Osaka Prefecture University

Bull. Univ. Osaka Pref., Ser. B, Vol. 33,
大阪府立大学紀要, 農学・生物学, 1981

Heavy Metal Contamination in Urban Soils I
Zinc Accumulation Phenomenon in Urban Environments as Clues of Study

Yutaka KOMAI

Laboratory of Soil Science and Plant Nutrition, College of Agriculture

(Received October 31, 1980)

Abstract

As an introduction of the continuing study on the heavy metal contamination in urban soils, zinc accumulation phenomenon observed in urban areas in south Osaka was reported. The survey of zinc concentration in soybean leaves taken in urban and suburban arable lands indicated its accumulation in a wide area. And a correlation between easy soluble zinc level in soils and leaf zinc content were shown. Zinc concentrations in suspended particles in air, falling dust and some water samples were checked, and the high levels of zinc were found also in them. In several city parks, significant higher concentrations of zinc and lead in surface than subsurface soils were found. The ratios of easily soluble to total zinc were markedly high in urban soils than rural soils. In Sakai, at sandy fields near the coast of Osaka Bay, where the rotation culture of vegetables prevailed, zinc accumulation in soils was notable among heavy metals. Where the renewal of sand as the vegetable field had been made at intervals, there was a significant increase in zinc contents from new sand to older sand materials. From the above data, it was considered that the zinc accumulation observed in the urban areas was a wide phenomenon through atmospheric pollution caused by zinc emission from various sources. The probability of emission sources of heavy metals in urban environments was discussed. With respect to the zinc contaminants, emissions from the melting of scrap in iron and steel producing factories, the wear of rubber tire occurred with motorization and the incineration of municipal wastes are regarded as the main sources.

Introduction

The so-called urban problem before the second world war had a meaning of social ills such as poverty and unemployment, however, after the rapid development of cities caused by recent industrialization the conservation of environmental quality has become an important problem in urban areas. As to urban environmental pollutions, the qualities of atmosphere and aquatic system have attracted public attention because of their direct and rapid influences. But in the understanding of urban environmental problems, it is necessary to consider the significance of urban soil.

The exposed soil area in cities has been markedly reduced due to the progress of urbanization, and the disappearance of green space has been a matter of concern. In current city plannings, the necessity of reserves for parks and other green areas is generally recognized. These areas not only provide places for citizens' recreation and give a psychologically pleasant effect through the maintenance of greens, but have a great physical effect in buffering the urban climate change. On the other hand, from the viewpoint of environmental pollution in urban areas, the capacity of soil to accumulate heavy metals functions as an indicator which provides information on long-term environmental quality. Heavy metal concentrations in air and water have wide variations at different times, and many determinations are necessary for average data; however, soil pollution may be con-

NII-Electronic Library Service

Osaka Prefecture University

sidered to have an integral nature, as it were.

The recent development of motorization has brought a notable source of heavy metal contamination into urban environments, and the enrichment of metals in soil-plant systems along roads with heavy traffic has attracted attention. YOSHIDA *et al.*[1], SHIMADA *et al.*[2] and ABE *et al.*[3] reported the contamination with lead and zinc, *etc.* in roadside soils and street trees. Also in foreign countries, lead pollution in roadside sites has been indicated. On the other hand, there are various instances of heavy metal pollution caused by emissions from specific and stationary sources such as metal producing factories and waste incinerating plants. However, there are not so many published reports on investigations of them. City parks and urban arable lands laid out scatteredly in most cities are considered to be affected by many and unspecific emission sources. Accordingly they are suitable sites for the general investigation of heavy metal contamination in urban soils. But the reports on them are very few, and the short papers by PURVES *et al.*[4,5,6] are the instances.

Recently, the author has investigated the behavior of heavy metals in the soil-plant systems of parks and arable lands, in many cities such as Osaka, Sakai, Amagasaki and Kishiwada. The contamination of heavy metals in urban soils in large cities is regarded as a very wide phenomenon brought through various kinds of atmospheric pollution. The sources of emission of heavy metals are very complex, and the estimation of their extent of participation in pollution is rather difficult. The object of our research, however, is not to explain local problems caused by some specific sources, but to elucidate the relationship between heavy metal contamination in urban environments and the general features of the cities, such as population density, industrial output, structure and plant scale of industry, road development and traffic volume, *etc.*

In this paper, the zinc accumulation phenomenon found in urban areas in earlier years are summerized as clues of the study. Determinations of metals were made by X-ray fluorescence and atomic absorption analyses. The author are grateful to Prof. M. YAMAGUCHI for his continuing interest and support and Messrs. Y. HARIMA and K. HAYAMA for their assistance in some experimental works.

