PERGAMON

Atmospheric Environment 34 (2000) 2755–2770

ATMOSPHERIC ENVIRONMENT

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The concentration, trend and seasonal variation of metals in the atmosphere in 16 Japanese cities shown by the results of National Air Surveillance Network (NASN) from 1974 to 1996

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Received 5 March 1999; received in revised form 28 June 1999; accepted 15 July 1999

Abstract

In this study, the data from National Air Surveillance Network (NASN) for selected crustal elements (Al, Ca, Fe, Mn, Sc and Ti), anthropogenic elements (As, Cu, Cr, Ni, Pb, V and Zn) and a marine element (Na) in atmospheric particulate matter have been evaluated over Japan from 1974 to 1996. The NASN had 16 sampling stations, Nopporo, Sapporo, Nonotake, Sendai, Niigata, Tokyo, Kawasaki, Nagoya, Kyoto-Hachiman, Osaka, Amagasaki, Kurashiki, Matsue, Ube, Chikugo-Ogori and Ohmuta in Japan, at which aerosols were regularly collected every month and samples collected by low volume air sampler were analyzed by neutron activation analysis (NAA) and X-ray fluorescence (XRF). Results of trend analysis showed that the annual average concentrations of crustal and anthropogenic elements decreased significantly between 1974 and 1996. Furthermore, the concentrations of selected crustal and anthropogenic elements were higher between 1974 and 1979 than between 1980 and 1989. Crustal elements displayed high concentrations in the "Kosa" period (the dust season from March to May) and in winter (November and December) and low concentrations in summer (July and August). The ratios of elemental average concentrations in the Kosa period to those of summer were 2–5 times higher and those of winter relative to those of summer were 1.5–3 times higher for crustally derived elements. Particularly, concentration of Sc in Matsue was 8 times higher during the Kosa period than during summer time. Some of anthropogenic elements also showed high concentrations in the Kosa Season and low concentrations in summer. The ratios of elemental concentration in the Kosa period to those of summer and those of winter relative to summer were 2–4 times higher for some anthropogenically derived elements. Especially, the concentration of Zn in Chikugo-Ogori was 5.7 times higher during wintertime than during summertime. © 2000 Elsevier Science Ltd. All rights reserved.

Keywords: Crustal elements; Anthropogenic elements; Marine element; Atmospheric particulate matter; National Air Surveillance Network (NASN) in Japan

1. Introduction

Once anthropogenic and natural emissions are released into the atmosphere, a complex mix of physical and chemical processes redistributes them locally, regionally and globally. The measurement of emissions from either individual or multiple sources is very important to understand and predict the present and future status of the air quality. In order to meet these needs, concentrations of air pollutants have been measured in either marine or continental atmospheres using different methods over the world.

Concerning this interest, studies on the transport and deposition of atmospheric trace elements have received increasing attention, especially with regard to the issue of

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PII: S1352-2310(99)00353-2

long range transport. Concentrations of trace elements associated with aerosol particles have been measured in remote (Tuncel et al., 1989; Blanchet, 1989; Artaxo et al., 1990; Savarino et al., 1994; Mukai and Suzuki, 1996), marine (Duce et al., 1980; Uematsu et al., 1983; Chester et al., 1991; Losno et al., 1992; Arimoto et al., 1995) and continental/urban areas (Amundsen et al., 1992; Lowenthal et al., 1992; Dokiya et al., 1995; Rahn et al., 1989; Fan et al., 1996), respectively. Most of these measurements were done for several years or only for a short time based on seasonal changes of the concern area due to limitations of equipment, personnel and funds. However, National Air Surveillance Network in Japan has sequentially collected air quality data including aerosol particles without any interruption for a long time on many measured items by almost the same methods of collection and analysis. From this point of view NASN network must be unique. Such a comprehensive network cannot be found elsewhere in the world.

From time to time, the data from NASN has been reported to explain long-range transport of air pollutants from the East Asian region to Japan for limited periods or for some selected location (Mukai et al., 1990,1994; Hashimoto et al., 1990,1994). However, there are a few researches, which were done using NASN data over Japan (Tanaka et al., 1983,1989,1990; Hashimoto et al., 1992). Such works focused on long-range transport of soil derived elements from East Asia region during the Kosa period and the inventory of some elements such as Al in the atmospheric air especially.

Nevertheless, there are no comprehensive studies of the NASN data except some reports published by Environmental Protection Agency (EPA) in Japan, which examine features in detail of the quality of air pollution over Japan. In fact, a large-scale NASN survey had been carried out regularly (1974–1996) in Japan to provide information on time trends and spatial changes of metals in atmospheric particulate matter. Regardless of the initial purposes of NASN, the data already produced by this network is very important and will greatly contribute to future works.

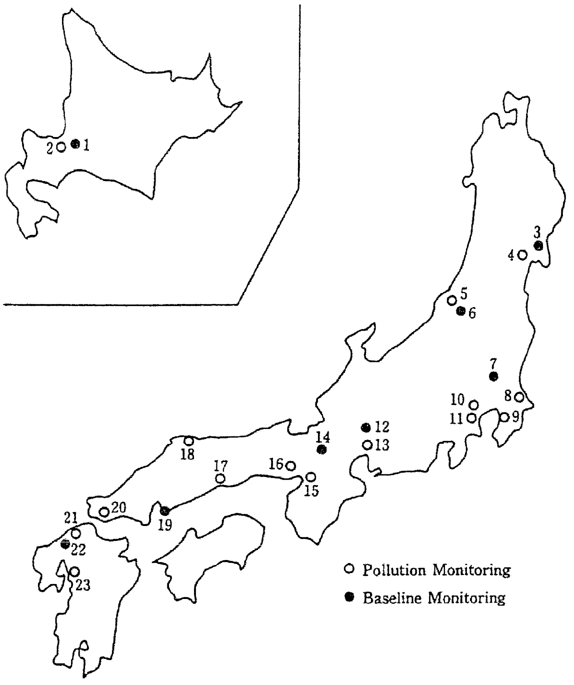
Therefore, the aim of this study is to review the history of NASN, introduce the outlines of NASN for other researchers and show the concentrations, the trend and seasonal variation of some selected crustal and anthropogenic elements including a marine element in atmospheric particulate matter observed for 23 yr across Japan.

2. National Air Surveillance Network (NASN)

A well-organized air monitoring network (National Air Surveillance Network of Japan) was established by the Japanese Government in 1965, following a similar air monitoring network in the US, and has continued to measure air quality throughout Japan since then.

At the beginning of NASN during the 1960s, the Japanese Government was facing serious problem of air pollution, which increased with the rapid progress of industries. NASN was created as an attempt to survey the whole status of air pollution in Japan for the purpose of legislating, controlling and preventing air pollution. The data from NASN, accumulated for more than 20 yr show the changes of air pollution both in degree and character in Japan. Minor changes have been made in the measured items and methods of measurement. One of the special characteristics of NASN is the application of neutron activation method of analysis (NAA) for the measurement of trace elements in atmospheric samples. This method and the X-ray fluorescence method measure concentrations of 31 elements including rare earths.

NASN can measure more than 15 kinds of gaseous and particulate matter, as well as meteorological observations of temperature, relative humidity, precipitation, and solar radiation. For monitoring suspended particulate matter, SPM, the 23 stations (Fig. 1) return



1. Nopporo 2. Sapporo 3. Nonotake 4. Sendai 5. Niigata 6. Niitsu
7. Tsukuba 8. Kashima 9. Ichihara 10. Tokyo 11. Kawasaki 12. Inuyama
13. Nagoya 14. Kyoto-Hachiman 15. Osaka 16. Amagasaki 17. Kurashiki
18. Matsue 19. Kurahashijima 20. Ube 21. Kita-Kyushu 22. Chikugo-Ogouri
23. Ohmuta

Fig. 1. Map of monitoring station of NASN in Japan.

collected filters to the central analytical laboratory (Japan Environmental Sanitation Center) located in Kawasaki. Among these stations, 16 stations collect SPM for multi-elemental neutron activation analysis on a monthly basis (25-days sampling period). One-third of the network, i.e., 8 stations, is for baseline monitoring, while the remaining 15 stations are for monitoring air pollutants in urban areas.

The first NASN results were published in 1966, one year after network operations started. Growth of the network with years, number of stations and changes in sampling and analytical methods, is shown as follows:

At the beginning of NASN (1966), three stations were established. These stations were Tokyo, Osaka and Amagasaki. Hi-Vol sampling (24-h sample twice a month) was done and analysis performed using emission spectrographic method for 10 elements (Be, Ti, V, Cr, Mn, Fe, Ni, Cu, Cd, Pb). In 1971, ten stations (Sapporo, Kashima, Ichihara, Kawasaki, Nagoya, Matsue, Kurashiki, Ube, Kita-Kyushu and Ohmuta) were added to these stations. Both Hi-Vol (24-h sample once a month) and Lo-Vol (25-days sample once a month) samplings were accomplished and atomic absorption spectrometry (AAS) was used to analyze for nine elements (V, Mn, Fe, Co, Ni, Cu, Zn, Cd, Pb).

