Particulate lead as an indicator of impacts of human activity on surrounding environment in the Philippines

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フィリピンにおける都市活動の環境への影響指標としての粒子態鉛

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本研究では、環境ホルモン作用があり毒性のある鉛に注目し、鉛が底泥に蓄積されていく過程を明らかにし、鉛の空間分布から、都市活動によって排出される粒子状物質による汚染の範囲を推定し、鉛がその粒子状汚染物質の輸送経路を解明するのに有用な指標であるかを検証することを主な目的とした。さらに、鉛以外の重金属や希土類についても調査を行い、金属ごとの挙動の特徴を明らかにした。本研究の結果は、フィリピンのマニラ湾の流動解析に基づく粒子状物質の輸送モデルの検証にも役に立つと考えられる。

1. Introduction

In 1960s, Japanese had severe experiences of pollution caused by heavy metals such as Itai-Itai disease from cadmium and Minamata disease from mercury. Rapid growth of the economy and rapid industrialization can be seen not only in Japan but also in developing countries like the Philippines. Most developed areas in the Philippines are concentrated around Manila bay and Pasig River. The analysis of bottom sediments is essentially important for the study of the water environment in the Philippines, because they may reflect pollution especially caused by particulate matters.

The objectives of this study are to identify lead-pollution area from the spatial distribution of lead as one of endocrine disrupters and to estimate the pathway of particulate pollutants by lead concentration. This study also will help the validation of modeling transportation of particles in Manila Bay.

2. Methods

2.1 Sampling

37 bottom sediment samples (1～42) were collected in August, 2001 , 17 sediment samples (P1～P5, L1～L11) were collected in March, 2002. In addition, 5 dry-deposits at the side of roads were collected. The sampling locations in Manila Bay were chosen not to be affected so much by waves, excluding steep slope places. Velocity of the formation of the sediments is said to be 1 cm/y in Manila Bay [1].8 sedimetn samples were collected in 2002 in Tama river as shown in Fig.2 In 2003, soil samples were taken in northern part of Luzon Island as shown in Fig.3. The area was used as agricultural field.

2.2 Grain size analysis

Samples of Manila bay were dispersed by soap and dried in an oven. A laser particle analyzer (LISST 100) was used for determination of grain size analysis for Manila Bay samples by Dr. Yasuyuki Nakagawa of the Port and Airport Research Institute, Yokosuka, Japan. Samples of Pasig river and Laguna lake were analyzed by SALD-3000S (Shimadzu).

2.3 Metal concentration analysis

Approximately 0.5 grams of the powdered sample was digested using 2ml of nitric acid and 4ml hydrochloric acid. The solution was heated at 130 degrees for 2 hours and filtered using GF/B mesh glass fiber filter. ICP/AES (Shimadzu ICPS-7000) was used for the determination of most of the metal concentrations and ICP/MS (Seiko SPQ-9000) was used for trace metal analysis as for Pb and Cd. However, Cd concentrations of most of the samples were below the detection limit. The values measured by ICP/MS were corrected by concentrations of Bi and Y as internal standards.