A survey of zinc concentration in plant and soil samples in south Osaka

This study started when an investigation of the chemical constituents of various plant samples taken from the Daisen Campus of the University of Osaka Prefecture was carried out with X-ray fluorescence analysis in 1967. It was before heavy metal problems in urban environments had attracted much attention. It was found that many samples contained a large amount of zinc compared with the values in the literatures. Accordingly, a wider survey of zinc concentration in plants and soils in southern Osaka Prefecture was made in 1968 using soybean leaves as an indicator.

Twenty-two samples, collected from arable lands distributed in urban areas around the midtown area in Sakai City, contained 300 ppm of zinc on average and the maximum of 470 ppm. Furthermore, the zinc concentration in the samples, taken in a radiating pattern from the above site towards the borders of the prefectural area, suggested that the accumulation of zinc in plants was a wider phenomenon than we had anticipated. The zinc level in soybean leaves at the border area, however, approached those in the literatures, below 100 ppm. Figures 1 and 2 show the locations of sampling points and the zinc concentrations in samples taken at the radiate points, respectively. Samples in the southwestern direction, that is, in the coastal zone, showed somewhat higher levels than

NII-Electronic Library Service

Osaka Prefecture University

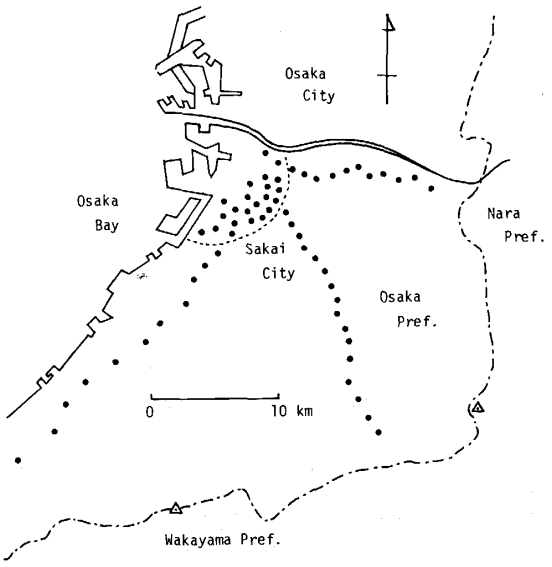

Fig. 1. Location of sampling points of soybean leaves.
Inside dotted line is urban area around midtown of Sakai City.

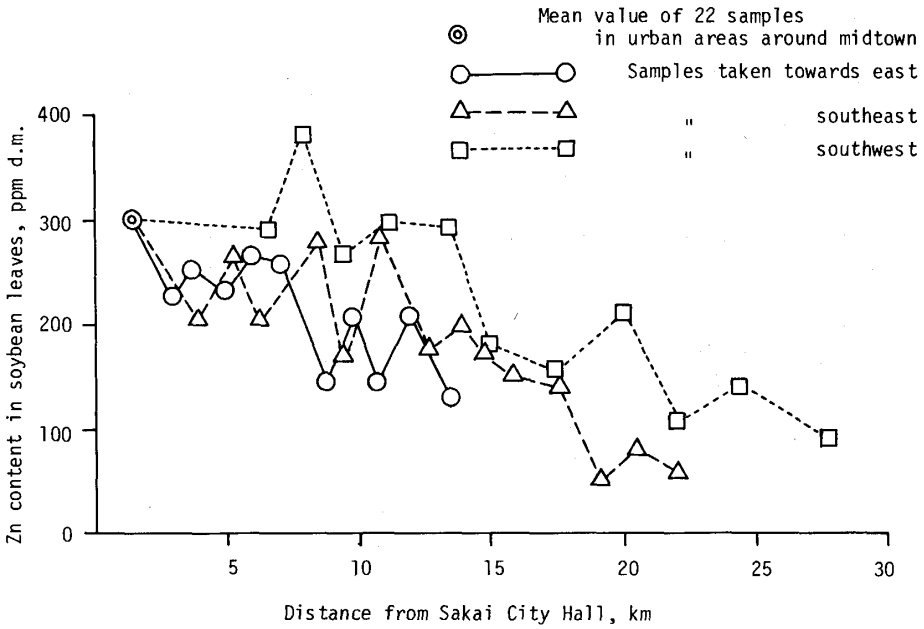

Fig. 2. Gradual decrease in zinc content in soybean leaves with distance from the center of urban area of Sakai.

those in other directions. Differences in the pH values and geological origin of the soils at sampling points of the above survey had not noticable relation to the variation of the zinc content of soybean leaves. But a correlation (r=0.751***) between the easy soluble zinc content in soils (extracted with 0.1N HCl) and the leaf zinc content were observed.