From 1974 to 1996, 16 stations for both atomic absorption spectrometry (AAS) and instrumental neutron activation analysis (INAA) were grouped into 2 parts: 12 stations for pollution monitoring (Sapporo, Niigata, Sendai, Tokyo, Kawasaki, Nagoya, Osaka, Amagasaki, Matsue, Kurashiki, Ube, Ohmuta) and four baseline stations (Nopporo, Nonotake, Kyoto-Hachiman, Chikugo-Ogori). Then, seven additional stations for only AAS analysis were grouped into three stations (Kashima, Ichihara, Kita-Kyushu) and four baseline stations (Niitsu, Tsukuba, Inuyama, Kurahashijima). Hi-Vol (24-h sample once a month at all 23 stations) and Lo-Vol (25 day samples once a month at the 16 stations) samplings were performed. Hi-Vol samples were analyzed by AAS for nine elements (V, Mn, Fe, Co, Ni, Cu, Zn, Cd, Pb); Lo-Vol samples were analyzed by XRF for three elements (Cd, Ni, Pb) and by using INAA for 28 elements (Ag, Al, As, Ba, Br, Ca, Ce, Cl, Co, Cr, Cs, Cu, Fe, Hf, K, La, Lu, Mn, Na, Sb, Sc, Se, Sm, Th, Ti, V, W, Zn).

3. Results and discussions

3.1. Average concentrations

The arithmetic mean concentrations and standard deviations of 14 metal elements of Al, Ca, Fe, Mn, Na, Sc, Ti, As, Cu, Cr, Ni, Pb, V and Zn in the atmospheric particulate matter observed in 16 cities over Japan for 23 yr from 1974 to 1996 at each sampling location are presented in Table 1. During evaluation, the annual average concentration of each element calculated based on 12 monthly average data between April (beginning of financial year in Japan) and March was taken from NASN data reports; then, the long-term (23 yr) average concentration was determined from the annual average concentration of each element.

As can be seen in Table 1, the highest average concentrations were observed in Kawasaki (Fe, Ti, Mn, Cu, Ni and V), Osaka (Na, Cr, Pb and Zn), Sapporo (Al and Sc), Ohmuta (Ca) and Niigata (As), respectively. These cities are either industrial or large cities of Japan. Conversely, their lowest average concentrations were noticed in Nonotake (Al, Ca, Fe, Sc, Ti, Cu, Cr, Ni, V and Zn) and Nopporo (Mn, As and Pb), as expected. Nonotake and Nopporo were assumed as baseline-remote area in Japan as shown in Fig. 1.

The period of measurement for NASN data is quite long. Therefore, it is not possible to compare the average concentrations for the long-term average concentrations of Japanese cities to those of other countries directly. Therefore, NASN data for selected periods was compared with results from elsewhere.

In comparing the concentrations with Beijing (Sekine et al., 1992), China, during 1 yr (January 1989–December 1989), concentrations of crustal elements such as Al, Fe, Mn were lower in cities in Japan for the mentioned period. For example, the average concentration of Al in Beijing was measured as 13 200 ng m^{-3} while the average concentration of Al was measured as 780 ng m^{-3} in Sapporo which was the highest value in Japan. The reason for the very high concentration of crustal elements in Beijing can be explained to the influence of the Kosa blown from the vast northern desert.

For the same period, anthropogenic elements such as Zn, Pb, Cu, Cd, Ni and V showed lower values in cities of Japan compared to those of Beijing. For example, the concentration of Pb displayed extremely high values (222 ng m^{-3}) in Beijing, while the range of Pb concentrations was between 8.6 and 97 ng m^{-3} in cities of Japan. The regulation of leaded gasoline first started in Japan in 1975 and its production has been stopped since 1987.

Another comparison can be made with Seoul, Korean data (Hashimoto et al., 1990) for the 2-yr period from April 1987 through March 1989. Atmospheric elemental concentrations in Seoul were generally higher than those in the two Japanese large cities (Tokyo and Kawasaki). Soil-derived elements such as Al, Sc showed particularly higher concentrations. In addition, concentrations of anthropogenic elements such as Ni, As and V, which were supposed to be emitted from fossil fuel combustion were also higher in Seoul compared to those in Tokyo and Kawasaki. A higher level of Pb in Seoul must be due to automobile exhaust emissions, since the consumption of

Table 1
Annual concentration of the elements in atmospheric particulate matter over Japan between 1974 and 1996 (sampling locations are depicted in Fig. 1)

