cape Hedo Atmosphere and Aerosol Monitoring Station (CHAAMS) (Takami et al. 2007) at Cape Hedo, Okinawa,

Japan, to monitor the chemical composition of aerosols transported from East Asia. Among the many kinds of aerosols collected at Cape Hedo, EC and OC were the most important chemical components. Data obtained from March 2004 to March 2008 were used for the analyses of EC and OC, as well as other air pollutants (Shimada et al. 2011). Since EC and OC at Cape Hedo are mainly transported from China, the concentrations of EC and OC were high during the spring and winter, when most air masses come from East Asia (EC: $0.14\text{--}0.19\text{ }\mu\text{g C m}^{-3}$, OC: $0.62\text{--}0.82\text{ }\mu\text{g C m}^{-3}$). Concentrations of other air pollutants were also high at this time, e.g., PM_{2.5}: $14.8\text{--}19.9\text{ }\mu\text{g m}^{-3}$, O₃: 43–55 ppbv, and CO: 183–221 ppbv. In contrast, the concentrations of these pollutants were low during summer, when a Pacific high-pressure zone covered all of Okinawa (EC: $0.03\text{--}0.08\text{ }\mu\text{g C m}^{-3}$, OC: $0.28\text{--}0.44\text{ }\mu\text{g C m}^{-3}$). Concentrations of other pollutants were also low, e.g., PM_{2.5}: $9.85\text{--}14.8\text{ }\mu\text{g m}^{-3}$, O₃: 14–23 ppbv, CO (carbon monoxide): 68–93 ppbv. Thus, there is observational evidence that most air pollutants at Cape Hedo, Okinawa, come from East Asia, including China.

The source of carbonaceous aerosols was evaluated by determining the OC/EC ratios and the correlation between EC and OC. The ratio was low in spring and winter (OC/EC: 5.7–8.0) but high in summer (OC/EC: 10.2–18.9). These results imply a conclusion similar to that noted above. The anthropogenic pollutants were transported to CHAAMS from Asia in spring and winter, whereas in summer, monsoon winds transported clean oceanic air masses. The high OC/EC ratio also enabled us to identify the photochemical oxidation reaction.

In Eastern Asia (from Pakistan, China, and Mongolia to Japan, and also India, Southeast Asia, Philippines, and Korea), EC and OC emissions from China accounted for 61 % and 49 %, respectively, of the total emissions from the entire area (Zhang et al. 2009). Concentrations of EC and OC in the major cities in China were high (Yang et al. 2005). Weekly carbonaceous concentrations varied over wide ranges in, for example, Beijing ($8.6\text{--}59\text{ }\mu\text{g m}^{-3}$ for OC and $1.5\text{--}25.4\text{ }\mu\text{g m}^{-3}$ for EC) and Shanghai ($5.1\text{--}38.4\text{ }\mu\text{g m}^{-3}$ for OC and $2.3\text{--}13.0\text{ }\mu\text{g m}^{-3}$ for EC).

Generally speaking, air pollutants are transported from China toward Japan in association with the north or northwest monsoon in winter. High levels of air pollutants in China in winter are due to higher emission and poorer dispersion owing to low boundary layers and low wind speed. In spring, large-scale transport of air pollutants is caused by the movement of a low-pressure system or a migratory high-pressure system (Uno et al. 1998; Shimada et al. 2011).

2.5 PAHs

In many aerosols, organic (carbon-containing) particles are very small (around $0.1\text{--}1\text{ }\mu\text{m}$ in diameter) and these particles are one of the main components of PM_{2.5}. Organic aerosols can be divided into two kinds: primary and secondary. Primary organic aerosols (POAs) are mainly emitted by fossil fuel combustion.