Simutaneously, a water culture test under regulated conditions on the effect of zinc addition on soybean growth was tried. Table 1 gives the result. Referring to the zinc concentration in leaves showing inhibited growth in this test, it is probable that soybean growth in the urban areas had suffered more or less from an excess of zinc. Also in

NII-Electronic Library Service

Osaka Prefecture University

Table 1. Water culture test of soybean added with zinc sulfate.

Added concentration of Zn in culture solution	Zn content in		Plant growth
	Leaves	Roots	
ppm	ppm d.m.		g d.m./pot
0	42	77	36.0
0.05	70	139	33.0
1	425	1860	15.7 (Injured)
3	564	2250	15.0 („)
10	508	2390	10.4 („)
30	467	18380	6.4 (Dead)

Two litter glass pot and Shive's nutrient solution were used in a regulated growth chamber. Growth period were 3 weeks before and 4 weeks after Zn addition.

another soil culture test, at a similar level of zinc content in the leaves, damage to growth was observed. A growth test on various vegetables under comparable conditions showed the zinc accumulation was more notable in leguminous and cruciferous plants.

Heavy metal concentrations in various environmental materials

With respect to the cause of zinc contamination in soil-plant system described in the preceding paragraph, heavy metal contents in suspended particles in air were checked. The zinc levels were actually high in them (Table 2). Referring to an official material[7], such contamination of zinc in aerosol was considered to be a wide phenomenon observed not only in Sakai but in other large cities in Japan (Table 3). Samples of falling dust

Table 2. Heavy metal concentrations in atmosphere of Sakai and Osaka.

$\mu g/m^3$

	Sakai City*		Osaka City**
	(Feb.)	(Nov.)	(av. in a year)
Suspended particles	307	204	298
Cu	0.23	0.04	0.09
Fe	11.4	1.79	5.3
Mn	0.37	0.09	0.40
Ni	–	–	0.09
Pb	0.97	–	0.74
Zn	1.65	1.44	1.8

Samples were taken by high volume air samplers in 1968.
* Taken near Univ. of Osaka Pref.
** Data from reference 7).

Table 3. Zinc concentration in atmosphere of major cities.

$\mu g/m^3$

City	Zn concentration*	
	1968	1969
Sapporo	1.1	0.95
Tokyo	1.2	1.50
Kawasaki	2.4	2.39
Nagoya	–	1.92
Osaka	1.8	2.23
Amagasaki	2.0	1.94
Okayama	–	0.61
Ube	1.1	0.60
Kitakyushu	1.1	1.66

* Values on average in the year. Data from reference 7).

taken in Sakai were also examined. As shown in Table 4, in the nearer site to industrial area, the higher concentrations of metals in dust were found. And most of soluble zinc in them was water soluble form in contrast to iron and manganese, *etc.* Further, some water samples from the environment such as river, rain and tap water were tested, and the zinc levels in them were also significant among the heavy metals (Table 5). The sewage

NII-Electronic Library Service

Osaka Prefecture University

Table 4. Amount of heavy metals contained in precipitation and falling dust in Sakai.

	Site A		Site B		Site C	
	H_2O*	HCl**	H_2O	HCl	H_2O	HCl
Fe	10.3***	815	1.1	589	1.2	245
Mn	10.0	119	6.9	76	2.4	32.2
Zn	69.9	7.7	48.2	5.2	10.3	3.0
Cu	3.1	3.0	15.4	2.3	8.9	0.7
Pb	–	16.4	–	4.8	–	3.0

* Water soluble.
** 6N-HCl soluble.
*** mg/m^2/25 days (Jan.-Feb., 1968).
Site A : near northwestern industrial area.
Site B : central commercial area.
Site C : southeastern residential area.
Samples were collected by deposit gauges.