Element	Napporo	Sapporo	Nonotake	Sendai	Niigata	Tokyo	Kawasaki	Nagoya	Kyoto-H.	Osaka	Amagasaki	Kurashiki	Matsue	Ube	Chikugo-O.	Ohmuta
Al	322.9 ± 176.8 (*n* = 18)	908.2 ± 406.6 (*n* = 23)	237.4 ± 94.3 (*n* = 18)	626.1 ± 375.6 (*n* = 23)	398.1 ± 205.2 (*n* = 21)	429.1 ± 164.5 (*n* = 23)	586.1 ± 195.9 (*n* = 23)	440 ± 129.2 (*n* = 20)	375.0 ± 149.4 (*n* = 16)	555.2 ± 153.9 (*n* = 23)	428.5 ± 119.7 (*n* = 20)	509.4 ± 127.3 (*n* = 18)	489.1 ± 184.7 (*n* = 23)	538.1 ± 165.0 (*n* = 16)	456.7 ± 218.9 (*n* = 18)	829.1 ± 192.4 (*n* = 23)
Ca	216.1 ± 128.7 (*n* = 18)	672.4 ± 281.8 (*n* = 21)	185.8 ± 69.6 (*n* = 16)	570 ± 292.9 (*n* = 21)	338.6 ± 148.8 (*n* = 21)	468.9 ± 238.9 (*n* = 21)	675.2 ± 154.4 (*n* = 21)	328.5 ± 99.3 (*n* = 20)	246.3 ± 67.8 (*n* = 15)	542.4 ± 138.0 (*n* = 21)	410.5 ± 129.4 (*n* = 16)	411.1 ± 92.8 (*n* = 18)	365.2 ± 140.5 (*n* = 21)	541.3 ± 202.9 (*n* = 16)	299.4 ± 119.1 (*n* = 18)	716.7 ± 214.1 (*n* = 21)
Fe	206.1 ± 102.8 (*n* = 18)	625.2 ± 263.7 (*n* = 23)	157.6 ± 48.9 (*n* = 16)	513.9 ± 362.9 (*n* = 23)	427.4 ± 241.5 (*n* = 21)	676.9 ± 202.5 (*n* = 23)	1246.1 ± 316.9 (*n* = 23)	418 ± 101.2 (*n* = 20)	407.5 ± 120.9 (*n* = 20)	940.9 ± 161.0 (*n* = 23)	916.0 ± 256.5 (*n* = 20)	626.1 ± 112.4 (*n* = 18)	364.3 ± 185.6 (*n* = 23)	481.2 ± 124.9 (*n* = 16)	301.1 ± 130.0 (*n* = 18)	729.1 ± 212.9 (*n* = 23)
Ti	21.5 ± 11.9 (*n* = 18)	53.3 ± 23.1 (*n* = 21)	15.4 ± 5.3 (*n* = 16)	42.5 ± 24.4 (*n* = 21)	31.6 ± 13.1 (*n* = 21)	40.2 ± 17.2 (*n* = 21)	55.2 ± 15.9 (*n* = 21)	34.2 ± 11.1 (*n* = 20)	32.7 ± 10.8 (*n* = 16)	51.7 ± 14.6 (*n* = 21)	39.5 ± 10.2 (*n* = 20)	39.3 ± 8.8 (*n* = 18)	32.1 ± 13.9 (*n* = 23)	41.2 ± 15.3 (*n* = 16)	28.8 ± 14.8 (*n* = 18)	47.6 ± 11.5 (*n* = 21)
Mn	6.47 ± 2.36 (*n* = 18)	16.8 ± 6.99 (*n* = 23)	6.61 ± 2.00 (*n* = 16)	18.6 ± 9.99 (*n* = 23)	63.1 ± 42.9 (*n* = 21)	40.1 ± 11.5 (*n* = 23)	70.2 ± 36.3 (*n* = 23)	26.4 ± 6.69 (*n* = 20)	21.0 ± 4.97 (*n* = 16)	50.7 ± 13.9 (*n* = 23)	52.5 ± 10.7 (*n* = 16)	33.9 ± 5.87 (*n* = 18)	14.4 ± 3.43 (*n* = 23)	25.9 ± 10.4 (*n* = 23)	12.2 ± 3.20 (*n* = 18)	24.4 ± 10.5 (*n* = 23)
Na	471.1 ± 181.8 (*n* = 18)	728.7 ± 194.6 (*n* = 23)	538.8 ± 169.4 (*n* = 16)	703.9 ± 185.9 (*n* = 23)	778.6 ± 260.6 (*n* = 21)	637.0 ± 172.5 (*n* = 23)	909.6 ± 153.5 (*n* = 21)	622.0 ± 146.5 (*n* = 20)	459.4 ± 126.4 (*n* = 16)	920.9 ± 181.7 (*n* = 23)	746.0 ± 173.1 (*n* = 20)	559.4 ± 110.8 (*n* = 18)	823.9 ± 151.9 (*n* = 23)	778.8 ± 186.0 (*n* = 16)	450.6 ± 167.9 (*n* = 18)	832.6 ± 174.3 (*n* = 23)
Sc	0.06 ± 0.03 (*n* = 18)	0.20 ± 0.09 (*n* = 21)	0.04 ± 0.01 (*n* = 16)	0.14 ± 0.10 (*n* = 21)	0.08 ± 0.05 (*n* = 21)	0.09 ± 0.04 (*n* = 21)	0.12 ± 0.04 (*n* = 18)	0.07 ± 0.02 (*n* = 20)	0.06 ± 0.03 (*n* = 16)	0.09 ± 0.03 (*n* = 21)	0.08 ± 0.03 (*n* = 20)	0.09 ± 0.01 (*n* = 18)	0.10 ± 0.05 (*n* = 21)	0.09 ± 0.03 (*n* = 16)	0.08 ± 0.04 (*n* = 18)	0.17 ± 0.04 (*n* = 21)
As	1.57 ± 0.59 (*n* = 18)	2.93 ± 1.56 (*n* = 21)	2.74 ± 0.73 (*n* = 16)	2.90 ± 0.94 (*n* = 21)	10.4 ± 8.11 (*n* = 21)	3.73 ± 0.95 (*n* = 21)	4.37 ± 1.92 (*n* = 23)	4.24 ± 1.06 (*n* = 23)	4.65 ± 1.58 (*n* = 16)	9.30 ± 3.85 (*n* = 21)	5.17 ± 1.62 (*n* = 20)	7.25 ± 2.81 (*n* = 18)	4.76 ± 1.42 (*n* = 21)	4.90 ± 1.69 (*n* = 16)	5.15 ± 1.86 (*n* = 18)	9.30 ± 3.85 (*n* = 21)
Cu	14.4 ± 8.33 (*n* = 18)	20.9 ± 11.3 (*n* = 23)	8.20 ± 2.83 (*n* = 16)	11.4 ± 2.84 (*n* = 23)	10.6 ± 4.69 (*n* = 21)	30.2 ± 6.04 (*n* = 23)	100.5 ± 76.7 (*n* = 23)	15.5 ± 3.35 (*n* = 23)	23.0 ± 5.09 (*n* = 18)	20.9 ± 11.3 (*n* = 23)	27.9 ± 5.23 (*n* = 20)	11.4 ± 2.84 (*n* = 23)	10.6 ± 4.69 (*n* = 21)	30.2 ± 6.04 (*n* = 23)	13.9 ± 4.22 (*n* = 18)	15.5 ± 3.35 (*n* = 23)
Cr	1.56 ± 0.75 (*n* = 18)	2.61 ± 0.99 (*n* = 23)	1.14 ± 0.61 (*n* = 16)	3.63 ± 2.66 (*n* = 23)	9.96 ± 6.18 (*n* = 21)	6.09 ± 1.74 (*n* = 23)	19.3 ± 4.79 (*n* = 23)	5.85 ± 1.72 (*n* = 20)	3.59 ± 1.11 (*n* = 16)	43.7 ± 12.0 (*n* = 23)	13.2 ± 2.56 (*n* = 20)	22.3 ± 2.56 (*n* = 18)	8.37 ± 3.53 (*n* = 23)	20.4 ± 22.3 (*n* = 16)	1.98 ± 0.79 (*n* = 18)	21.2 ± 3.72 (*n* = 23)
Ni	2.40 ± 1.55 (*n* = 18)	3.81 ± 2.20 (*n* = 23)	1.48 ± 0.69 (*n* = 16)	2.84 ± 2.04 (*n* = 23)	7.76 ± 2.12 (*n* = 21)	5.63 ± 2.04 (*n* = 23)	13.6 ± 3.79 (*n* = 23)	5.58 ± 1.22 (*n* = 20)	4.36 ± 19.9 (*n* = 16)	8.70 ± 2.54 (*n* = 23)	12.4 ± 2.72 (*n* = 20)	4.94 ± 1.31 (*n* = 18)	2.65 ± 1.41 (*n* = 23)	7.33 ± 1.43 (*n* = 16)	3.47 ± 1.42 (*n* = 18)	4.76 ± 1.76 (*n* = 23)
Pb	17.5 ± 13.2 (*n* = 18)	43.9 ± 35.6 (*n* = 23)	23.2 ± 10.8 (*n* = 16)	39.2 ± 26.3 (*n* = 23)	73.4 ± 53.1 (*n* = 18)	124.7 ± 66.8 (*n* = 23)	146.1 ± 94.1 (*n* = 23)	71.5 ± 20.0 (*n* = 20)	70.8 ± 13.2 (*n* = 18)	173.6 ± 107.8 (*n* = 23)	87.6 ± 37.0 (*n* = 20)	64.3 ± 20.6 (*n* = 18)	31.3 ± 12.2 (*n* = 23)	56.9 ± 18.3 (*n* = 16)	44.7 ± 9.88 (*n* = 18)	126.0 ± 72.8 (*n* = 23)
V	3.52 ± 1.66 (*n* = 18)	5.19 ± 3.24 (*n* = 23)	2.44 ± 0.79 (*n* = 16)	4.00 ± 2.00 (*n* = 23)	5.98 ± 2.36 (*n* = 21)	8.90 ± 3.29 (*n* = 23)	14.9 ± 7.81 (*n* = 23)	9.59 ± 2.35 (*n* = 20)	5.94 ± 1.89 (*n* = 16)	14.3 ± 7.33 (*n* = 23)	13.5 ± 4.02 (*n* = 20)	8.35 ± 2.37 (*n* = 18)	4.33 ± 2.57 (*n* = 23)	9.85 ± 1.33 (*n* = 16)	5.89 ± 1.49 (*n* = 18)	7.30 ± 1.87 (*n* = 23)
Zn	39.6 ± 7.6 (*n* = 18)	149.3 ± 34.8 (*n* = 23)	38.3 ± 12.1 (*n* = 16)	181.9 ± 100.7 (*n* = 23)	259.9 ± 62.2 (*n* = 23)	298.7 ± 89.9 (*n* = 23)	190 ± 34.3 (*n* = 20)	196.3 ± 70.8 (*n* = 16)	196.3 ± 70.8 (*n* = 16)	327.8 ± 78.6 (*n* = 23)	247.5 ± 35.7 (*n* = 20)	116.0 ± 17 (*n* = 18)	40.7 ± 4.8 (*n* = 23)	111.4 ± 15.7 (*n* = 16)	234.1 ± 132.4 (*n* = 18)	290.4 ± 86.2 (*n* = 23)

Note: Concentrations are given in ng m^{-3} of air.

leaded gasoline had not been regulated in Seoul when these samplings were made at the monitoring station. The regulation of leaded gasoline started in 1988 in Seoul and as reported by Hashimoto et al. (1994), a steady decrease in Pb levels was observed except for the period before October 1988.

An additional comparison can be made with the data (Lee et al., 1994) in Walsall in UK for the 13-yr period from 1976 through 1989. For example, concentrations of soil elements such as Al, Fe, Sc in Walsall considered as an industrial city were higher than those in industrial cities in Japan except Ohmuta for Al. The concentration of Al was measured as 838 ng m^{-3} in Walsall whereas in Niigata, Kawasaki, Amagasaki and Ube it was measured as 494, 643, 469 and 629 ng m^{-3}, respectively. The concentration of Al in Ohmuta (898 ng m^{-3}) was found slightly higher than in Walsall at the same time. In the case of Fe concentration, in Walsall it was significantly higher than those in all industrial cities of Japan. At the same time, anthropogenic elements such as As, V, Ni, Zn, and Pb in Walsall also showed considerably higher values compared to those in industrial cities of Japan. For example, a very high concentration of Pb (1316 ng m^{-3}) was measured in Walsall whereas concentration of Pb was 94.7 ng m^{-3} in Niigata. In connection with this interest, related data for other urban and rural data from Europe, North America and Asia are reviewed elsewhere (Lee et al., 1994).

3.2. *Trend of concentration changes for 23 years*

In order to see the variation of annual average concentrations of metal elements in atmospheric particulate matter in Japan with time, 16 Japanese cities in NASN were classified into four groups. The population and main industry of each city are shown in Table 2. These groups are (1) five industrial cities (Niigata, Amagasaki, Kawasaki, Ohmuta, Ube), (2) five large cities (Sapporo, Sendai, Tokyo, Nagoya, Osaka), (3) four small cities (Kyoto-Hachiman, Kurashiki, Chikugo-Ogori, Matsue) and (4) two baseline-remote sites (Nopporo, Nonotake), respectively. Nopporo is located in the suburb of Sapporo at an altitude of 100 m and Nonotake is located on a mountain of an altitude of 223 m far from Sendai. Therefore, it is considered that these sites are not affected by the polluted area. Then, the trend of metal element concentration for each group city was investigated for both six crustally derived metal elements (Al, Ca, Fe, Mn, Sc, and Ti) and seven anthropogenically derived elements (As, Cu, Cr, Ni, Pb, V and Zn) for 23 yr periods from 1974 through 1996. In addition, concentrations of Na and suspended particulate matter in 16 cities in NASN were also evaluated. During the work, the large data set of 4416 samples collected by low volume sampler every month and 31 elements analyzed in each sample by neutron activation method and X-ray fluorescence method were taken into account.