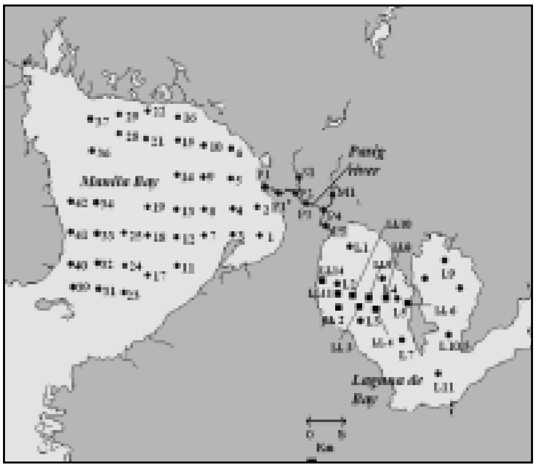

Fig. 1 Sampling Locations in the Philippines

Figure.2 Sampling Locations in Tama River

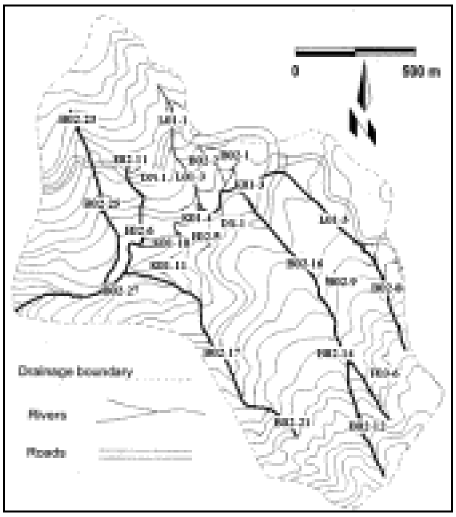

Fig.3 Northern part of Luzon Island which was used as agricultural field

3. Results and discussion

Metal concentrations and the grain size of sediments in Manila bay, in Laguna lake, in Pasig river, in Marikina river, in San Juan river and in Tama river were measured. A concentration profile of metals of sediments in Laguna lake was quite uniform as shown in Table.2. High metal concentrations were found with sediments of Pasig river area. In the case of Pb, the highest (87.5mg/kg) was found at the location of the junction of San Fan River to Pasig River.

Due to salt intrusion, coagulation takes place at the river mouth area, resulting that the precipitation of iron hydroxide enriches sediment concentration.(Jinming Duan. et al 2002).

Our study in Table.1 shows that relatively uniform concentration of iron around 40g/kg was observed possibly because iron is a main composition of suspended particles originated from both natural and anthropogenic sources.

The average grain size in this study was relatively small, showing that the sediments were mainly consisted of clay and silt.

Table.1 Summary of results of metal concentrations of sediments

location \ element	Cr	Co	Ni	Cu	Zn	Zr	Pb	Fe	Ti	Mn	Cd	average	clay	silt	sand
	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	g/kg	g/kg	g/kg	mg/kg	µm	%	%	%
1	15	18	11	98	128	39	27	43	3	0.8	0.5	10.7	21	72	7
2	34	20	20	94	218	25	41	44	2	0.6	0.7	8.3	28	72	0
3	9	21	7	109	94	38	24	50	3	1.3	0.0	11.9	23	69	8
4	25	14	12	87	198	17	27	40	2	0.6	0.9	8.5	29	68	3
5	34	17	17	96	219	21	41	48	2	0.9	0.6	7.6	31	67	2
6	38	20	21	89	173	21	37	44	2	0.8	0.6	8.0	27	72	1
7	14	18	11	72	84	26	20	44	3	1.3	0.0	10.6	18	76	6
8	28	19	26	80	110	28	23	43	2	1.0	0.0	9.8	25	67	8
9	30	15	15	80	153	18	23	44	2	1.0	0.6	8.7	26	69	5
10	45	22	24	94	148	23	22	44	2	1.0	0.4	9.2	27	67	6
11	12	20	10	42	63	22	16	44	2	1.0	0.0	17.1	9	82	9
12	17	18	15	58	81	21	17	40	2	1.3	0.0	11.8	18	76	6
13	19	13	13	66	87	17	13	37	2	1.1	0.5	9.2	24	74	2
14	26	14	15	72	105	17	16	43	2	1.1	0.5	12.4	18	70	11
15	28	13	14	62	106	10	14	38	1	0.9	0.4	9.8	23	70	7
16	31	18	19	59	94	14	12	45	2	1.0	0.0	9.2	29	66	5
17	15	17	13	47	68	20	16	40	2	1.3	0.0	20.2	7	81	13
18	21	18	18	63	88	20	16	42	2	1.4	0.0	9.7	24	70	6
19	21	13	14	62	89	13	12	38	2	1.3	0.0	11.4	18	75	7
21	35	20	23	75	113	21	18	46	2	1.0	0.5	9.1	28	66	6
22	27	14	15	63	89	7	12	37	1	0.9	0.0	9.4	27	68	5
23	12	16	10	33	59	23	17	45	2	1.1	0.0	20.5	7	78	14
24	25	22	23	76	104	21	18	43	2	1.6	0.0	12.0	19	72	9
25	22	14	17	63	90	15	15	40	2	1.4	0.0	12.3	18	75	7
28	38	22	27	88	124	18	16	45	2	1.1	0.0	9.6	24	71	5
29	31	16	18	68	99	11	11	43	2	1.0	0.0	9.6	25	69	6
31	23	23	21	58	91	22	17	43	2	1.4	0.0	21.5	7	76	16
32	21	14	17	63	92	12	14	39	2	1.9	0.0	10.4	17	73	10
33	30	21	26	80	113	21	15	43	2	1.8	0.0	12.3	17	77	6
34	40	25	33	101	147	21	18	43	2	1.6	0.5	11.0	20	75	6
36	44	24	30	93	134	21	12	41	2	1.0	0.0	9.2	27	67	6
37	29	15	17	65	91	11	10	40	1	0.9	0.0	8.9	24	72	3
39	20	13	16	61	91	17	18	38	2	1.7	0.0	10.6	23	70	7
40	24	15	17	101	120	12	15	40	2	1.9	0.7	8.4	32	63	5
41	23	13	15	99	115	9	15	37	1	1.2	0.0	9.4	29	63	8
42	23	13	15	63	92	8	13	36	1	1.1	0.0	8.2	35	58	7
mean	26	17	18	74	113	19	19	42	2	1.2	0.2	11.0	22	71	7
median	25	17	17	72	104	20	16	43	2	1.1	0.0	9.8	24	71	6
st.dev.	9	4	6	18	39	7	8	3	0	0.3	0.3	3.5	7.0	5.0	3
max	45	25	33	109	219	39	41	50	3	1.9	0.9	21.5	35	82	16
min	9	13	7	33	59	7	10	36	1	0.6	0.0	7.6	7	58	0