Table 5. Heavy metal concentrations in water samples in Sakai.

ppm

Sample		Fe	Mn	Zn	Cu
Rain water*					
Jan. 30**	Initial 2 hrs	0.02	0.01	0.16	0.09
	Middle 2 hrs	0.01>	0.01>	0.94	0.05
	Final 2 hrs	0.01>	0.05	0.19	0.02
Feb. 8		0.01>	0.01	0.10	0.04
River water					
Feb. 2 - 11	(av. of 4 determinations)				
	Yamato River	0.18	0.10	0.05	0.01
	Ishizu River	0.43	0.20	0.69	0.04
	Kudara River	0.87	0.21	0.45	0.04
Tap water*					
Feb. 2 - 11	(av. of 4 determinations)	0.01	0.04	0.25	0.04

* Sampled at Univ. of Osaka Prefecture.
** 1968

sludge from a municipal treetment plant also contained a large amount of zinc.

Heavy metal accumulation in park and farm soils

In several parks in both Sakai and Osaka and some rural areas the concentrations of five heavy metals in soils were investigated in 1970. Table 6 shows some of the results. Significantly higher levels of metal contents of zinc and lead in surface soils than subsurface soils were observed. The concentrations of total and easily soluble (0.1N HCl extractable) zinc in some park soils at different depths are given in Table 7. The ratios of easily soluble to total zinc in the soils of urban areas had a tendency to rise with the increase in total zinc, and the ratios in some rural areas used as controls were below 20%.

In Sakai, on sandy fields near the coast of Osaka Bay, rotation culture of soft leaf vegetables, such as spinach, garland chrysanthemum and Welsh onion has prevailed under

NII-Electronic Library Service

Osaka Prefecture University

Table 6. Distribution of heavy metals in surface and subsurface soils in parks and gardens.

Park*	Soil**	Fe	Mn	Zn	Cu	Pb
Sakai City						
Kinzei	Surface	221 (1.4)	210 (1.3)	951 (2.4)	119 (1.4)	122 (1.1)
	Sub.	155	165	398	88	115
Xavier	Surface	160 (1.0)	159 (0.7)	1060 (1.5)	108 (1.1)	189 (1.5)
	Sub.	162	221	703	96	125
Daisen	Surface	134 (0.7)	206 (0.6)	719 (7.3)	83 (1.0)	102 (1.3)
	Sub.	206	334	99	86	81
Nakamozu***	Surface	201 (1.0)	232 (1.2)	275 (1.9)	87 (1.0)	85 (1.1)
	Sub.	206	193	146	89	76
Osaka City						
Nozaki	Surface	123 (1.3)	186 (2.2)	1010 (7.1)	119 (1.3)	370 (4.4)
	Sub.	97	83	143	89	85
Ōgimachi	Surface	96 (1.4)	127 (1.2)	380 (4.3)	79 (0.9)	125 (1.4)
	Sub.	70	105	88	86	90
Control****						
Farm soil	Mixed	100	100	100	100	100

Determinations were made by X-ray fluorescence analysis.
Values were the indexes compared with control as 100.
Figures in parentheses show the ratios of surface to subsurface soils.
* Except with stars.
** Surface soil : 0-2 cm, subsurface soil : 3-5 cm in depth.
*** Campus of Univ. of Osaka Prefecture.
**** Farm soil in the above univ.

Table 7. Zinc concentration in park soils at different depths in Sakai.

Park	Depth of soil	Soil pH	Total Zn* in soil (A)	0.1 N-HCl soluble Zn (B)	Ratio (B)/(A)
	cm		ppm	ppm	%
Ōhama	0 - 1	5.6	696	438	63
	1 - 4	5.6	134	61	45
	4 - 7	6.0	98	40	41
Xavier	0 - 2	5.9	344	194	56
	2 - 5	5.9	500	297	59
	5 - 10	5.8	276	140	51
	10 - 15	6.7	191	74	39
	15 - 20	7.3	190	58	31
Shukuin	0 - 2	6.7	1070	662	64
	15 - 20	7.9	1960	1365	70
Kinzei	0 - 2	7.2	611	408	67
	15 - 20	8.1	202	69	34

* Determined by X-ray fluorescence analysis.

intensive management. The concentrations of five heavy metals in the soils at three representative sites in the above area, Sanbō, Minato and Ishizu, were determined in total and easily soluble forms in 1971. The result on zinc is shown in Fig. 3. At Sanbō, in the

