

Distribution Function of Heavy Metals in River Sediment

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Along with remarkable industrial development and an increase in city populations, environmental pollution has become a serious problem.

The authors have studied environmental analysis of heavy metals in river sediment to understand the status of river pollution (Nishida et al.1982; Nishida et al.1983). In these studies, one of the most important problems is to determine the probability distribution function of each heavy metal in river sediment. Few of the studies concerned with the distribution of heavy metals have discussed the distribution function.

In this paper, we discuss the distribution function of heavy metals as found in upstream and downstream sediments of first class rivers in Japan. The first class rivers were designated by the Japanese government in 1963 as important in the national land conservation and economic program.

MATERIALS AND METHODS

We obtained random samples of unpolluted upstream sediment and of downstream sediment by means of an Ekman-Biedge instrument at ten points from the right to left banks of each of 92 rivers in Japan. The upstream sediments were taken at unpolluted places and those downstream were taken about 1 km from the river mouth. Each of these sediments was stirred well and dried at 80°C. We used 15g each of dry sediment passed through a 20 mesh sieve, and measured the quantities of heavy metals soluble both in total and in 0.5N-HCl. We used the heavy metals that were soluble in dilute hydrochloric acid because nongeological metals, due to the working of chelate agents or dilute acid, are more easily removed than metals contained as minerals (Tada et al.1976)

In order to decide the distribution function of each heavy metal, its AIC was calculated. The AIC is a statistical decision procedure for model fitting which was introduced

by Akaike (1974). This procedure uses statistics of the following type:

$$AIC = -2\log_e(\text{Maximized Likelihood}) + 2K \quad (1)$$

where K is the number of parameters within the model which are adjusted to attain the maximum likelihood. We adopted the model which minimizes the AIC within a set of possible alternatives.

In general, models which can be fitted to the distribution of environmental substances may be chosen from a set of probability distributions around the normal distribution. We have selected five models; Gamma distribution, Weibull distribution, Beta distribution, Hypergamma distribution and Lognormal distribution. Each of these distributions converges to the normal distribution under certain limiting conditions. The probability density function (p.d.f) of each model is shown below.

p.d.f of Gamma distribution

$$F(x:\alpha, \beta) = \frac{\alpha^\beta}{\Gamma(\beta)} x^{\beta-1} \exp(-\alpha x) \quad (2)$$

p.d.f of Weibull distribution

$$F(x:\alpha, \beta) = \alpha \beta x^{\alpha-1} \exp(-\beta x^\alpha) \quad (3)$$

p.d.f of Beta distribution

$$F(x:\alpha, \beta) = \frac{1}{B(\alpha, \beta)} x^{\alpha-1} (1+x)^{-(\alpha+\beta)} \quad (4)$$

p.d.f of Hypergamma distribution

$$F(x:\alpha, \beta, \gamma) = \frac{\alpha \beta^{\gamma/\alpha}}{\Gamma(\gamma/\alpha)} x^{\gamma-1} \exp(-\beta x^\alpha) \quad (5)$$

p.d.f of Lognormal distribution

$$F(x:\alpha, \beta) = \frac{\beta}{\sqrt{2\pi} x} \exp\left\{-\frac{\beta^2}{2} (\log x - \alpha)^2\right\} \quad (6)$$

The AIC was calculated by our program (Miyai et al. 1982).

RESULTS AND DISCUSSION

We measured the quantities of heavy metals soluble both in total and in 0.5N HCl of Cu, Pb, Cd, Zn, Cr and Ni of the upstream and the downstream sediment (the average of ten measurements) of 92 first class rivers. The average and the variance of each heavy metal are shown in Table 1. Frequency distributions of the data used for calculating AIC for each heavy metal are shown in Figure 1. The AIC's calculated by (1) for each heavy metal are shown in Table 2.