Table.2 Summary of results of metal concentrations of sediments in Laguna Lake in 2002

location \ element	Cr	Co	Ni	Cu	Zn	Zr	Pb	Fe	Ti	Mn	Cd	average
	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	g/kg	g/kg	g/kg	mg/kg	μm
L1	31	24	18	116	129	35	21	58	3	1.2	0.4	8.6
L2	13	21	10	127	106	36	27	54	3	1.9	0.5	9.0
L3	13	24	10	138	106	50	22	58	4	1.4	0.3	10.7
L4	18	21	11	121	102	37	20	54	3	1.7	0.3	7.2
L5	18	25	12	132	107	39	22	55	3	1.7	0.3	9.9
L7	13	23	9	119	91	35	18	56	3	1.5	0.6	11.2
L9	19	23	14	124	110	36	21	57	3	1.3	0.4	8.7
L10.5	13	21	10	121	90	33	19	56	3	1.3	1.2	11.5
L11	11	52	9	79	119	32	16	88	3	4.0	1.2	16.0
mean	17	26	12	120	107	37	21	60	3	1.8	0.6	10.3
median	13	23	10	121	106	36	21	56	3	1.5	0.4	9.9
st.dev.	6	10	3	17	12	5	3	11	0	0.9	0.4	2.5
max	31	52	18	138	129	50	27	88	4	4.0	1.2	16.0
min	11	21	9	79	90	32	16	54	3	1.2	0.3	7.2

Table.3 Summary of results of metal concentrations of sediments in Laguna Lake in 2003

