

Background values for evaluation of heavy metal contamination in sediments

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Abstract

The contamination of sediments can be evaluated from comparison of the current concentration with the estimated background value. Although it is known that grain size affects background concentrations, there have been difficulties in evaluating this effect. The specific surface area of sediments, which can be calculated from the grain size distribution, is introduced as a property index for correlation of background values. The results show that for sandy-to-sandy silt sediments, the background values of metals are expressed in terms of fines content, which can be correlated with the corresponding specific surface area. However, for silty clay or clayey silt sediments, which would have a high metal retention capacity, the level of the background may be constrained by the relatively low concentrations of metals in seawater. Finally, background levels of Zn, Cu and Pb are presented.

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1. Introduction

The background concentration of hazardous substances in sediments is an important factor for evaluating the degree of contamination. It has often been concluded that surface sediments were contaminated from the fact that deeper sediments show relatively low concentrations of pollutants, as shown in Fig. 1 [1]. However, this assumption does not hold if the grain size of sediments varies significantly with depth, in which the background value also varies with depth, as shown in Fig. 2 [2]. This is because the background concentration of contaminants depends on many factors, including the particle characteristics, sorption/desorption capacity, and existing substances in nature.

Background values were found to agree well with average values for gneisses rocks [3]. However, the background values for copper, zinc, and lead do not necessarily agree with values

previously obtained. Therefore, to allow an accurate estimation of background values, various techniques have been used. Fukue et al. [1] used carbonate content as a normalizing substance. On the other hand, Cobelo-García and Prego [4] obtained baseline relationships between the concentrations of iron and pollutants, whilst Din [5], Cortesão and Vale [6] and Santschi et al. [7] used aluminum (Al) to normalize the concentrations of heavy metals in sediments. Titanium has also been used as a normalizing element in the field of geochemistry. All the techniques introduced here require data for non-contaminated sediment samples.

Santschi et al. [7] mentioned that if sediment is richer in Al, it is composed of smaller-grained particles. This is because Al is a constituent of clay minerals, which can adsorb a greater amount of heavy metals than coarse particles.

The fate of a pollutant depends on the surface activity factors of sediment particles, such as charge density, cation exchange capacity (CEC), specific surface area [8,9], and the equilibrium (natural) concentration of contaminants [10,11]. Since the particle characteristics of sediments are strongly influenced by the particle size, the grain size distribution may reflect the true nature in practice.

Many species of pollutants exist in nature and their concentrations vary widely. Therefore, it is necessary to estimate the background values to evaluate contamination. One easy and

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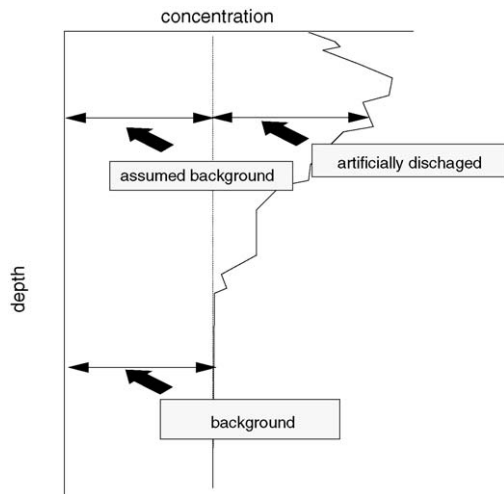


Fig. 1. Metal concentrations and constant background in a homogenous sediment profile.

economical method may be to use the grain size distribution, because it reflects the characteristics of particles, such as the specific surface area, which may govern the electrical charge density, CEC, etc. [9]. For example, if the size of a particle is very small, it may be a clay mineral, which has a relatively high charge density and high CEC. These characteristics mostly result from the high specific surface area.

To express the grain size distribution of soils, representative indices such as the clay content, coefficient of uniformity, and grain size at 10% fines have been used [12]. These values are significant when they are used in correlation with a particular property of soils or sediments. For example, the grain size at 10% fines has been used to predict the permeability of sands [13].

In this study, to characterize the grain size distribution, the concept of “an ideal specific surface area” is estab-

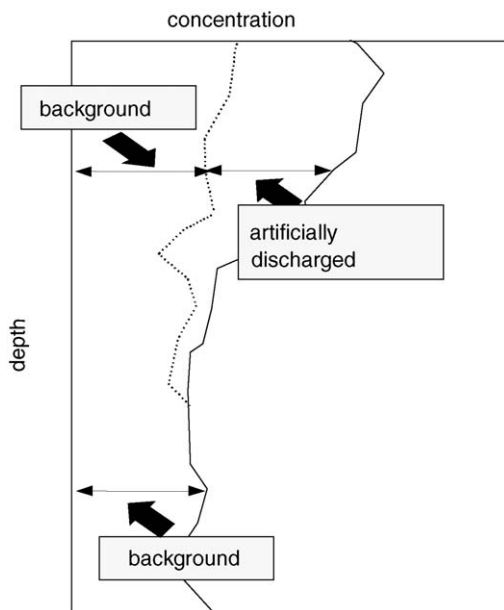


Fig. 2. Illustration of variable background concentrations with depth.

lished, whereby the grain size distribution of sediments can be expressed by a single constant value. This value is correlated to the background value of heavy metals in sediments.