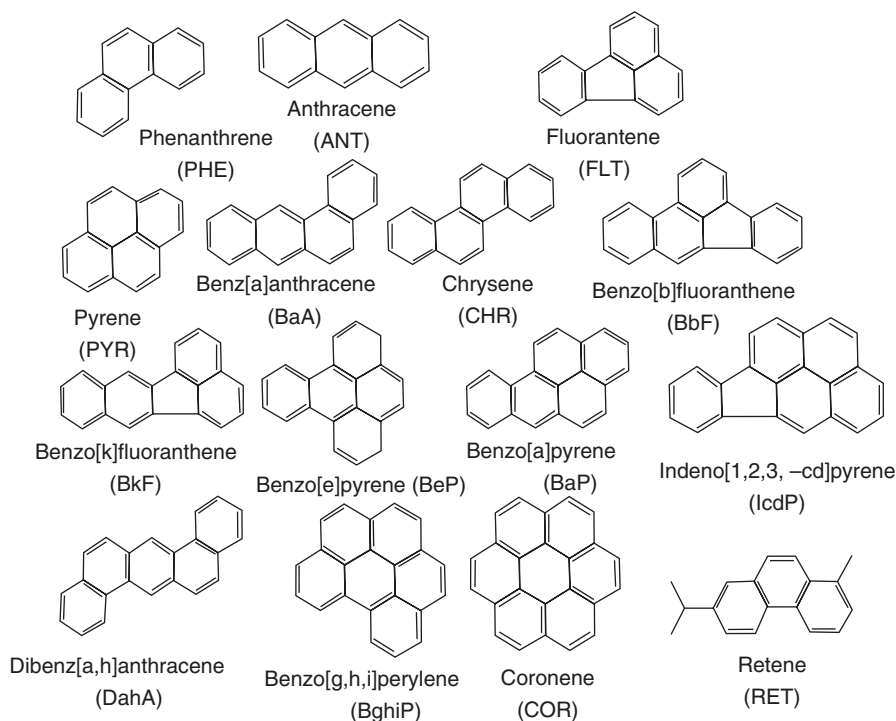


Fig. 2.8 Typical polycyclic aromatic hydrocarbons (PAHs) measured in this study

Cooling of high-temperature emission gas produces POA compounds, owing to the coagulation of organic compounds with dozens or more carbon atoms. Such compounds are emitted into the air as fine particles (0.1–0.5 μm in diameter). Secondary organic aerosols (SOAs) are produced in the atmosphere by the photo-oxidation of semi-volatile organic compounds forming very fine particles. Organic aerosols contain compounds such as fatty acids, low-molecular-weight di-carboxylic acids, long-chain alkanes, and PAHs. Since these organic compounds have origins of their own, they can be used as tracers to obtain information about the origins of aerosols.

PAHs (typical examples are shown in Fig. 2.8) are formed by the aromatization of parts of low-molecular-weight organic compounds that are produced by the thermal decomposition of organic compounds in the combustion process. PAHs are emitted mainly by the incomplete combustion of fossil fuels and bio-fuels, and often contain harmful compounds that are carcinogenic or mutagenic. For example, benzo[a]pyrene is classified as IARC Group 1 (carcinogenic to humans). PAHs represent 35–82% of the carcinogenicity of aerosols (Pedersen et al. 2004). They acquire higher toxicity or carcinogenicity by transformation to nitro-substituted PAHs or by transformation to quinones by atmospheric reaction with ozone, OH radicals, and NO_2 .