	Na	Mg	K	Ca	Ti	Mn	Fe	Sr	Cr	Ni	Cu	Zn	Zr
	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	%	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg
LL2	975	4060	1230	2269	2949	1437	4.9	50	13	121	143	92	36
LL3	1996	4283	1357	2721	2867	1454	4.9	60	24	9.9	148	89	33
LL4	1011	4262	1233	2618	2848	1494	4.6	58	13	9.1	142	82	33
LL6	1539	4673	1645	4716	2602	1190	4.5	73	13	22.8	151	97	32
LL8	1242	5483	1328	2334	2583	1384	4.7	57	12	10.3	165	45	43
LL9	1383	4725	1257	2709	2222	1632	4.8	63	11	17.1	145	31	39
LL10	2191	5640	1591	2337	2787	1232	5	56	12	17	165	55	39
LL11	2037	5271	1437	2516	2166	1604	4.5	63	12	31.3	152	43	32
LL12	6693	6074	3235	4162	3615	1771	6.3	79	20	133	182	144	38
LL14	2570	5356	2251	4843	2507	2629	5.3	85	15	12.9	157	138	31
mean	2164	4983	1656	3122	2715	1583	5	64	14	38	155	82	36
media	1768	4998	1397	2664	2695	1474	4.85	61.5	13	17.1	151.5	85.5	34.5
stdev.	1677	676.5	633.9	1027	412	407.6	0.534	11.1	4.2	47.2	12.56	38.83	4.01
max	6693	6074	3235	4843	3615	2629	6	85	24	133	182	144	43
min	975	4060	1230	2269	2166	1190	4	50	11	9	142	31	31

Table.4 Summary of results of metal concentrations of sediments in Pasig River

location \ element	Cr	Co	Ni	Cu	Zn	Zr	Pb	Fe	Ti	Mn	Cd	averag
	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	g/kg	g/kg	g/kg	mg/kg	μm
P1	44	29	25	133	311	19	71	59	3	0.6	0.6	16.1
P2	43	31	26	114	239	15	51	58	2	0.9	0.4	13.6
P3	30	24	17	167	186	11	29	55	3	1.3	1.4	11.2
P4	42	33	21	76	148	17	18	56	3	0.8	0.2	15.7
P5	29	28	16	60	113	20	13	47	2	1.0	0.3	11.7
S1	27	21	15	114	195	23	88	57	3	0.9	0.4	10.0
M1	47	33	25	128	209	11	23	63	3	0.8	1.0	15.9
mean	37	28	21	113	200	16	42	56	3	0.9	0.6	13.5
median	42	29	21	114	195	17	29	57	3	0.9	0.4	13.6
st.dev.	8	5	5	36	64	4	29	5	0	0.2	0.4	2.5
max	47	33	26	167	311	23	88	63	3	1.3	1.4	16.1
min	27	21	15	60	113	11	13	47	2	0.6	0.2	10.0

3.1 Correlation coefficients of metal concentration versus grain size

In order to understand the relationships between metal concentration and grain sizes, correlation coefficients were calculated and listed in Table.5. High correlation coefficients were found among Ti, Mn and Fe, and also between Cr and Ni.

Mean grain size did not have a high correlation coefficient with any of the metal concentrations. In this study, grain size of all sediments was in the fine range and has a little difference among locations. High correlation coefficients between metal concentrations and grain size were not observed.

Table.5 Correlation coefficients in Philippines

	Cr	Co	Ni	Cu	Zn	Zr	Ti	Mn	Pb	Fe	Cd
Cr											
Co	0.20										
Ni	0.85	0.17									
Cu	0.15	0.37	0.03								
Zn	0.66	0.34	0.42	0.53							
Zr	-0.46	0.34	-0.38	0.45	-0.11						
Ti	-0.11	-0.53	0.09	-0.61	-0.30	-0.20					
Mn	-0.08	-0.57	0.22	-0.63	-0.41	-0.43	0.83				
Pb	0.27	0.21	0.10	0.46	0.76	0.13	-0.27	-0.41			
Fe	0.02	-0.60	0.19	-0.66	-0.29	-0.38	0.95	0.87	-0.33		
Cd	0.11	0.43	-0.11	0.61	0.53	0.16	-0.46	-0.55	0.33	-0.48	
mean grain size	-0.14	0.37	-0.02	-0.27	-0.08	-0.03	0.00	0.01	0.00	-0.08	-0.08