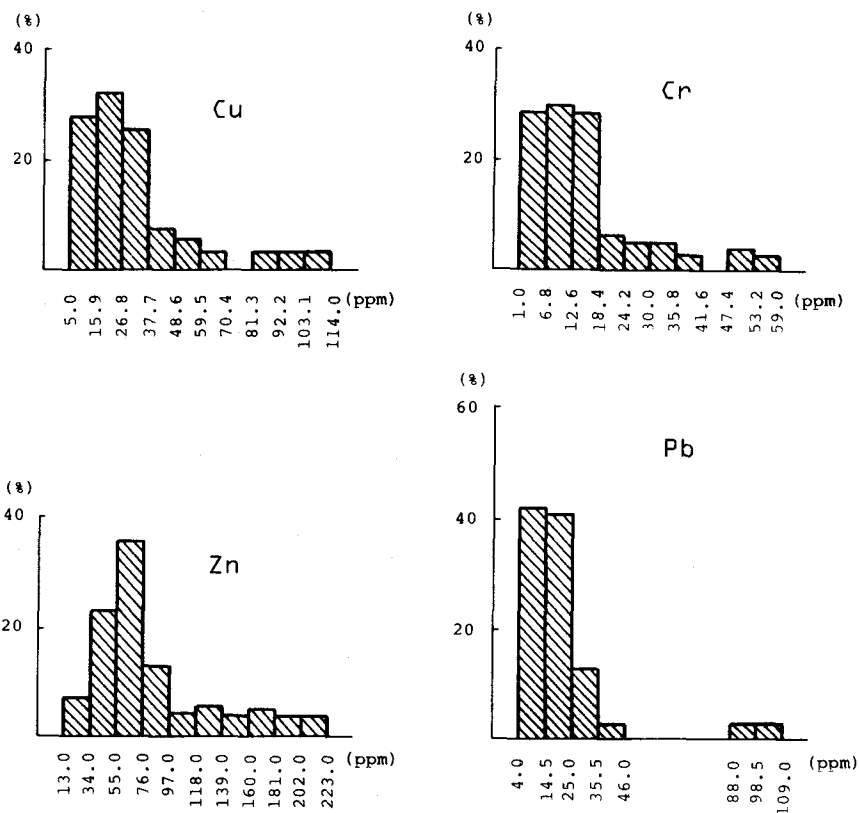
Table 1. The average(AV) and the variance(VR) of each heavy metal.

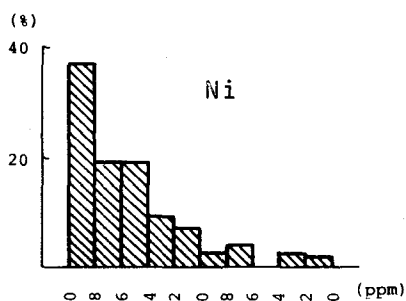
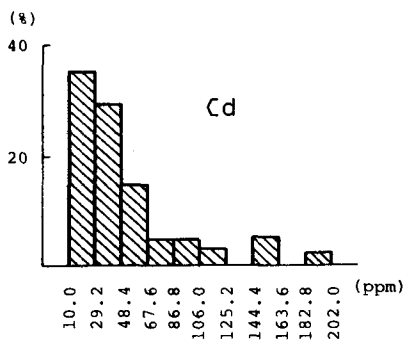
<u>Total soluble</u>						
<u>Upstream</u>	Cu	Cr	Zn	Pb	Cd	Ni
AV	27.57	14.13	79.89	19.13	0.50	18.79
VR	401.55	132.36	1984.16	254.14	0.16	283.05
<u>Downstream</u>						
AV	48.47	32.38	180.76	26.79	1.18	22.63
VR	1998.76	642.91	28288.16	457.66	1.41	268.49

<u>0.5N-HCl soluble</u>						
<u>Upstream</u>						
AV	8.86	1.49	20.06	9.74	0.24	2.25
VR	59.28	2.80	440.75	137.03	0.08	5.83
<u>Downstream</u>						
AV	24.72	5.32	76.58	15.40	0.63	5.41
VR	2305.07	97.52	11175.96	241.55	0.78	30.05