Fig. 2 shows selected crustally derived element concentrations of Al and Sc in the atmospheric particulate matter observed in cities in NASN from 1974 to 1996. As can be seen in Fig. 2, the concentration of Al declined clearly in large cities compared to the industrial and small cities, and baseline sites until 1990 and mean concentrations of Al ranged between 1200–50, 1600–170, 1100–140 and 610–54 ng m^{-3}, in industrial, large and small cities, and baseline sites, respectively. Concentrations of Sc also show similar trends to that of Al in large

Table 2
Population and main industry of each city

Group name	City name	Population (March 31, 1996)	Main industry
Industrial cities	Niigata	486 001	Petrochemical industry
	Amagasaki	477 698	Iron and steel industry
	Kawasaki	1 188 981	Iron and steel industry
	Ohmuta	173 494	Petrochemical industry
	Ube	144 237	Petrochemical industry
Large cities	Sapporo	1 750 627	—
	Sendai	946 652	—
	Tokyo	7 963 613	—
	Nagoya	2 093 705	—
	Osaka	2 486 357	—
Small cities	Kyoto-Hachiman	74 966	—
	Kurashiki	426 458	—
	Chikugo-Ogori	51 559	—
	Matsue	146 153	—
Baseline-remote sites	Nopporo	117 073	—
	Nonotake	20 466	—

cities particularly. In the case of mean concentrations of Sc, it varied between 0.26–0.012, 0.43–0.039, 0.27–0.026 and 0.14–0.054 ng m^{-3}, in industrial, large and small cities, and baseline sites, respectively.

On the other hand, concentrations of suspended particulate matter over Japan between 1974 and 1996 showed no significant increasing or decreasing trends as shown in Fig. 3. The reason for this is that suspended particulate matter has been emitted continuously from automobile exhaust especially in large cities besides other sources. Mean concentrations of SPM ranged between 62–12, 58–15, 45–19 and 33–7.3 μg m^{-3}, in industrial, large and small cities, and baseline sites, respectively. As can be seen in Fig. 3, concentrations of Ca decreased gradually in large cities before 1990. After 1990, concentrations did not display any peak concentration and continue to decrease in most cities. Mean concentrations of Ca varied between 1400–60, 1100–97, 770–64 and 500–64 ng m^{-3}, in industrial, large and small cities, and baseline sites, respectively. Moreover, concentrations of Ti decreased rapidly in industrial cities. Concentrations of Mn obviously reduced in both industrial and large cities.

In general, small cities and baseline-remote sites did not show any clear trend compared to large and industrial cities. In addition, concentrations of crustal-derived elements were relatively higher during the period of 1974–1979 than the period of 1980–1989. The source of crustal-derived elements in the atmosphere is mainly wind-blown soil particles; however, in industrial and/or urban areas, human activities such as metal industry, and fuel combustion can be origins of soil-derived elements in the atmosphere. Therefore, it is difficult to evaluate the cause for the trend in crustal elements. Consequently, where many sources of crustal elements could exist in the area, the chemical mass balance method would be better applied to clarify the influence of sources as suggested by Hashimoto et al. (1992).

Fig. 4 shows selected anthropogenically derived element concentrations of As and V in the atmospheric particulate matter collected across Japan from 1974 to 1996. Concentrations of As, mainly derived from smelting and fuel combustion, declined rapidly in Niigata and Ohmuta. Mean concentrations of As ranged between 26–1.3, 16–0.92, 13–2.2 and 3.8–0.69 ng m^{-3}, in industrial, large, small and baseline cities, respectively. Concentrations of V, emitted from oil combustion, decreased in both industrial and large cities clearly. Mean concentrations of V varied between 32–2.6, 31–1.4, 12–1.9 and 6.8–1.4 ng m^{-3}, in industrial, large and small cities, and baseline sites, respectively as shown in Fig. 4.

Concentrations of Pb, derived principally from the combustion of petrol containing lead additivities and from smelting, decreased in both industrial and large cities clearly and varied between 390–22, 400–12 , 140–13 and 60–8.6 ng m^{-3}, in industrial, large and small cities, and baseline sites, respectively as shown in Fig. 5. However, concentrations of Zn, originated from coal combustion, smelting operations, incineration and wood combustion and from the wearing down of vehicle tyres, did not display any clear inclination in large cities and ranged between 510–66, 480–41, 520–30 and 61–26 ng m^{-3}, in industrial, large and small cities, and baseline sites, respectively, as shown in Fig. 5.

Concentrations of Na, assumed to be entirely marine derived and crustal derived slightly decreased and ranged between 1400–200, 1200–260, 1100–180 and 770–170 ng m^{-3}, in industrial, large and small cities, and baseline sites, respectively, as shown in Fig. 6.

The decline in the mean concentrations of elements is presented in Table 3 by comparing the average of the first two years (1974–1976) of the NASN data with that of the last two years (1994–1996). The ratios in Table 3 show the reduction from 1974 to 1976 levels. The reduction ratios of concentrations for selected elements such as Al, Fe and Mn ranged between 0.20 and 0.73 in industrial, large and small cities of Japan. This can probably be explained by reductions of fly-ash emissions from industrial and the gradual switch from coal to oil, gas and electricity as a domestic energy source for space heating. On the other hand, concentrations of both Al and Fe in Tokyo also did not show any decline.

The reduction rates of concentrations varied between 0.20 and 0.89 for V, Pb and Zn in industrial, large and small cities of Japan. This decline can probably be attributed to reductions in industrial and domestic emissions over the last 23 yr. However, concentrations of Zn did not show any declining either in Matsue or in Sapporo.

3.3. *Seasonal variation*

As can be seen in Fig. 7, monthly average concentrations of crustal elements for the 8 yr period from 1989 through 1996 as represented by Al for eight selected cities displayed high concentrations in March to May (the commonly known Dust season or Kosa Period) and low concentrations in July and August (summer). As mentioned before, the high concentrations for crustal elements in March to May were attributed to the presence of dust storms that occurred in the arid regions in central Asia. The soil and/or mineral particulate matter is lifted from these arid regions and transported to Japan by westerly winds. However, there are some exceptions for some elements and cities. For example, the seasonal average concentration of Fe observed in summer (1133 ng m^{-3}) is slightly higher than that in spring (1060 ng m^{-3}) in Kawasaki during an 8 yr period (1989–1996). This case can be explained by the fact that the steel industry has strong influence on the elemental pattern of Fe particularly in Kawasaki.

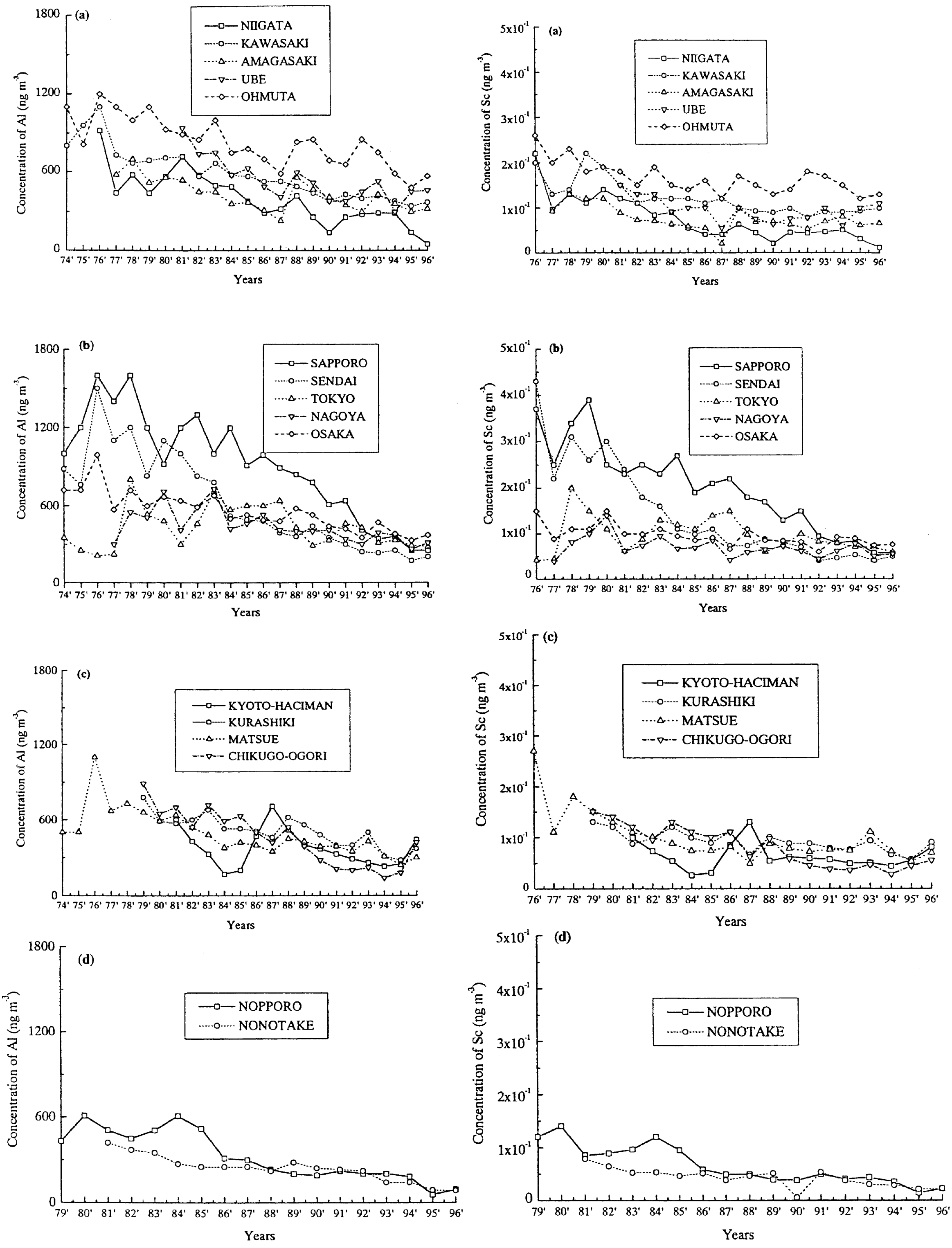


Fig. 2. Variation of annual average concentrations of Al and Sc, in industrial cities (a), in large cities (b), in small cities (c), in baseline cities (d), for 23 yr from 1974 to 1966.