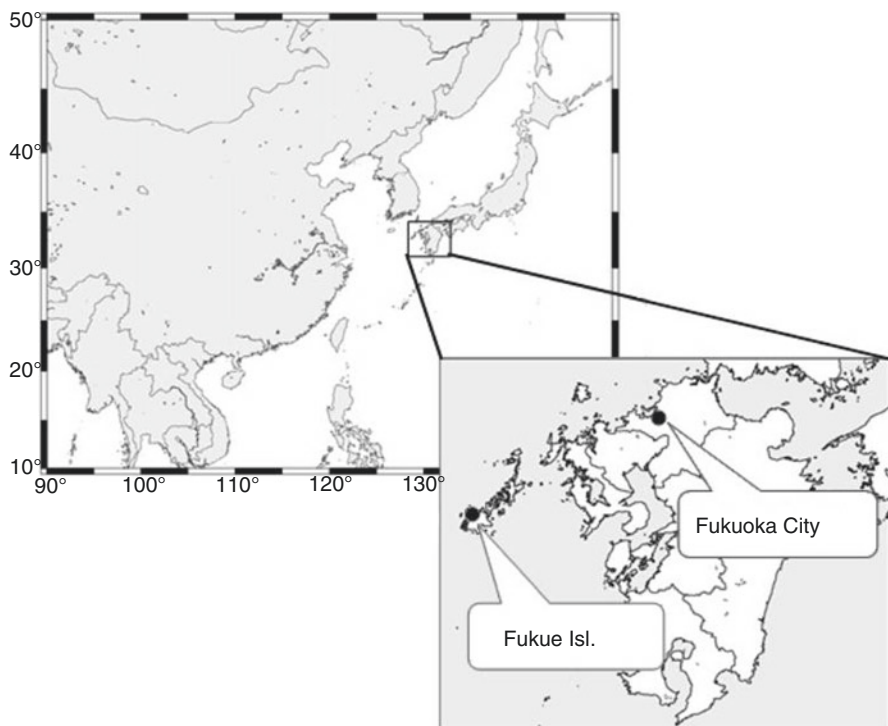


Fig. 2.9 Locations of Fukue Island and Fukuoka

The emission of PAHs in China accounts for 20% or more of world emission (Zhang and Tao 2009). The annual average concentrations of benzo[a]pyrene in Beijing and Xi'an are 11.6 and 15.6 ng m^{-3} , respectively, levels which exceed Chinese air quality standards (10 ng m^{-3} ; the WHO guideline is 1 ng m^{-3}) (Ma et al. 2011; Okuda et al. 2010).

Since Japan is located downwind of East Asia, aerosols emitted in East Asia can be easily transported to Japan. It is expected that domestic emission in Japan will decrease owing to the air quality regulations, but, nonetheless, an increase in PAH concentrations is expected because of their long-range transport. In western areas of Japan, such as Fukue Island, Nagasaki, and Fukuoka, the monthly average concentration levels and variations of $\text{PM}_{2.5}$ are quite similar and short-term increases in concentration are mainly due to $\text{PM}_{2.5}$ transport from East Asia (Kaneyasu et al. 2010, 2011). The same situation can be expected for PAHs.

From this viewpoint, the author and co-workers continually observed 15 PAHs (as shown in Fig. 2.8) on Fukue Island and in Fukuoka (Fig. 2.9) from 2009 to 2011 (Ogawa et al. 2012; Sato et al. 2007, 2009; Yoshino et al. 2011).

The seasonal average concentrations of 14 of these PAHs, except for retene (Fig. 2.8) on Fukue Island were found to be in the following order: summer (0.12 ng m^{-3}) < fall (1.68 ng m^{-3}) < spring (1.96 ng m^{-3}) < winter (3.26 ng m^{-3}) (Fig. 2.10).

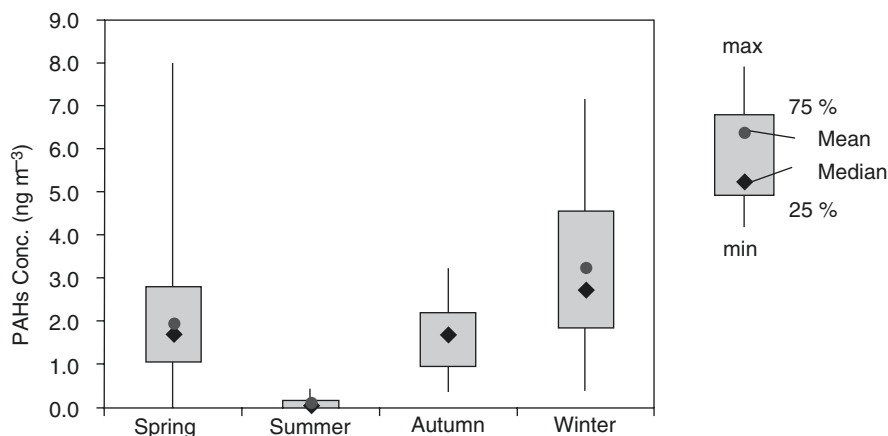


Fig. 2.10 Seasonal average concentrations of the PAHs shown in Fig. 2.8 (with the exception of retene) on Fukue Island

In winter, compared with the concentrations in spring and fall, the concentrations of CHR (chrysene), BbF (Benzo[b]fluoranthene), BkF (Benzo[k]fluoranthene), and BeP (Benzo[e]pyrene) (acronyms are shown in Fig. 2.8) were high, which suggests an increase in coal combustion. This is reasonable because in winter the use of coal for heating increases in northern China.