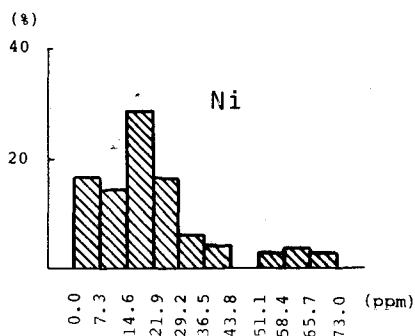
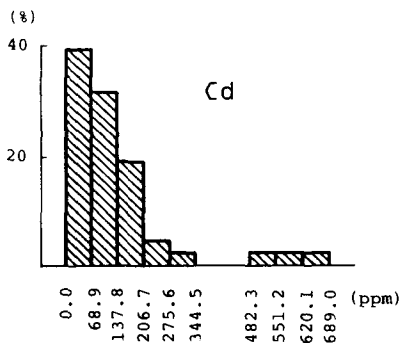
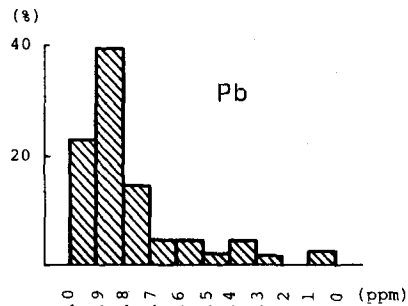
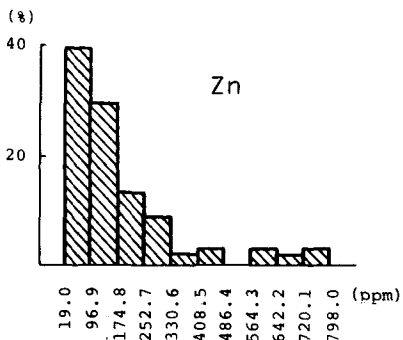
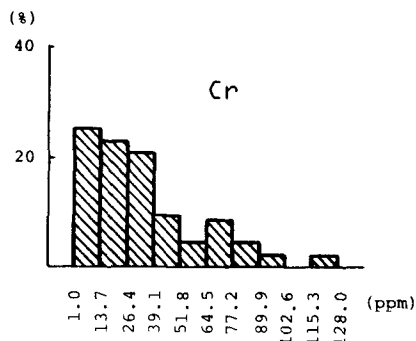
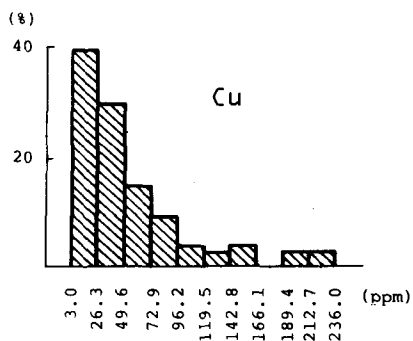
Figure 1. Frequency distribution of each heavy metal.

Total soluble (upstream)

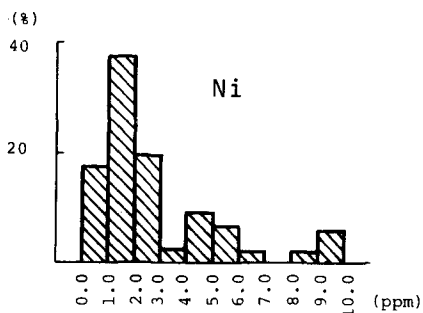
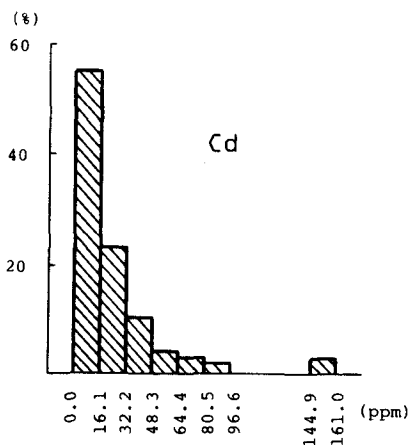
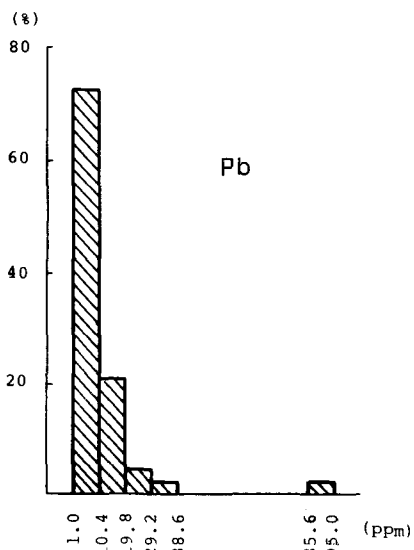
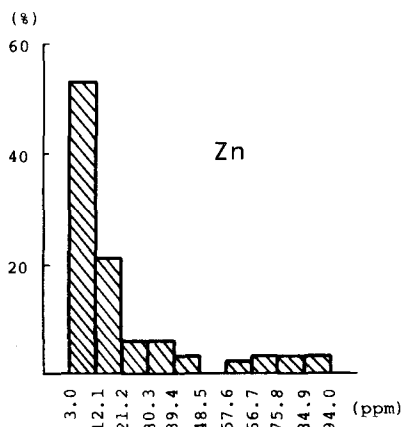
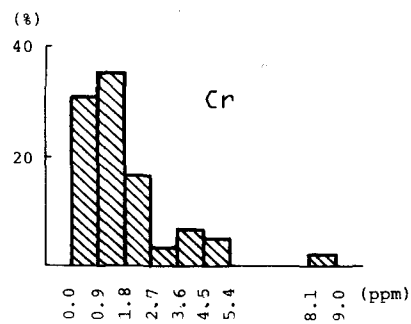
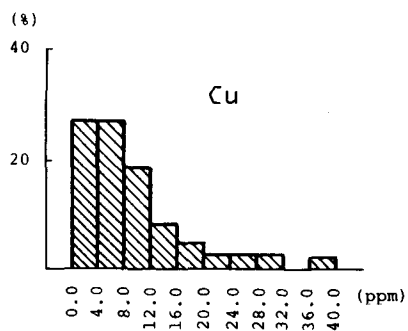