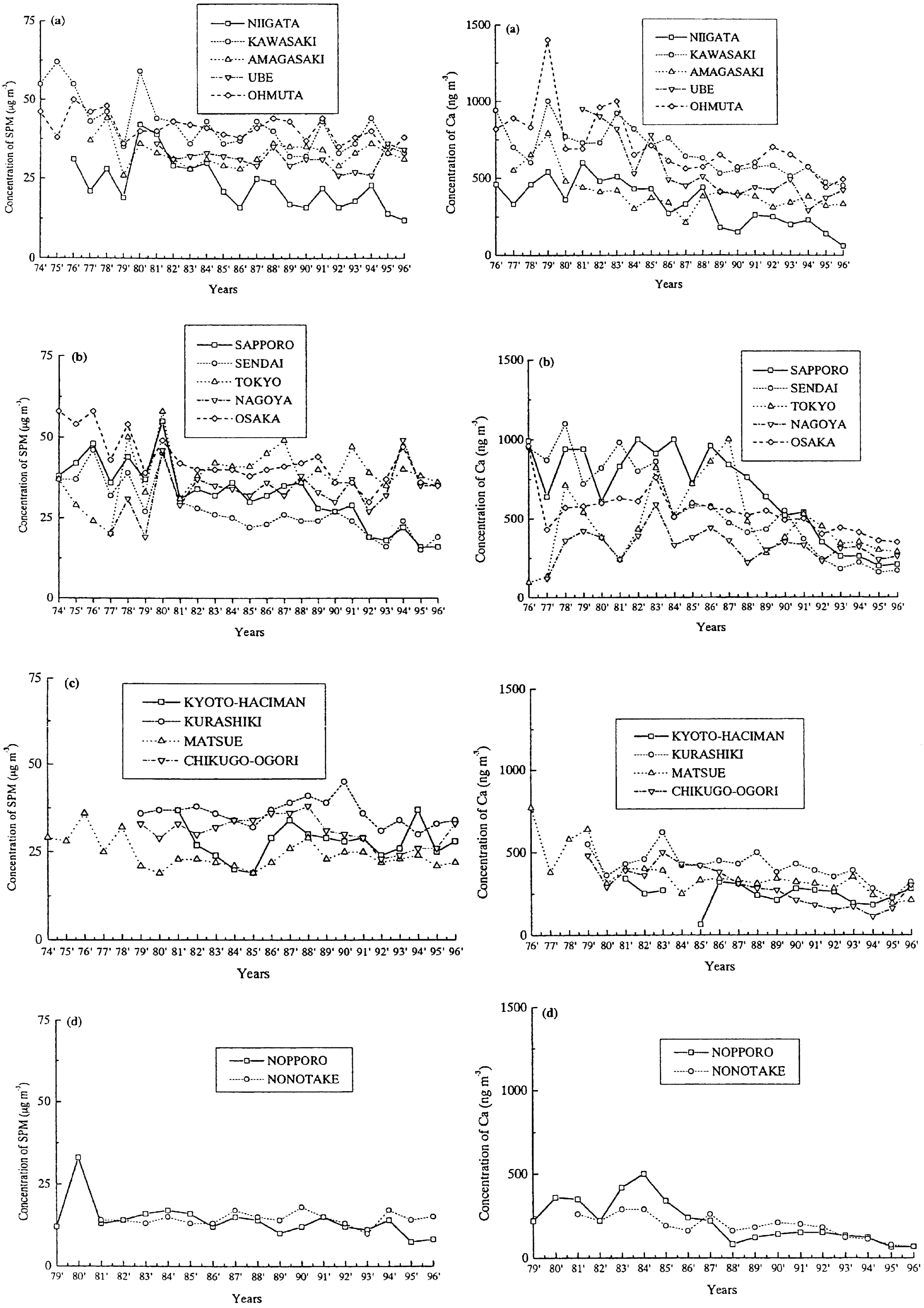


Fig. 3. Variation of annual average concentrations of SPM and Ca, in industrial cities (a), in large cities (b), in small cities (c), in baseline cities (d), for 23 yr from 1974 to 1996.

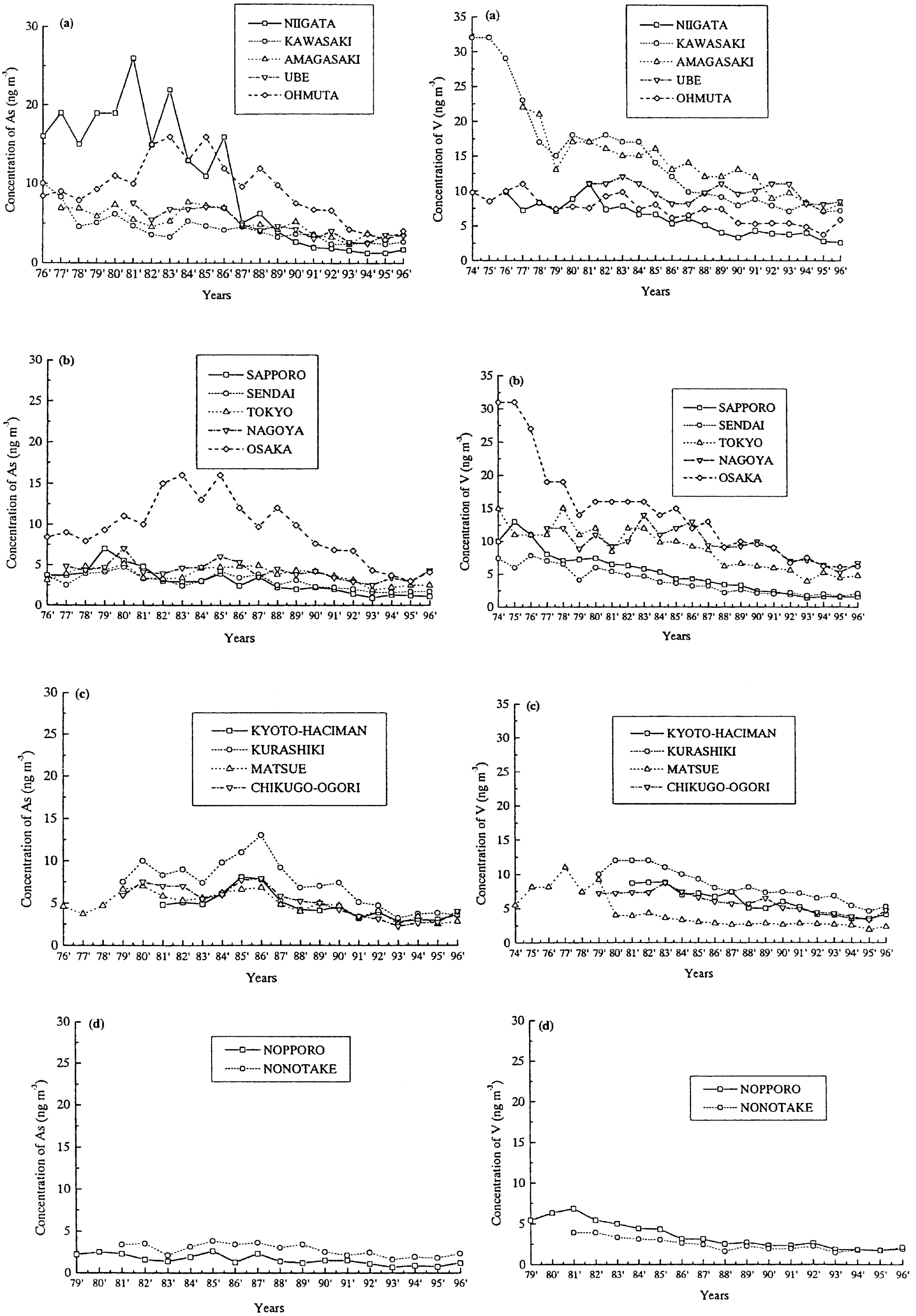


Fig. 4. Variation of annual average concentrations of As and V, in industrial cities (a), in large cities (b), in small cities (c), in baseline cities (d), for 23 yr from 1974 to 1996.