3.2 Estimation of polluted area in Manila Bay from the spatial distribution of relative concentration (RC) to the mouth of Pasig River which was most polluted by anthropogenic source

In the case of Pb and Zinc, the highest location in Manila Bay was found at the mouth of Pasig River. The distribution of Pb and Zn concentration illustrates the diffusion of those in Manila Bay originating from the mouth of Pasig River. To identify the polluted area in Manila Bay, Relative concentration (RC) of concentration of each location to concentration of P1 was calculated and the relative concentration (RC) of each location was compared with the RC of Laguna Lake which was not affected by anthropogenic pollution. From the Fig.4, area polluted by lead was extent of about 10km from the mouth of Pasig River. Fig.5 shows the relative concentration of zinc distributed in the same way as lead.

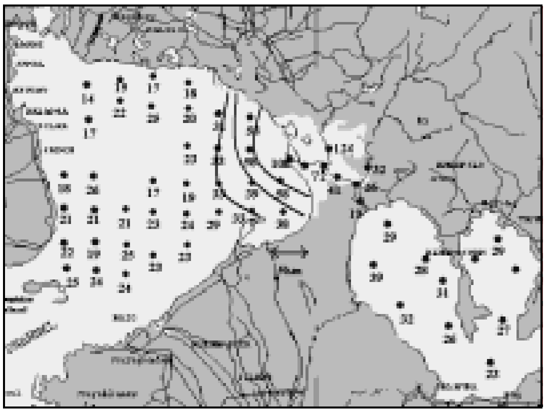

Fig.4 Relative lead concentration to P1concentration

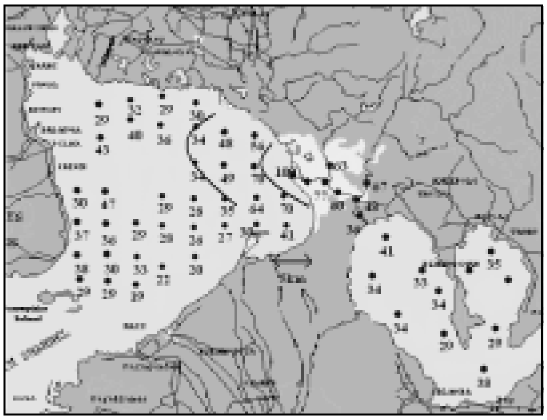

Fig.5 Relative zinc concentration to P1 concentration

3.3 Correlation coefficient between lead and zinc

Zinc was identified as hazardous elements to aquatic lives in water environment quality standard in Japan last year.

As is demonstrated in the last section, lead and zinc showed similar spatial distribution of concentrations in Manila Bay. To evaluate the relations between two elements were plotted against zinc concentration in Fig.6. In Tama River, in Manila Bay, in Pasig River, positive correlation was shown.

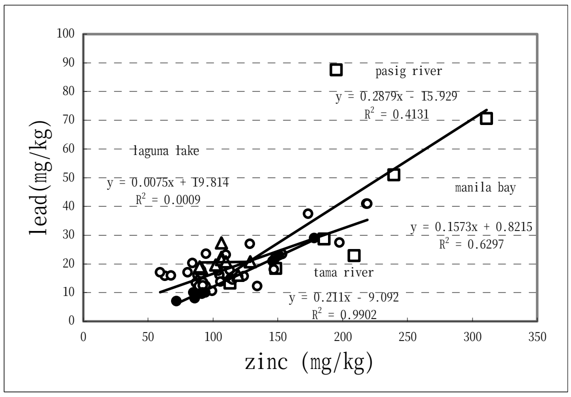

Fig.6 Correlation coefficient between lead and zinc

3.4 Metal associated with SS(suspended solids) in Pasig river, Marikina river and San Juan river.

Metals associated with sediment are possibly originated from metals with SS(suspended solids) in water. To investigate the metal concentration with SS and to study the use of lead concentration as a indicator of particulate polluted matter, concentration of each metal contained in SS was measured and compared each other.