Total soluble (downstream)



0.5N-HCl soluble (upstream)



0.5N-HCl soluble (downstream)

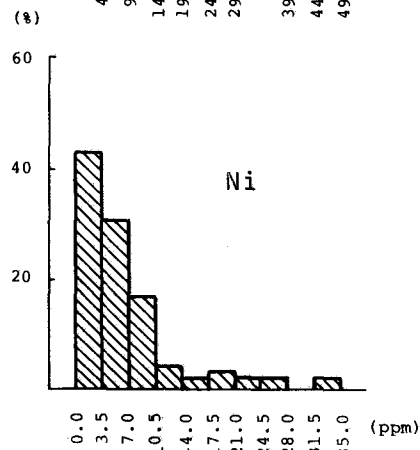
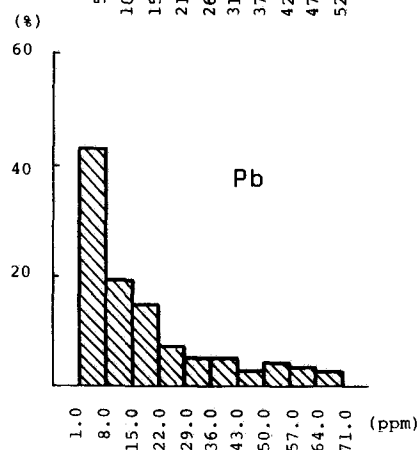
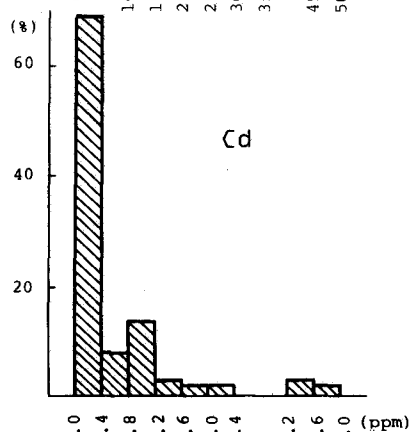
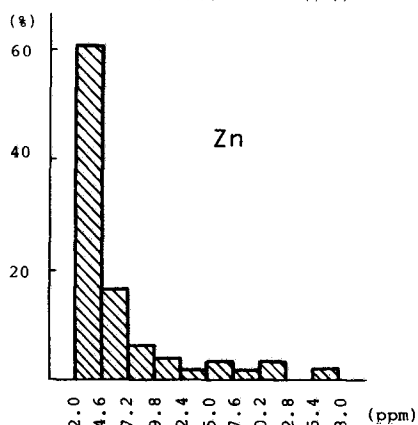
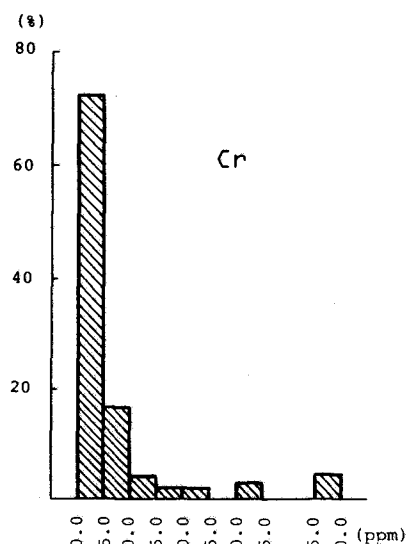
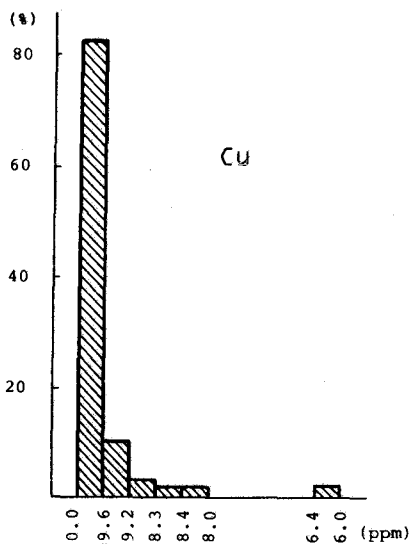


Table 2. The AIC of each heavy metal.