In contrast, in the Japanese megacity of Fukuoka, in addition to the PAHs arising from long-range transport, local-origin PAHs originating from automobiles have been shown to contribute to high PAH concentrations. If we assume that all PAHs measured on Fukue Island arrive by long-range transport, we can use the ratio of PAHs in Fukue to those in Fukuoka to estimate the fraction of transported PAHs in Fukuoka. The ratios in spring and winter are 0.66 ± 0.40 and 0.75 ± 0.20 , respectively, which means that the effect of long-range PAH transport in Fukuoka is at a level that cannot be disregarded.

2.6 Metallic Elements and Source Apportionment

Although the proportion of metallic elements in aerosols is not very large (less than 17%, as shown in Fig. 2.1), these elements are among the most important components of PM_{2.5}. The fine particles contain many elements. Some of these elements have been identified as being carcinogenic or mutagenic, such as Cr, Ni, Cd, and As (IARC 2015). In addition, metallic elements can serve as indicators of the origin of aerosols, because they do not participate in chemical reactions during long-range transport.

The concentrations of some metallic elements in Beijing (Yu et al. 2013) are listed in Table 2.1, where they are compared with those in Fukue Island, Nagasaki, and Fukuoka (Suzuki et al. 2014). The concentrations in Beijing are from 10 to more than 50 times higher than those in Fukuoka.

Table 2.1 Concentrations of metallic elements in Beijing, China, and Fukue Island and Fukuoka, Japan

Elements	Beijing ^a (ng m ⁻³)	Fukue ^b (ng m ⁻³)	Fukuoka ^b (ng m ⁻³)	Beijing ^c Fukuoka
V	15.3	1.48	1.97	10.3
Cr	22.4	0.54	0.84	41.5
Mn	62	4.74	6.83	13.1
Fe	1089	96.5	152	11.3
Ni	28.1	1.21	1.18	23.2
Cu	36.8	1.85	2.13	19.9
Zn	313	26.7	29.3	11.7
As	37.9	1.23	1.35	30.8
Se	38.1	0.67	0.67	56.9
Ba	88.2	1.53	4.73	57.6
Pb	117.3	8.41	10.1	13.9

^aAverage for four seasons^bAverage for three seasons (spring, summer, and winter)^cRatio of the concentration at Beijing against that at Fukuoka

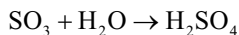
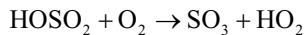
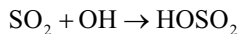
Recently, Yu et al. (2013) evaluated the origin of aerosols by the positive matrix factorization (PMF) method (Paatero 1997). Their analysis included not only metallic elements but also non-metallic elements such as S, Si, P, Cl, and Br. They optimized the aerosol origin factors and categorized them into seven types: secondary sulfur (13.8 $\mu\text{g m}^{-3}$, 26.5 %), vehicle exhaust (8.9 $\mu\text{g m}^{-3}$, 17.1 %), fossil fuel combustion (8.3 $\mu\text{g m}^{-3}$, 16 %), road dust (6.6 $\mu\text{g m}^{-3}$, 12.7 %), biomass burning (5.8 $\mu\text{g m}^{-3}$, 11.2 %), soil dust (5.4 $\mu\text{g m}^{-3}$, 10.4 %), and metal processing (3.1 $\mu\text{g m}^{-3}$, 6.0 %). They also reported that: “Fugitive dusts (including soil dust and road dust) showed the highest contribution of 20.7 $\mu\text{g m}^{-3}$ in the spring, doubling those in other seasons. On the contrary, contributions of the combustion source types (including biomass burning and fossil fuel combustion) were significantly higher in the fall (14.2 $\mu\text{g m}^{-3}$) and in the winter (24.5 $\mu\text{g m}^{-3}$) compared to those in the spring and summer (9.6 and 8.0 $\mu\text{g m}^{-3}$, respectively).”