NII-Electronic Library Service

Osaka Prefecture University

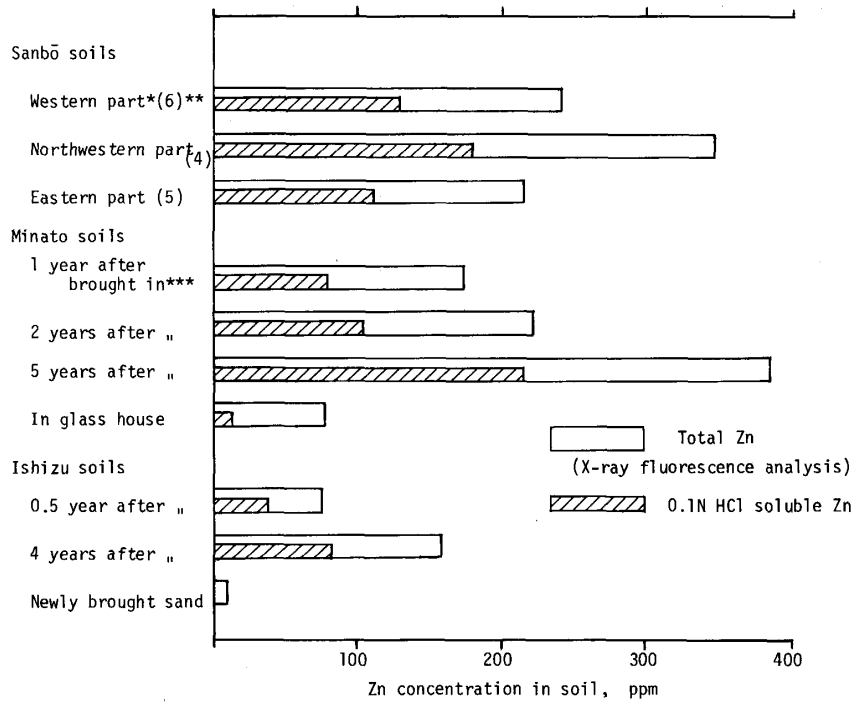

Fig. 3. Zinc concentration in vegetable farm soils in coastal area in Sakai.

* Directions from Sanbō Primary School.
** Number of sampling sites.
*** In these sites, new sand has been brought in as culturing field with intervals of 4 or 5 years.

northernmost part near Osaka, a higher level of metals in sandy soils was observed. At Minato and Ishizu, situated in the middle and southern parts of the area, respectively, new sand has been brought in to culture vegetables at intervals of four or five years, since vegetable growth on unchanged sand material declines with the lapse of time. There was a significant increase in zinc contents from new sand to older sand materials. A similar difference in zinc contents was also observed in vegetables grown there. The sand in vinyl greenhouses at Ishizu did not show an increase in its zinc contents so much as in open fields at Minato. These findings suggest that annual increments of the metal have probably been added through the air in this area. Also in the copper and lead contents in these samples, rather similar patterns were observed, but they were less clear than those of zinc, and the ratios of their easily soluble to total forms in soils were lower than zinc.

Discussion

The source of heavy metal pollutants in urban environments includes a wide variety, but may be divided into three main categories as follows: (1) factories concerning metal and chemical industries, (2) automobiles, (3) municipal wastes and their incinerating plants.

Among metal works, in the case of nonferrous metals, such as zinc and copper, their smelters are generally located near mines away from large cities, since their ores have been obtained more or less domestically. Factories producing iron and steel, however, are concentrated in the coastal areas of large cities with trade ports for the convenience of

NII-Electronic Library Service

Osaka Prefecture University

transportation of the raw materials and products. Without distinction between ferrous and nonferrous, secondary metal product works are primarily located near or in city areas. These factories may directly participate in the spread of heavy metals in urban environments through the emission of dust and aerosol. On the other hand, chemical works concerned in heavy metal pollution are galvaniziag and production of paints, chemicals, plastic stabilizers and batteries, *etc.* Although the main route of metal discharge from these chemical sources is drainage, emissions into the air also occur in the case of metals with low boiling points.

Automobiles are thought to be the mobile source of heavy metal contamination. In Japan, as the addition of 4-methyl lead to gasoline has been largely limited since 1973, the emission of lead to the environment has been reduced recently, but that emitted in the past still remains in large amounts in urban environments.