Because most part of Laguna Lake area was not affected by anthropogenic sources, metals associated with SS in each river were normalized with metal concentrations with SS in Laguna Lake, as shown in Fig.7.

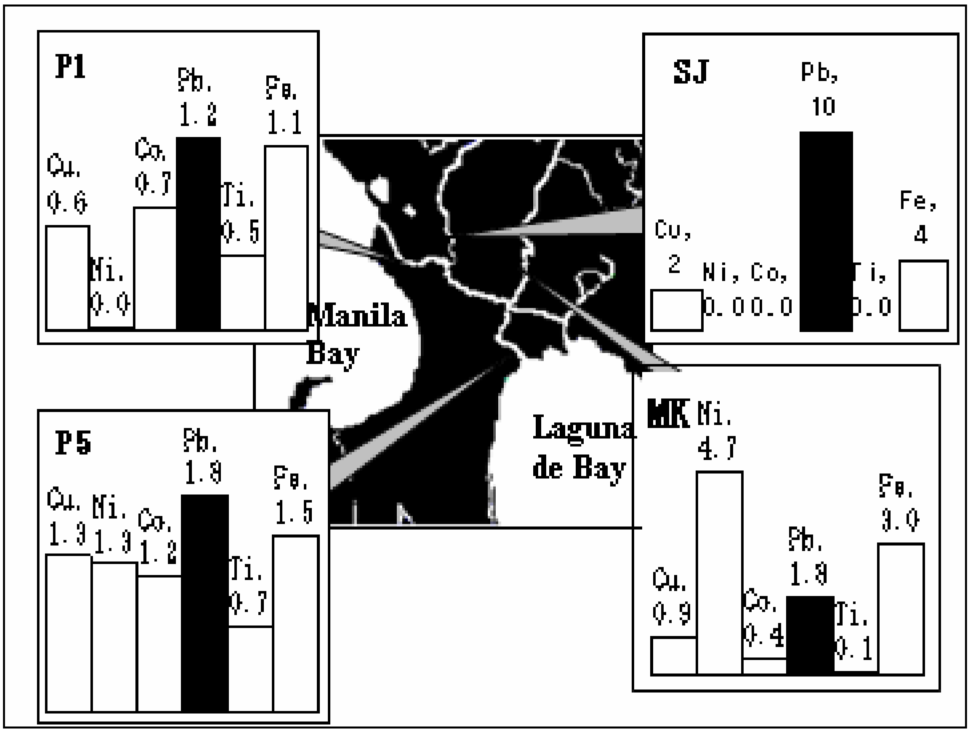

Fig.7 Distribution of particulate metal concentration in Rivers normalized with particulate metal concentration in Laguna Lake.

Metal concentration of road-side deposits was measured as a possible metal source of SS in river water. In Japan, about twenty years ago, addition of Pb to gasoline was forbidden while in Manila only a few years have passed since then. However, small differences were found between Pb concentration of deposits in Tokyo and that in Manila. This striking result could be interpreted that the amount of lead absorbed to particles were similar regardless of sampling countries, though the main source of Pb is consider to be fuel, even if other origins are taken into account (M.Legret. et al 1999).

3.5 Deposits at the sides of roads as a source of metals of sediments

Northern part of Luzon Island is mainly agricultural area which was not affected by industrialized human activity. Metals associated with road deposits in urban area were normalized with metals with soils of the agricultural area to compare the increase amount of each metal by involving mainly the dusts from auto exhaust.

The relative concentration of lead in the road side deposits was the highest among the metals. Enrichment of lead in urban area was identified from the Fig.8 From the results of relative concentration (RC) of SS in river water and in road side deposits, lead seems to be the best indicator of particulate pollutants in urbanized area.