In contrast, Suzuki et al. (2014) did not include non-metallic elements, and they determined five aerosol source factors for Fukue Island and four factors for Fukuoka. The five aerosol source factors for Fukue Island were *kosa*, sea salt, heavy oil combustion, coal combustion, and road dust, and the four factors for Fukuoka were *kosa*, heavy oil combustion, coal combustion, and road dust. Suzuki et al. (2014) also analyzed the source area of aerosols by using the total potential source contribution function (TPSCF) method (Hopke et al. 1995). At both sites, they found that *kosa* and emissions from coal combustion, both of which were evaluated on the basis of PMF analyses, originated in continental Asia, especially in China. A TPSCF plot of the road dust factor shows distributions over large cities such as Shanghai and Seoul. TPSCF plots of the oil combustion factor indicate that emissions from marine ships are also important at both sites.

2.7 Sulfate, Nitrate, and Ammonium Aerosols

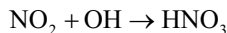
The most common aerosols in the atmosphere consist of sulfate (SO_4^{2-}) and nitrate (NO_3^-). These are not primary aerosols; they are secondary aerosols that form after precursor gases (SO_2 and NO_x) emitted into the air are oxidized by atmospheric reactions. In the atmosphere, there are both natural aerosols produced from gases emitted by, e.g., volcanos, and anthropogenic aerosols produced from gases produced by fossil fuel combustion.

Gaseous SO_2 is mainly oxidized in the air by OH radicals as follows:

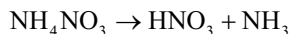
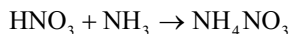


The resulting sulfuric acid (H_2SO_4) has such low volatility that it forms very fine particles. Moreover, it readily reacts with surrounding ammonia gas to form fine particles of ammonium sulfate $[(\text{NH}_4)_2\text{SO}_4]$, which are carried across wide areas by winds.

Similarly, NO_x is oxidized in air to form nitric acid (HNO_3) mainly by OH radical reactions, as follows:



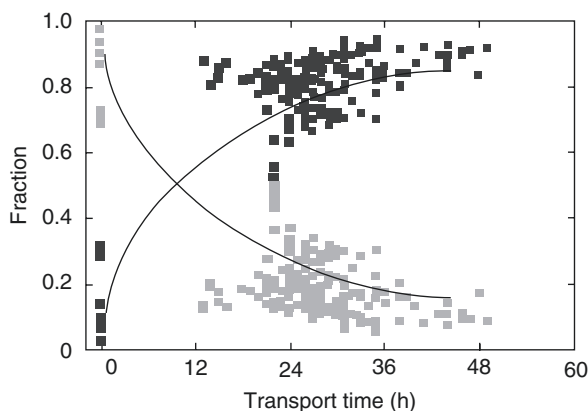
Nitric acid, however, is a gas under normal atmospheric conditions. Fine particles are created by its reaction with ammonia gas in the surrounding environment.



Unlike ammonium sulfate, ammonium nitrate is not very stable. It thermally decomposes during transport to form nitric acid gas and ammonia gas. The resulting nitric acid gas is adsorbed on the surfaces of coarse particles such as soil and sea-salt particles, and nitrate salts such as calcium nitrate $[\text{Ca}(\text{NO}_3)_2]$. Thus, nitrate is contained in coarse particles after long-range transport. As shown in Fig. 2.11, most nitrates are initially contained in fine particles, but they are primarily contained in coarse particles in Okinawa after long-range transport (Takiguchi et al. 2008). These findings show that air pollutants and atmospheric aerosols can be transformed during long-range transport.

More detailed size distributions of ionic species in aerosols were obtained by size-segregated collections and analyses of aerosols (Yumoto et al. 2015). Samples were collected by using a cascade impactor sampler with five stages. Aerosols in the size ranges of >10 , 2.5–10, 1–2.5, and 0.5–1 μm were collected on Teflon filters,

Fig. 2.11 Variations in nitrate fractions of particles with transport time. *Gray squares*: fraction of fine particulate nitrate in PM_{2.5}. *Black squares*: fraction of coarse particulate nitrate in PM₁₀. The transport time was calculated based on back trajectory analyses, by assuming that time 0 was when the trajectory left mainland China



and aerosols in the size range of 0.1–0.5 μm were collected with a stainless steel fiber filter (Otani et al. 2007).