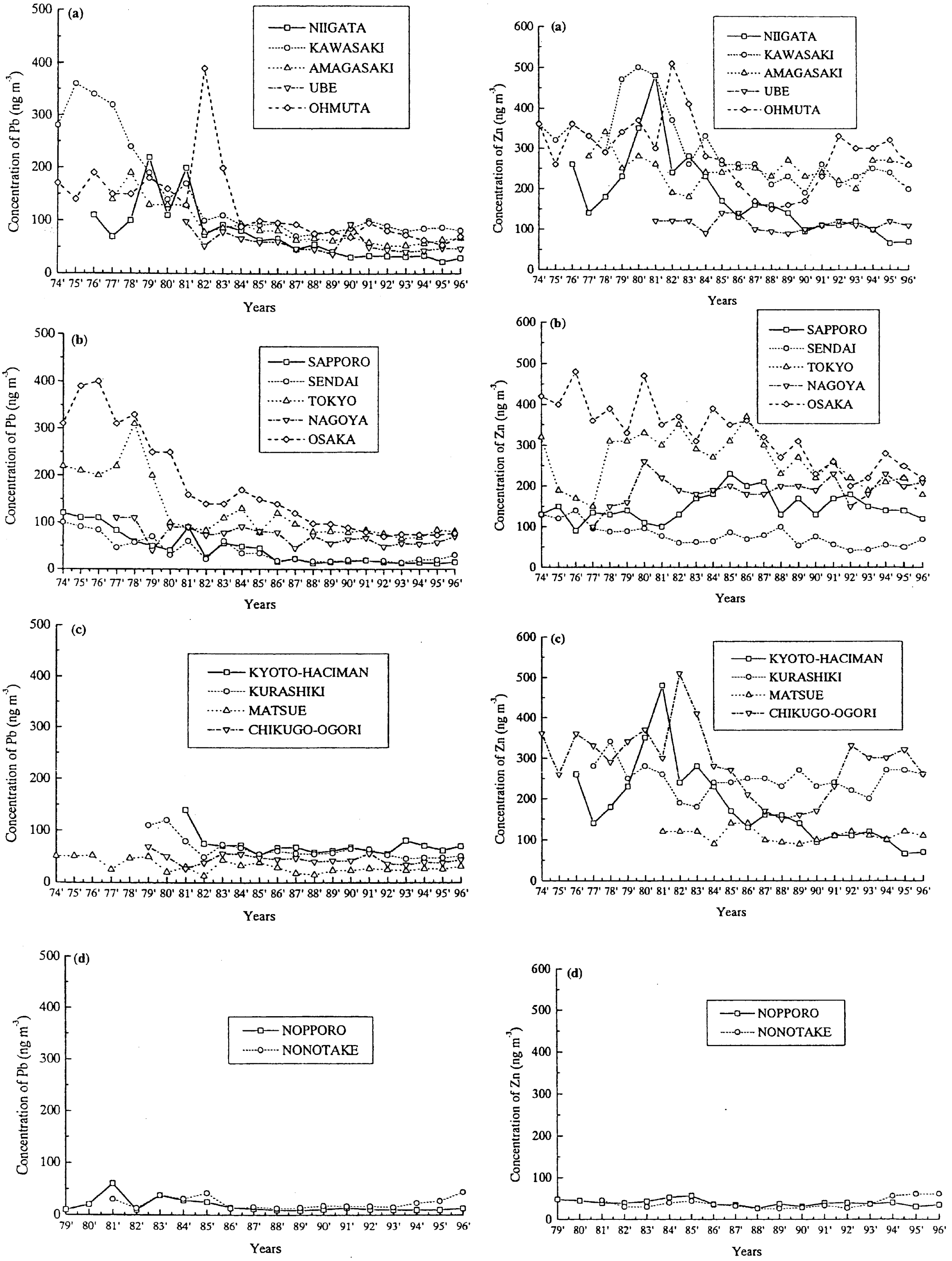


Fig. 5. Variation of annual average concentrations of Pb and Zn, in industrial cities (a), in large cities (b), in small cities (c), in baseline cities (d), for 23 yr from 1974 to 1996.

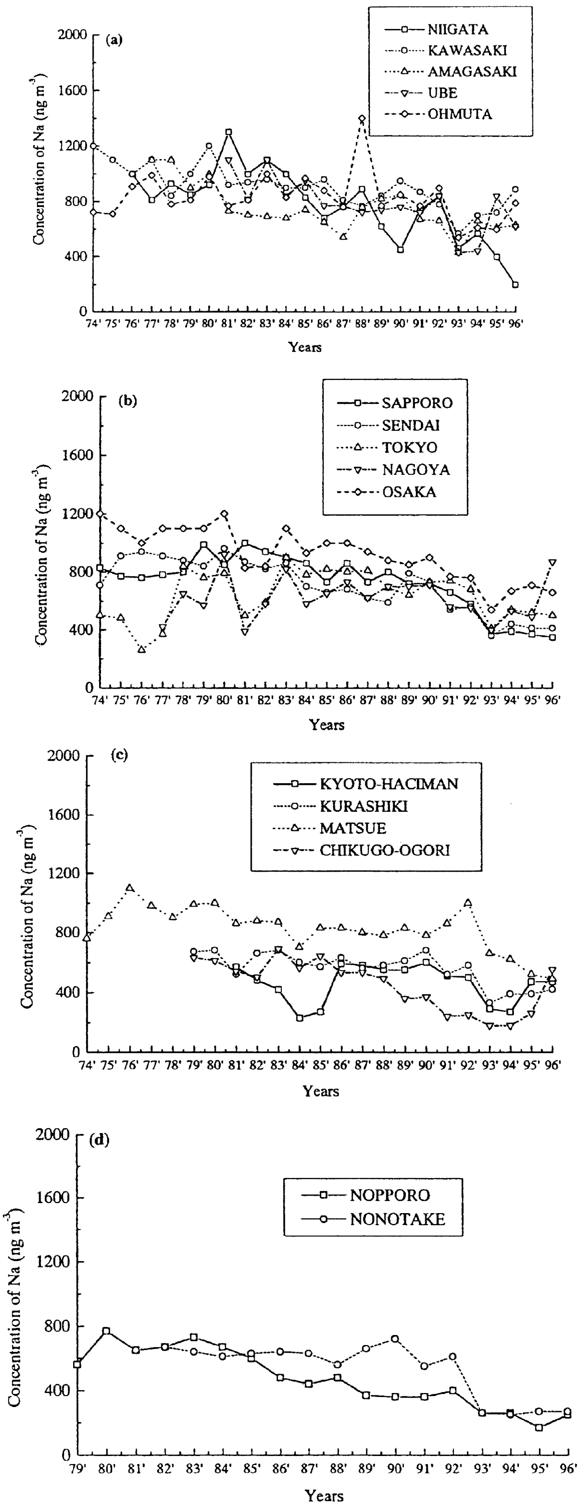


Fig. 6. Variation of annual average concentration of Na, in industrial cities (a), in large cities (b), in small cities (c), in baseline cities (d), for 23 yr from 1974 to 1996.

In winter (November and December), c rustal elements steadily showed higher concentrations than in summer except for some industrial cities such as Kawasaki and Niigata. Either a local source makes more contribution to concentrations of crustal elements during wintertime or meteorological conditions show highly stable conditions in those cities. Consequently, seasonal average concentrations of some crustal elements such as Al, Fe and Sc are higher constantly in winter than in summer.

On the other side, monthly average concentrations of some of the anthropogenic elements such as As, Zn and Pb for the 8 yr period from 1989 through 1996 as represented by As (Fig. 8) for selected cities showed higher concentrations in March to May (spring) than in July and August (summer). However, elements such as Cr, Cu, Ni and V displayed higher concentrations during summer time than during the Kosa period at some cities.

Seasonal average concentrations and the elemental average concentration ratio in the dust season and in winter relative to those in summer are given in Table 4 for selected locations and elements. As mentioned earlier, each value Zn has been averaged for each month for 8 yr (1989–1996). As seen in Table 4, the ratios of elemental average concentrations in the Dust Season to those in summer were 2–5 times higher and the ratios of elemental concentration in winter to summer were 1.5–3 times for crustally derived elements generally. This fact provides a very clear evidence that the crustally derived elements are brought from the Asian region to Japan during the Kosa period. In particular the concentration of Sc in Matsue, which originates from the soil, was 8 times higher during the Kosa period than during the summer period.

It is interesting to note that the maximum ratios of elemental average concentrations were found in Ohmuta, Matsue, Chikugo-Ogori and Ube, respectively. These cities were located on the western coast of Japan. In other words, they are under direct influence of long range transport of crustal elements from the Asian region during the Kosa period. This finding is in good agreement with the previous result reported by Tanaka et al. (1983). The work indicated that Kosa dust strongly affects the atmospheric composition at the Ube, Matsue and Kyushu areas including Ohmuta and Chikugo-Ogori, depositing 1.5–2.3 t km^{-2} month^{-1} of crustal elements.