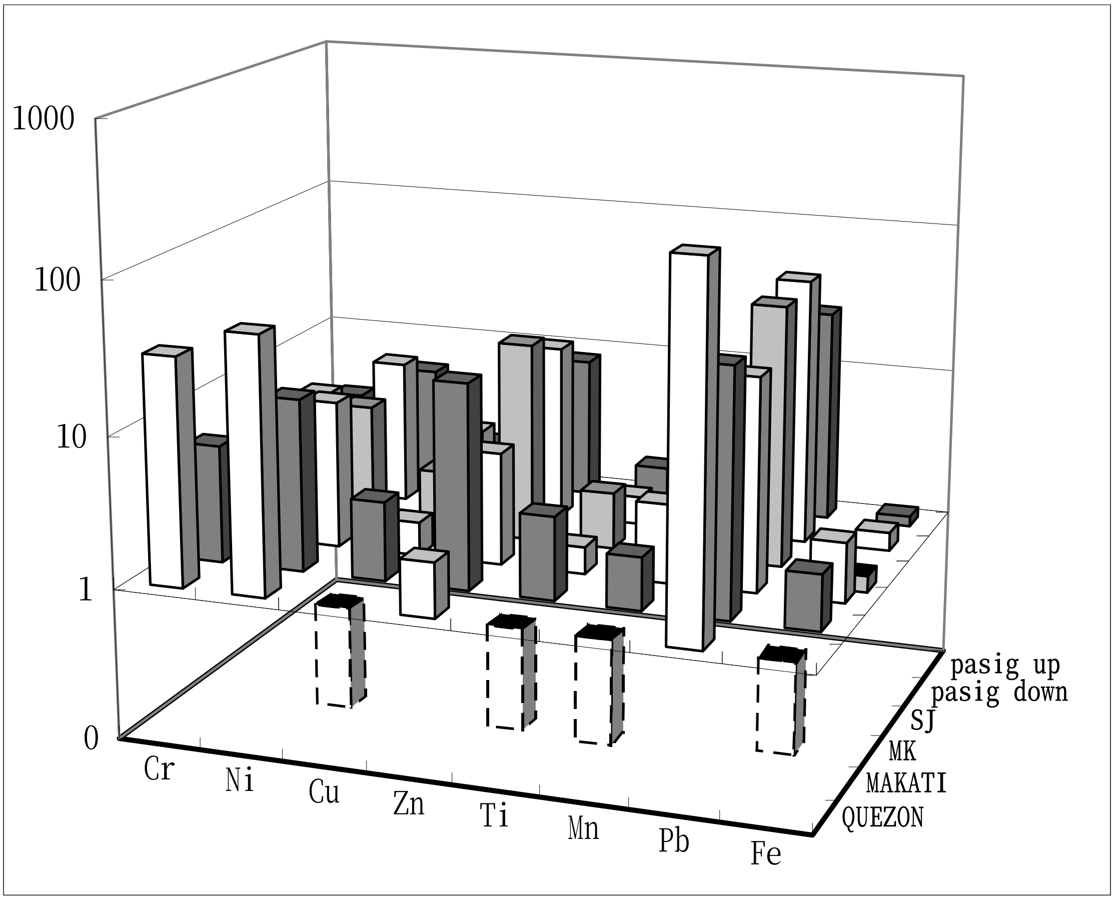

Fig.8 Relative concentration of each metal associated with road deposits to each metal concentration of soils in agricultural area

4 Calculation of transport distance of particles

4.1 transport of particles in Pasig River

Shields derived the following equation in 1936 for the relationship between shear velocity and critical bet flow.

$$\frac{\tau_{0cr}}{(\gamma_s - \gamma)d} = function\left(\frac{U_* d}{\nu}\right)\cdots(1)$$

Where τ_{0cr} is critical bed flow, γ is weight of water unit volume, U* is shear velocity and d is diameter of particles.

In the case of general river flow, usually Re number is high. In that case, equation (1) can be reduced to eq(2).

$$\frac{\tau_{0cr}}{(\gamma_s - \gamma)d} = 0.06\cdots(2)$$

From the balance of the forces acting on water body in uniform flow, eq (3) was derived.

$$\tau_b = \gamma h S_o \cdots(3)$$

where

τ_b is bed flow force, γ is weight of water of unit volume, h is water depth and S_0 is gradient of bed of river.