During the sampling periods of April, October, and December 2012 and April 2013 at Cape Hedo, species from anthropogenic sources, such as NH_4^+ and SO_4^{2-} , were dominant among fine-mode particles (0.5–1, 0.1–0.5 μm). In contrast, species from natural sources, such as Ca^{2+} and Na^+ , were dominant among coarse-mode particles (>10, 2.5–10 μm). SO_4^{2-} was found in nearly all the samples. SO_4^{2-} in fine mode was non-sea salt, whereas in coarse mode it was sea salt. The size distribution of NO_3^- indicated that NH_4NO_3 had decomposed to gaseous NH_3 and HNO_3 during transport, and the resulting HNO_3 had been adsorbed on coarse particles by reacting with sea salt or crustal particles.

2.8 Trends of Ionic Species in Aerosols Transported from East Asia Measured by Aircraft

Anthropogenic emission in East Asia has been increasing because of rapid economic growth in this area. Atmospheric pollutants emitted in East Asia are observed even in North America, and they affect the entire northern hemisphere climate (UNECE 2011). Not only are the global aspects of long-range transport of pollutants very important, but so also are the regional aspects. From this perspective, we have continued aerial observations over the sea between continental Asia and Japan for 20 years in order to analyze the transport and transformation of long-range-transported atmospheric pollutants. The areas covered are the East China Sea, Sea of Japan, and the Yellow Sea. During this period, there was remarkable economic growth in East Asia, particularly in China, and as a result, the emission of atmospheric pollutants increased tremendously. Here, the results of our analyses of the long-term trends in the ionic components of aerosols are presented.

For the analyses we used 11 data sets, which were obtained in October 1991, November 1992, March and December 1994, January and December 1997, February

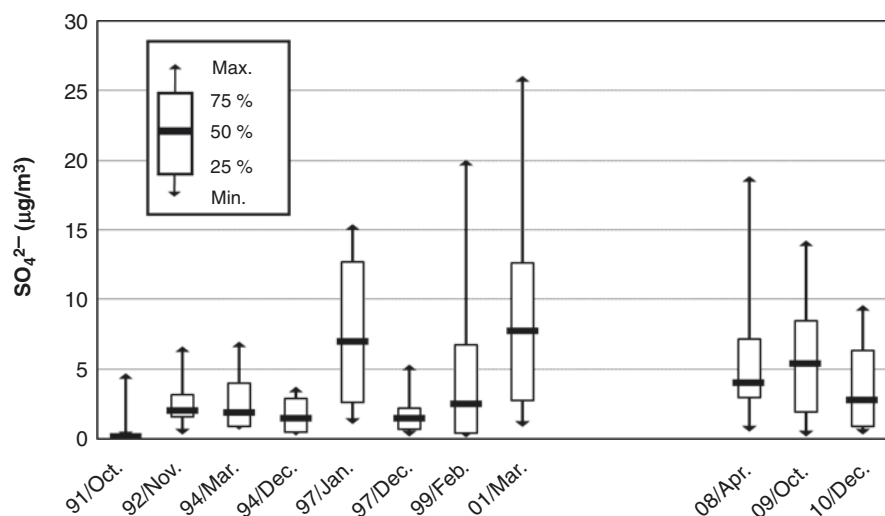


Fig. 2.12 Trend of sulfate in aerosols collected by aircraft

1999, March 2001, March–April 2008, October 2009, and December 2010 (Hatakeyama et al. 1995a, b, 1997, 2000, 2001, 2004, 2011, 2014).

Aerial observation is usually event-based and is difficult to continue over a long time. Many large projects have measured atmospheric aerosols all over the world (Clarke and Kapstin 2010). Although a lot of good data are available, long-term trends are not the usual target of such aerial observations. The greatest advantage of ground-based observations is that long-term trends and seasonal variations of air pollutants are usually the main targets of such studies. However, we continued aerial observations for about 20 years in almost the same area. Such observations have seldom been made elsewhere in the world, and we obtained very rare data.