The ratios of elemental concentrations in the dust season to those in summer were 1.6–3.9 times higher for some anthropogenically derived elements such as As, Pb, and Zn in Matsue, Ohmuta, Chikugo-Ogori and Ube, respectively. On the other hand, concentrations of elements such as Cr, Ni and V were slightly higher in dust season compared to those in summer and did not show any significant difference between winter and

Table 3
Comparison of average concentrations of six elements at industrial, large and small cities in Japan between the past period from 1974 to 1976 and the recent period from 1994 to 1996

Cities	Al			Fe			Mn		
	Concentration (ng m^{-3}) A	Concentration (ng m^{-3}) B	Ratio B/A	Concentration (ng m^{-3}) A	Concentration (ng m^{-3}) B	Ratio B/A	Concentration (ng m^{-3}) A	Concentration (ng m^{-3}) B	Ratio B/A
Kawasaki	953	363	0.38	1667	1067	0.64	133.3	35	0.26
Ohmuta	1037	547	0.53	1020	590	0.58	41	14.7	0.36
Sapporo	1267	287	0.23	770	257	0.33	24.6	6.7	0.27
Sendai	1047	207	0.20	1023	233	0.23	32	6.4	0.20
Tokyo	270	300	1.11	423	660	1.56	32.3	23.7	0.73
Osaka	810	360	0.44	1100	813	0.74	71.7	19.7	0.41
Matsue	700	300	0.43	607	257	0.42	18	10.1	0.56
	V			Pb			Zn		
Kawasaki	31	7.53	0.24	326.7	84.3	0.26	347	230	0.66
Ohmuta	9.4	4.87	0.52	166.7	62.3	0.37	327	293	0.89
Sapporo	11.3	1.57	0.14	113.3	13.7	0.12	123	133	1.08
Sendai	7.1	1.9	0.27	91.3	23.7	0.26	130	58	0.45
Tokyo	12.3	4.8	0.39	210	77.7	0.37	227	203	0.89
Osaka	29.7	6.17	0.21	366.7	74.7	0.20	433	250	0.58
Matsue	7.2	2.23	0.31	51.3	28	0.55	39	43	1.10

A: Average concentration from 1974 to 1976.
B: Average concentration from 1994 to 1996.

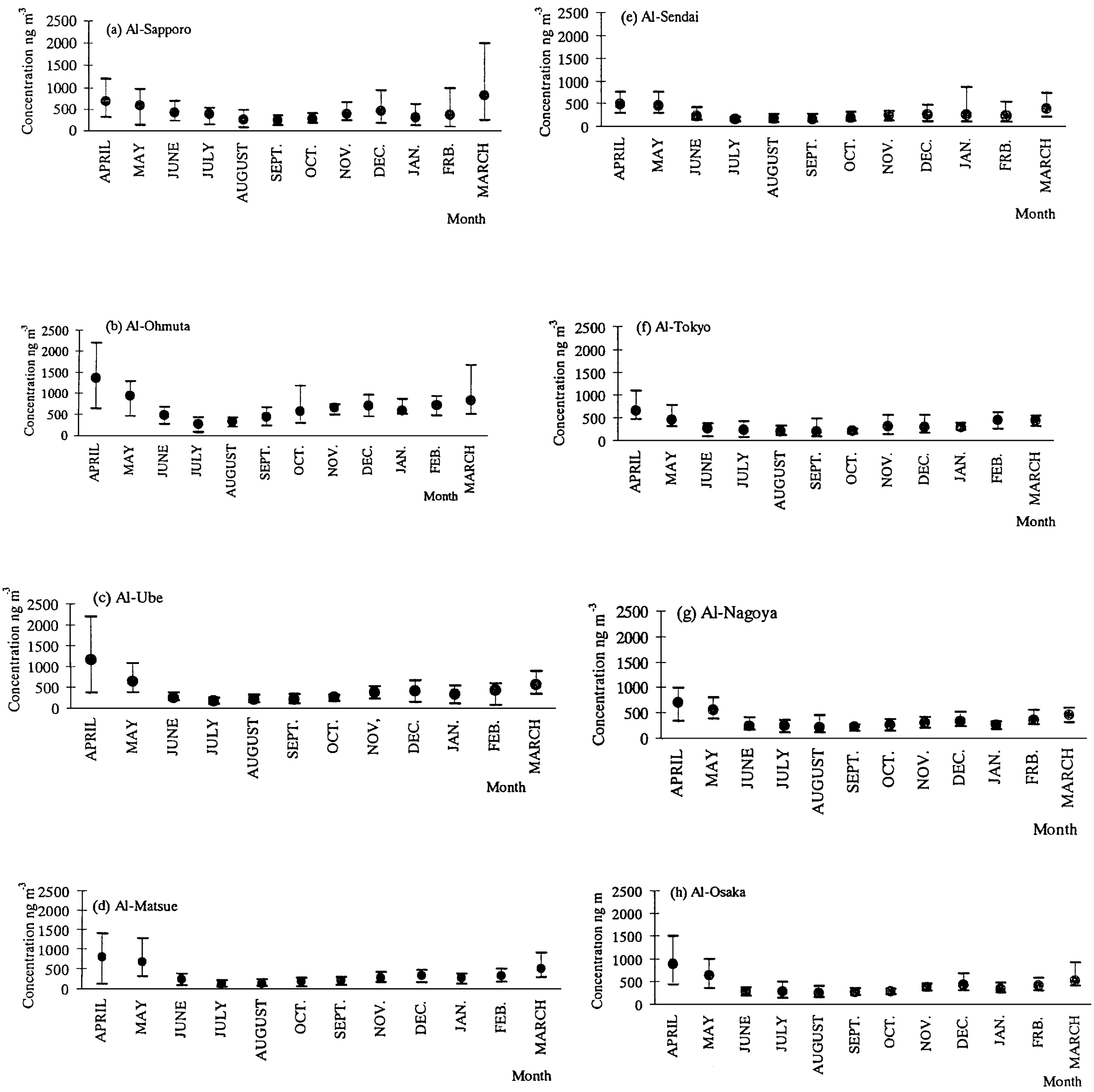


Fig. 7. Monthly variation on the average concentrations of Al for eight selected cities between April 1989 and March 1996.

summer. Furthermore, the ratios of elemental concentration of some of them such as As, Pb, and Zn were 1.6–5.7 times higher in winter than in summer for these four locations. Particularly, concentration of Zn in Chikugo-Ogori was 5.7 times higher in winter than in summer time. The reason for this fact may be that transport of anthropogenic elements such as As, Pb and Zn from the Asian continent to these four cities located at the western part of Japan is due to westerly winds which continuously occur in winter. Therefore, the high concentrations of these anthropogenic elements were recorded not only in the Dust Season but also in winter.

4. Conclusions

As the results of National Air Surveillance Network (NASN) data from 1974 to 1996 for metals in the atmospheric particulate matter over Japan, it was found that annual average concentrations of selected crustal and anthropogenic elements decreased

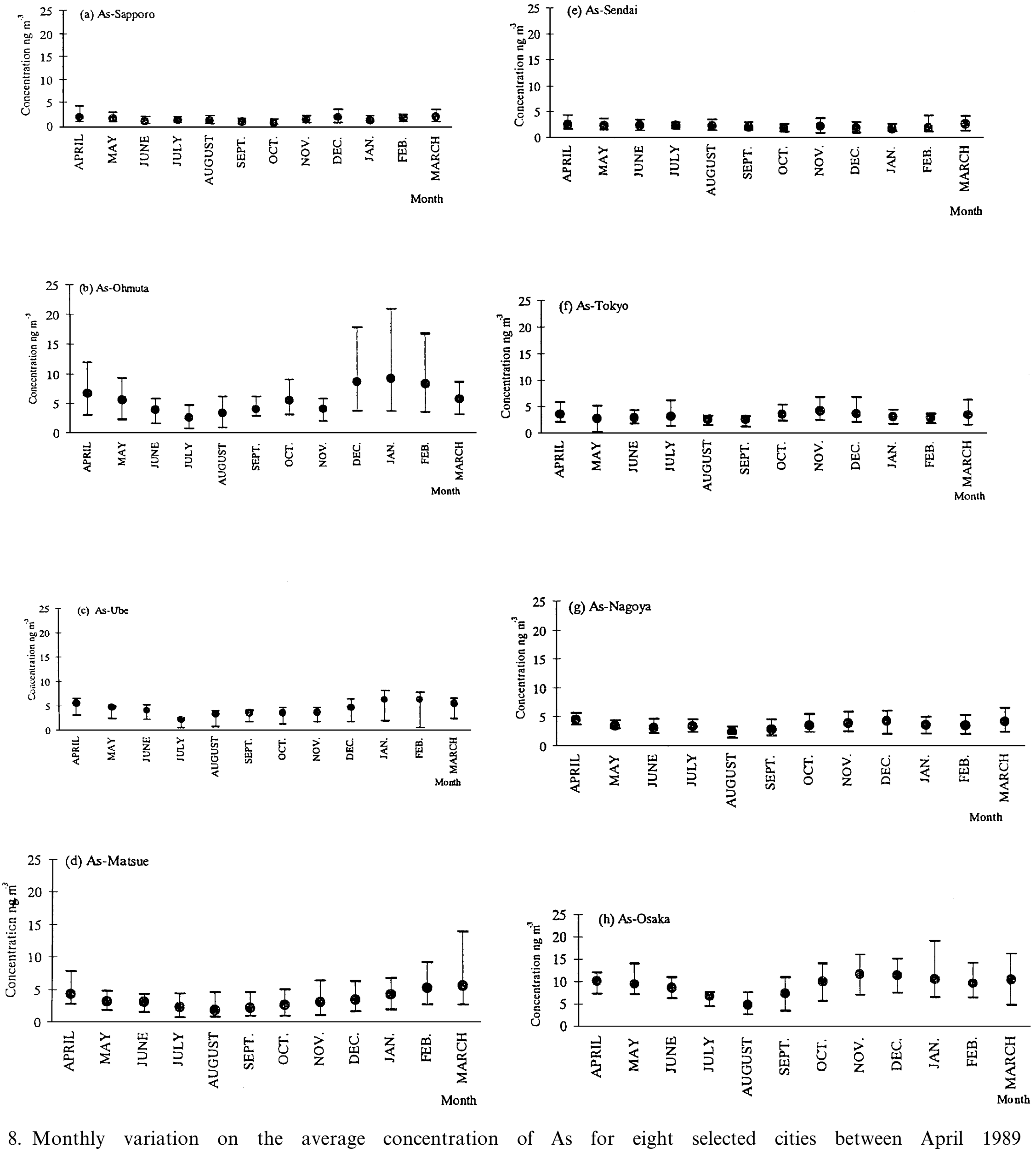


Fig. 8. Monthly variation on the average concentration of As for eight selected cities between April 1989 and March 1996.

substantially from 1974 to 1996. On the other hand, concentration of marine element reduced slightly between 1974 and 1996.