Because gradient of the river bed is small in Pasig River, energy gradient can be used as the gradient of the bed of river.

In this study, energy gradient was calculated from the difference of surface water level between Laguna Lake and Manila Bay (5m) divided by the distance of Pasig River between the Lake and the Bay (25km).

$$from(3) \quad \tau_b = 1.0[gf/cm^3]\times 10^6 \times 9.8[m/s^2]\times 5[m]\times \frac{5[m]}{25000[m]}\cdots(3')$$

$$from(2) \quad \tau_c = 0.06\times(2.75-1.0)[gf/cm^3]\times 10^6 \times 9.8[m/s^2]\times d \times 10^{-6}[m]\cdots(2')$$

By substituting (2')=(3'), we obtained the solution below.

d＝9523(μm)=1(cm)

The calculation above shows that particle whose diameter is less than 1cm is carried to down stream by river flow. Among sampling locations, there was an area where take samples. Though all road side deposits could not be carried out to Manila Bay, there was spatial distribution in the river. Most particles were considered to be carried down to the vicinity of Manila Bay.

Sediment of P1 was consisted in particles less than 1mm and ratio of sand fraction to all fraction was only 16.5%. No particles more than 200μm existed in sediments of P1. most of the particles larger than 200μm was deposited to the sediments is the down stream region of the Pasig river.

4.2 transport of particles in Manila Bay

The distance of transportation of particles from the mouth of Pasig river was calculated in Manila Bay.

In the case of small Reynolds number and in the case of clay fraction the settling velocity of particles can be obtained by the Stokes equation.

$$V_s = \frac{D^2 g(\rho_s - \rho_f)}{18\nu\rho_f}\cdots(4)$$

where Vs is settling velocity, D is diameter of particle, g is accelerated velocity, ρ_s is density of particle, ρ_f is density of fluid and ν is kinematical viscosity.

In this study, we assumed that the water temperature was 30 Celsius degree and the mean grain size at P1 used for the diameter of the particle. The density of clay was assumed to be the density of the particles.

D＝16μm (mean grain size at mouth of Pasig river)
g＝9.8m/s^2 (accelerated velocity)
ρ_s＝2.75gf/cm^3（density of clay）
ρ_f＝1.0gf/cm^3 (density of water)
ν：0.8×10^{-6} m^2/s （kinematical viscosity at 30 Celsius degree）
Ho：17m（mean Manila Bay depth）
Substituting these values into (4), we got

$$\frac{H_o}{V_s} = t = \frac{18\times 17\times 0.8\times 10^{-6}\times 1.0\times 10^6}{(16\times 10^{-6})^2\times 9.8\times(2.75-1.0)\times 10^6} = 15.4[h]$$

We could obtain the horizontal diffusion distance by multiplying average surface velocity0.5m/s and 15.4 hours.

0.5×15.4×3600=27.7km

The clay particle could be transported 27.7km from the mouth of Pasig River in Manila Bay.

From the spatial distribution of lead concentration, the particles were carried out to 10km distance from the mouth of Pasig River. There was the difference between calculated distance and the estimated distance from the distribution because there were fluctuations due to tidal current water flow and direction. In addition, bottom water flow direction is different from that of surface water.

5. Conclusion

Heavy metal concentration contained in bottom sediments were measured for 37 samples taken in Manila bay, 9 samples taken in Laguna lake, and 8 samples taken in Pasig river. Soil samples were taken in northern part of Luzon Island, an agricultural area.

Lead and zinc distribution indicates that the urban run-off from Metro Manila is the main source of lead and zinc in sediments in Manila bay. Judging from spatial distribution, the area polluted by lead and zinc was identified. The area within the distance of 10km from the mouth of Pasig River was polluted.

The results of relative concentration (RC) of metals in SS (suspended solids) in river water and the results of RC in road side deposits show that lead seems to be the best indicator to examine the fate of particulate pollution in water in surrounding area of Metro Manila.

6. Acknowledgement

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