The aircraft employed were a Cessna 404 and a Fairchild Swearingen Merlin IV, chartered from Showa Aviation Company (Yao, Japan), and a Beechcraft King Air 200 T, chartered from Diamond Air Service Incorporation (Toyoyama, Japan). The target area of the observations was the lower troposphere (below 3,000 m).

The common observations during 20 years of experiments were of gaseous species, such as ozone, SO_2 , and NO_x or NO_y (total reactive nitrogenous compounds: gaseous HNO_3 , organic nitrates, and inorganic nitrate salts in addition to NO_x), 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. We determined the trends of anthropogenic species in the aerosol phase (SO_4^{2-} , NO_3^- , and NH_4^+).

Figures 2.12, 2.13, and 2.14 present the trends of sulfate, ammonium, and nitrate ion concentrations, respectively, in aerosols collected on board the aircraft. Sulfate and ammonium ions showed quite similar trends, which indicates that these compounds existed in the form of $(\text{NH}_4)_2\text{SO}_4$ or $(\text{NH}_4)\text{HSO}_4$. Sulfate seems to have decreased from 2001, but unfortunately, since we lack data from 2002 to 2007

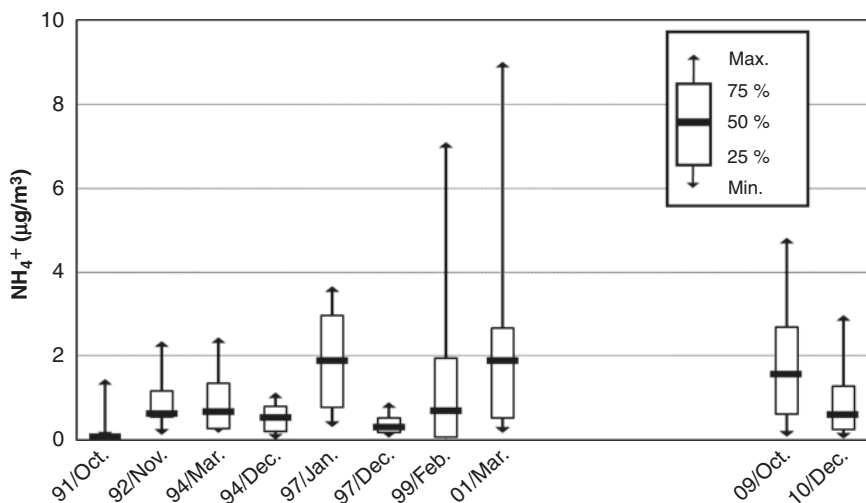


Fig. 2.13 Same as Fig. 2.12, for ammonium

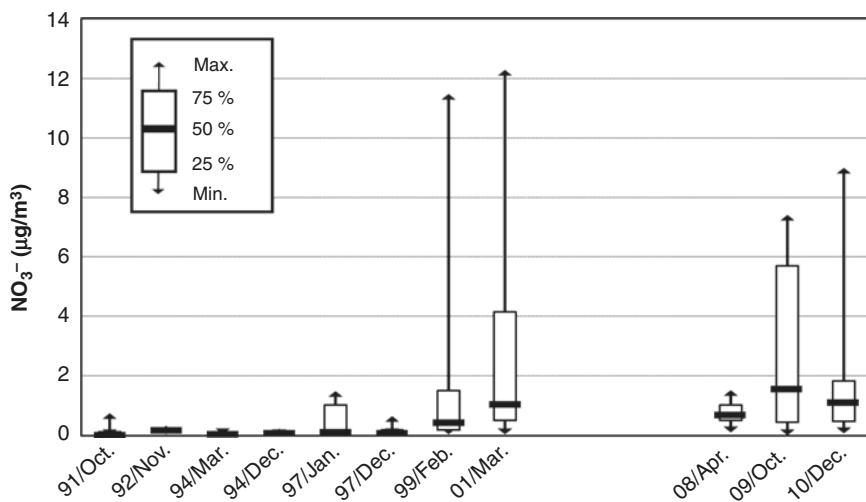


Fig. 2.14 Same as Fig. 2.12, for nitrate

(we carried out aerial observations over mainland China during that period), the trend is not clearly demonstrated. However, it has been reported that the emission of SO_2 in China has been decreasing since 2006 (Lu et al. 2010). Our results appear to agree with this finding, although a decreasing trend of SO_2 is not entirely evident in their work.