Crustal elements such as Al, Fe, and Sc displayed high concentrations in Dust Season (March–May) and in winter (November and December) and low concentrations in summer (July and August). Anthropogenic elements such as As, Pb, and Zn also showed high concentrations in the Kosa Season and in winter and low concentrations in summer. It was found that these elements were strongly influenced by their transport from the Asian continent to Japan by westerly winds. Only for summer, the westerly winds were replaced by the oceanic wind that dilute both crustal and some anthropogenic elements and therefore their concentrations were significantly low in Japan. However, anthropogenic elements such as Cr, Ni, and V did not show any significant seasonal concentration variation.

Table 4

Seasonal average concentrations of elements calculated from monthly averages and average concentration ratios in the Kosa season (spring) and in winter relative to those in summer between 1989 and 1996, Japan

Element	City	Concentration (ng m^{-3}) Spring, March–May	Concentration (ng m^{-3}) Summer, July and August	Concentration (ng m^{-3}) Winter, November and December	Conc.ratio $C_{\mathrm{Spr.}}/C_{\mathrm{Sum.}}$	Conc.ratio $C_{\mathrm{Win.}}/C_{\mathrm{Sum.}}$
Al	Matsue	665.0 ± 343.0 ($n = 24$)	189.3 ± 94.1 ($n = 15$)	302.0 ± 103 ($n = 15$)	3.51	1.59
	Ube	797.8 ± 462.1 ($n = 23$)	230.0 ± 62.9 ($n = 16$)	412.0 ± 145.7 ($n = 15$)	3.47	1.79
	Chikugo-O.	451.8 ± 332.9 ($n = 22$)	150.7 ± 75.1 ($n = 16$)	212.2 ± 140.1 ($n = 16$)	2.99	1.41
	Ohmuta	1060.0 ± 511.7 ($n = 24$)	393.2 ± 176.1 ($n = 15$)	695.6 ± 140.87 ($n = 15$)	2.69	1.77
Sc	Matsue	0.16 ± 0.10 ($n = 24$)	0.02 ± 0.01 ($n = 16$)	0.06 ± 0.03 ($n = 15$)	8.00	3.00
	Ube	0.17 ± 0.09 ($n = 23$)	0.04 ± 0.01 ($n = 15$)	0.08 ± 0.03 ($n = 15$)	4.25	2.00
	Chikugo-O.	0.08 ± 0.06 ($n = 22$)	0.02 ± 0.01 ($n = 14$)	0.04 ± 0.03 ($n = 15$)	4.00	2.00
	Ohmuta	0.22 ± 0.10 ($n = 24$)	0.06 ± 0.02 ($n = 15$)	0.16 ± 0.06 ($n = 16$)	3.67	2.67
Fe	Matsue	572.9 ± 371.6 ($n = 24$)	116.8 ± 30.0 ($n = 14$)	247.3 ± 82.4 ($n = 15$)	4.90	2.12
	Ube	725.0 ± 964.9 ($n = 23$)	254.0 ± 84.6 ($n = 15$)	404.7 ± 188.4 ($n = 15$)	2.85	1.59
	Chikugo-O.	292.9 ± 160.6 ($n = 21$)	97.9 ± 52.1 ($n = 13$)	216 ± 115.6 ($n = 12$)	2.99	2.21
	Ohmuta	853.3 ± 313.5 ($n = 24$)	246.7 ± 79.2 ($n = 15$)	730.0 ± 151.2 ($n = 15$)	3.46	2.96
As	Matsue	4.36 ± 2.54 ($n = 24$)	2.18 ± 1.22 ($n = 16$)	3.40 ± 1.60 ($n = 15$)	2.00	1.56
	Ube	4.28 ± 1.27 ($n = 23$)	2.22 ± 0.90 ($n = 15$)	3.58 ± 1.29 ($n = 15$)	1.93	1.61
	Chikugo-O.	4.13 ± 1.61 ($n = 22$)	2.10 ± 1.32 ($n = 15$)	3.71 ± 2.23 ($n = 16$)	1.97	1.77
	Ohmuta	6.08 ± 2.52 ($n = 24$)	3.01 ± 1.90 ($n = 15$)	6.44 ± 2.23 ($n = 16$)	2.02	2.14
Pb	Matsue	32.0 ± 8.71 ($n = 24$)	15.2 ± 3.76 ($n = 15$)	29.3 ± 6.82 ($n = 15$)	2.10	1.92
	Ube	52.5 ± 13.4 ($n = 24$)	33.1 ± 23.2 ($n = 15$)	85.8 ± 61.6 ($n = 15$)	1.59	2.59
	Chikugo-O.	43.7 ± 8.19 ($n = 24$)	24.8 ± 7.28 ($n = 15$)	58.4 ± 23.2 ($n = 16$)	1.76	2.35
	Ohmuta	73.6 ± 16.1 ($n = 24$)	29.3 ± 11.0 ($n = 15$)	104.9 ± 50.1 ($n = 16$)	2.51	3.58
Zn	Matsue	49.75 ± 14.46 ($n = 24$)	29.0 ± 5.12 ($n = 15$)	47.9 ± 15.44 ($n = 15$)	1.72	1.65
	Ube	133.4 ± 57.1 ($n = 23$)	79.8 ± 34.9 ($n = 15$)	130.5 ± 41.9 ($n = 15$)	1.67	1.64
	Chikugo-O.	183.7 ± 144.3 ($n = 22$)	47.4 ± 9.55 ($n = 15$)	270.9 ± 212.0 ($n = 14$)	3.88	5.72
	Ohmuta	245.2 ± 89.06 ($n = 24$)	97.8 ± 43.0 ($n = 15$)	360.0 ± 104.1 ($n = 14$)	2.51	3.68
Cr	Matsue	2.73 ± 1.22 ($n = 21$)	3.17 ± 2.28 ($n = 10$)	3.10 ± 1.82 ($n = 12$)	0.86	0.98
	Ube	4.46 ± 4.14 ($n = 18$)	3.21 ± 2.26 ($n = 11$)	3.73 ± 1.72 ($n = 10$)	1.39	1.16
	Chikugo-O.	4.18 ± 2.21 ($n = 20$)	2.30 ± 0.92 ($n = 9$)	4.13 ± 1.82 ($n = 11$)	1.82	1.79
	Ohmuta	3.92 ± 2.52 ($n = 24$)	4.60 ± 2.77 ($n = 14$)	4.74 ± 2.98 ($n = 16$)	0.85	1.03
Ni	Matsue	2.80 ± 1.01 ($n = 22$)	1.87 ± 0.77 ($n = 14$)	2.21 ± 0.55 ($n = 13$)	1.49	1.18
	Ube	8.30 ± 2.08 ($n = 23$)	7.70 ± 4.53 ($n = 15$)	7.92 ± 2.03 ($n = 15$)	1.08	1.03
	Chikugo-O.	2.76 ± 0.78 ($n = 22$)	1.89 ± 1.21 ($n = 13$)	3.21 ± 1.50 ($n = 15$)	1.46	1.69
	Ohmuta	4.46 ± 1.05 ($n = 24$)	3.37 ± 2.07 ($n = 14$)	4.69 ± 1.26 ($n = 16$)	1.32	1.39
V	Matsue	3.16 ± 0.73 ($n = 24$)	2.40 ± 0.45 ($n = 16$)	2.44 ± 0.63 ($n = 15$)	1.32	1.02
	Ube	11.50 ± 3.75 ($n = 23$)	10.92 ± 3.18 ($n = 15$)	9.10 ± 2.90 ($n = 15$)	1.05	0.83
	Chikugo-O.	4.86 ± 1.30 ($n = 22$)	3.50 ± 1.09 ($n = 15$)	5.88 ± 2.43 ($n = 16$)	1.39	1.68
	Ohmuta	5.34 ± 1.14 ($n = 24$)	4.99 ± 3.60 ($n = 15$)	5.98 ± 1.24 ($n = 16$)	1.07	1.19

Note: n indicates the number of data considered while the evaluation.

Acknowledgements

Authors wish to thank the Japan Society for the Promotion of Science. The first author being a JSPS fellow was supported generously by JSPS foundation during the research. This study was supported by funding from the Core Research for Evolutional Science and Technology (CREST) program by Japan Science and Technology Corporation (JST). The authors thank T. Ootoshi and T. Nedu of Japan Environmental Sanitation Center in offering the NASN.

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