Municipal wastes from citizens' homes and industries also include heavy metals in various forms. They are contained as alloys, surface coatings, paints and stabilizers in solid wastes, and are often concentrated in sewage sludge. Besides direct land reclamation and burying in the soil, these wastes have been incinerated in high proportions, and the emission of metals into the air and the discharge into drainage or exudate taking place in the above processes cause contamination in urban soils.

Zinc accumulation phenomenon in urban environments of Sakai and its environ was shown in this paper as an introduction of the continuing study on the heavy metal contamination in urban soils. From the data, it is considered as a wide phenomenon through atmospheric pollution caused by zinc emission from various sources.

Concerning from where the zinc contaminants have been brought widely into these urban and suburban areas, many sources can be considered as described above. However, the participation of emission from scrap, which usually contains a large amount of zinc, used as a raw material in iron and steel production may be the most probable. The melting point of iron is 1535°C and the boiling point of zinc is 907°C, and when raw materials are melted down in electric or open hearth furnaces zinc in scrap easily volatilize as oxide. Zinc oxide has a very fine diameter (about a micron), and after emitted into air it will be suspended and spread until falling down with precipitation. It may be a reason why its contamination in the environment is a wide phenomenon. Of course, the countermeasure for its emission from factories has been taken properly, but may not be perfect. And most of iron and steel factories usually more or less make use of various scraps as a raw material. HAMAMURA *et al.*[8] reported the heavy metal concentrations in suspended particles, collected leeward of electric furnaces of a steel producing factory, and also in soils around the plant. They observed a marked accumulation of zinc in the samples.

Besides those from metal producing factories, the emissions from the wear of rubber tires and the combustion of lubricating oil, both which contain zinc as an additive, may have caused a certain part of the contamination due to recent development of motorization. Further, the emission from the incineration of municipal wastes also cannot be ignored.

There are many instances of metal pollution caused by the infiltration of waste drainage from factories. Although they have more concentrated influence compared with cases *via* air, they may be phenomena having more local nature.

As well known, national resources of Japan are very limited in various industries, but as to zinc ore we have fairly large amount of its domestic resource. Zinc metal production in our country ranks second in the world next to Soviet Union, and more than 40%

NII-Electronic Library Service

Osaka Prefecture University

of its raw materials are obtained domestically. This fact may have a relation to the observed contamination, and it is supposed that zinc contamination in urban environments is a common phenomenon extending over many large cities (*cf.* Table 3).

In the foregoing paragraphs, the nature of heavy metal contamination in soils as an indicator of environmental quality has been given attention. But, simultaneously, the behavior of metals in soils such as the change of chemical form with the lapse of time and the influence on living things in urban area are necessary to be considered. The data on them are being arranged for reports.

References

1) YOSHIDA, T. and YAMADA, T. (1971). Environmental pollution with lead. *Environ. Pollution and Japanese Sci.*, **2**, 23-26. (*in Japanese*)
2) SHIMADA, N., SUMIYISHI, M., TOYODA, S., SATO, Y. and KOJIMA, M. (1973). Contamination of roadside soil and vegetation with lead, zinc and cadmium. *Tech. Bull. Faculty of Horticulture, Chiba Univ.*, **21**, 36-52. (*in Japanese*)
3) ABE, Y., TAKAHASHI, Y. and SEKI., T. (1975). *Reports on the protection of street trees in Sendai City* (YOSHIOKA, K. ed.), 3-17. (*in Japanese*)
4) PURVES, D. (1967). Contamination of urban garden soils with copper, boron, and lead. *Plant and Soil*, **26**, 380-382.
5) PURVES, D. and MACKENZIE, E. J. (1969). Trace-element contamination of parklands in urban areas. *J. Soil Sci.*, **20**, 288-290.
6) PURVES, D. and MACKENZIE, E. J. (1970). Enhancement of trace-element content of cabbages grown in urban areas. *Plant and Soil*, **33**, 483-485.
7) *Prime Minister's Office* (1971, 1972). Kōgai Hakusho (Environmental Quality in Japan). (*in Japanese*)
8) HAMAMURA, N., ISHIDA, K., TOMITA, B., NOMA, M., SHIMIZU, Y., NAKAMURA, K., HAYAKAWA, M., WATANABE, Y. and TOYOSHIMA, K. (1972). On the air pollution by electric arc furnace steel making factory. *J. Japan Soc. Air Pollution*, 7, 13-18. (*in Japanese*)

NII-Electronic Library Service