In contrast, nitrate was low before December 1997 (maximum $1.5 \mu\text{g m}^{-3}$), but it increased after 1999, with maximum concentrations exceeding $7 \mu\text{g m}^{-3}$. This seems to show an increasing trend. The emission of NO_x , which is a precursor of NO_3^- , is

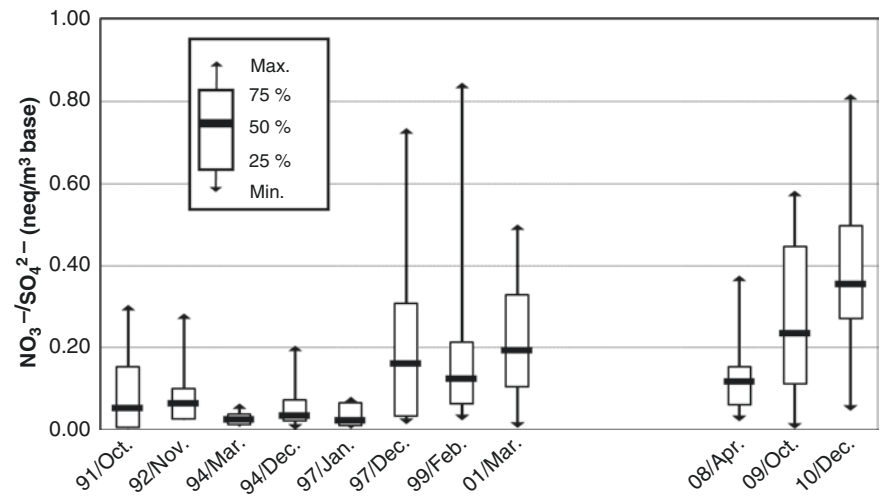


Fig. 2.15 Same as Fig. 2.12, for the ratio of nitrate/sulfate

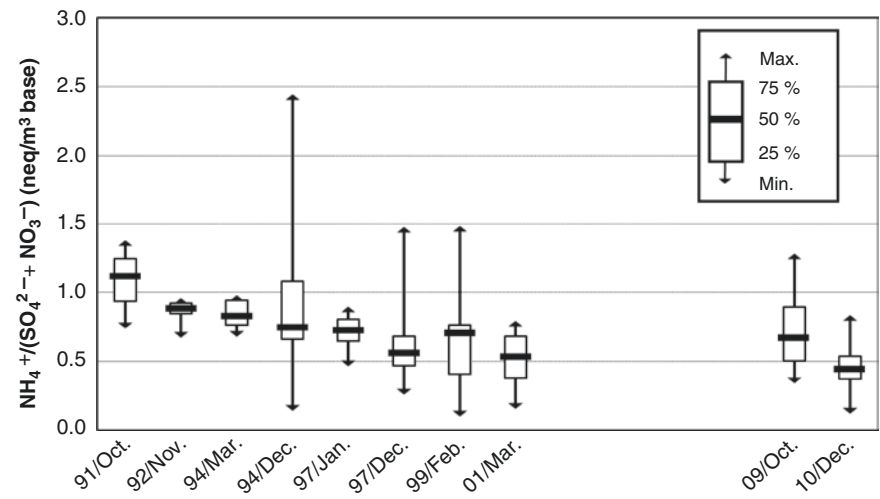


Fig. 2.16 Same as Fig. 2.12 for the ratio of ammonium/(sulfate + nitrate)

increasing in China (Kurokawa et al. 2013), and this could cause NO_3^- to increase over the East China Sea. More clearly, the ratio of $\text{NO}_3^-/\text{SO}_4^{2-}$ has increased, and this reflects a decreasing trend of sulfate and an increasing trend of nitrate (Fig. 2.15). Furthermore, the ratio of NH_4^+ to the sum of SO_4^{2-} and NO_3^- is steadily coming down (Fig. 2.16). This means that the acidity of aerosols over the sea between continental Asia and Japan is still growing, and this could lead to more serious acid deposition in this area.

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