<u>Total soluble</u>						
<u>Upstream</u>						
Distribution	Cu	Cr	Zn	Pb	Cd	Ni
Gamma	600.6	°515.1	730.1	539.5	692.5	°565.2
Weibull	606.3	518.8	737.8	554.1	698.0	°566.8
Beta	602.9	520.5	733.1	*524.4	*684.3	569.8
Hypergamma	*592.2	°515.9	*723.9	572.0	698.9	°566.5
Lognormal	°597.5	*512.7	°727.5	°525.1	*684.5	*562.4
<u>Downstream</u>						
Gamma	°690.6	*631.6	875.4	°597.8	822.1	*577.4
Weibull	692.9	°632.1	879.1	601.7	822.6	*577.1
Beta	698.9	658.4	°871.3	599.3	905.0	611.9
Hypergamma	698.3	°633.6	887.5	°596.9	*816.9	583.3
Lognormal	*687.7	638.3	*866.9	*593.7	842.6	588.8
<u>0.5N-HCl soluble</u>						
<u>Upstream</u>						
Gamma	°453.9	209.3	574.2	466.0	*604.2	265.1
Weibull	456.6	214.0	577.8	472.4	*604.2	267.9
Beta	°454.8	*194.9	*549.7	°456.9	624.6	*250.4
Hypergamma	456.2	214.9	588.2	488.2	610.7	273.8
Lognormal	*450.7	*194.7	°556.4	*452.9	*604.6	*250.8
<u>Downstream</u>						
Gamma	769.5	491.1	981.7	°690.3	947.4	493.9
Weibull	762.4	482.8	987.4	°690.9	944.9	496.5
Beta	751.2	*447.2	972.3	693.5	965.5	493.8
Hypergamma	774.1	498.2	986.6	693.8	953.9	504.4
Lognormal	*744.9	°452.1	*965.1	*685.5	*938.3	*488.3

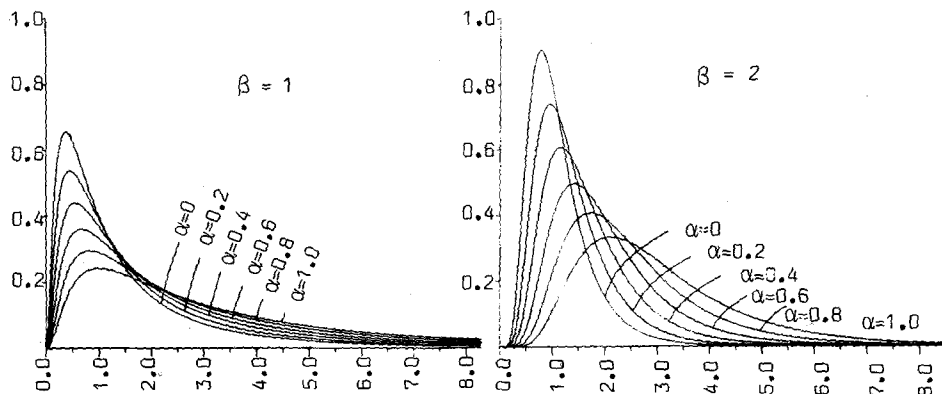
* The minimum AIC

° The approximate minimum AIC

In Table 2 each heavy metal of upstream sediment in total soluble is distributed lognormally or nearly lognormally. However, each downstream heavy metal is not always distributed lognormally. Cr and Ni are distributed according to the Gamma function and Cd according to the Hypergamma function. Each heavy metal soluble in 0.5N-HCl both upstream and downstream is distributed lognormally or nearly lognormally.

In general, total soluble is considered to be derived from both geological and nongeological sources. On these samples of Japanese rivers, the point from which the upstream sediments were taken was not populated; the downstream sediments were taken from a point about 1 km from the mouth of a river. Thus, each heavy metal of downstream sediment in total soluble is distributed according to both functions based on the geological and nongeological sources. Consequently,

Figure 2. Lognormal Distribution.



each heavy metal of downstream sediment is not always distributed according to the same function. Because each heavy metal of upstream sediment is considered to be derived from only geological matter, they are distributed according to the same function, i.e., lognormal. The same holds true for all 0.5N-HCl soluble heavy metals. From these results we postulate that each heavy metal of river sediment derived from a single source, is distributed lognormally.

The general form of lognormal distribution function is shown in Figure 2.

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