When the background, B_G , is known, the degree of pollution, P_d , is defined as:

$$P_d = \frac{C - B_G}{B_G} \quad (1)$$

where C is the current concentration of a substance or element [1].

2. Concept of ideal specific surface area

The particles of actual soils or sediments show a wide range of shapes and sizes. With regard to size, they can be classified as gravel, sand, silt or clay fractions. The characteristics of soil particles have been assumed to be a function of the specific surface area, because most reactions, such as sorption, desorption, and interactions between particles, occur on the surface of the particles. Since it is not easy to determine the experimental specific surface area, a calculated specific surface area is used in this study. This calculated value is considered to be one of the property indices.

Soil particles have been assumed to be spherical in conventional grain size analysis. Therefore, it is not uncommon to use the specific surface area for particles assumed to be spherical. The calculated specific surface area is called “the ideal specific surface area” to distinguish it from an experimentally measured value.

The specific surface area of a sphere is defined by:

$$SS = \frac{S}{m_s} \quad (2)$$

where SS is the specific surface area, defined as the summation of the surface area of the individual particles divided by the total mass of the sample, S is the surface area, and m_s is the mass of a spherical particle. Furthermore, sediments are assumed to be composed of spherical particles. Since sediments consist of a large number and various sized particles, they are separated into groups, as shown in Fig. 3, and thus the specific surface area of

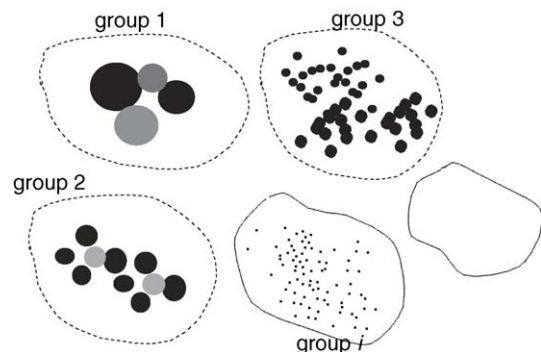


Fig. 3. Grouping of sediment particles for establishing ideal specific surface area.

sediments is given by:

$$SS = \sum \alpha_i SS_i \tag{3}$$

where α_i is the weight percent of group i .

3. Evaluation of SS

3.1. Grain size distributions

Various types of sediments were arbitrarily chosen to obtain the ideal specific surface area, SS, as shown in Fig. 4. Fig. 4a shows the results of the laser diffraction scattering method for particle-size analysis, which is based on two theories, i.e., Fraunhofer diffraction theory and Mie scattering theory [14]. The abrupt drops near 0.001 mm may result from disagreement between the two theories, because the analysis for particles smaller than 0.001 mm was carried out using the Mie theory. At present, there is no assurance that results using the Mie theory are correct, because there is no comparable analysis.

There is another type of analysis that can be applied to a small amount of sample. Fig. 4b shows results obtained using X-ray extinction [15]. This method is applied to particle sizes greater than 0.001 mm. Therefore, distributions for smaller particles (colloid sizes <0.001 mm) were assumed by extending the curves obtained. In this case, the curves were smoothed, as

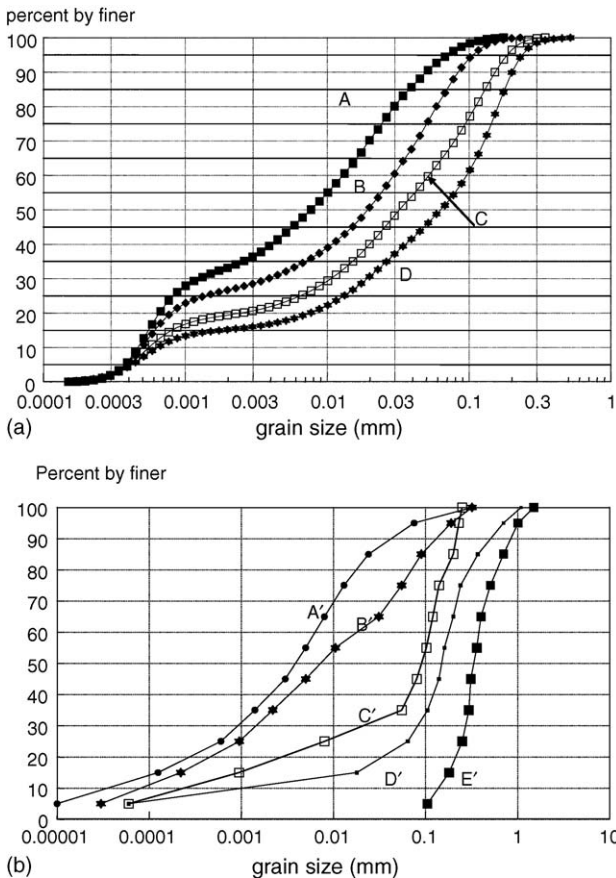


Fig. 4. (a) Grain size distribution of typical marine sediments, analyzed using the laser diffraction scattering method. (b) Smoothed grain size distributions of marine sediments used to determine relationships between the specific surface area and fines content.

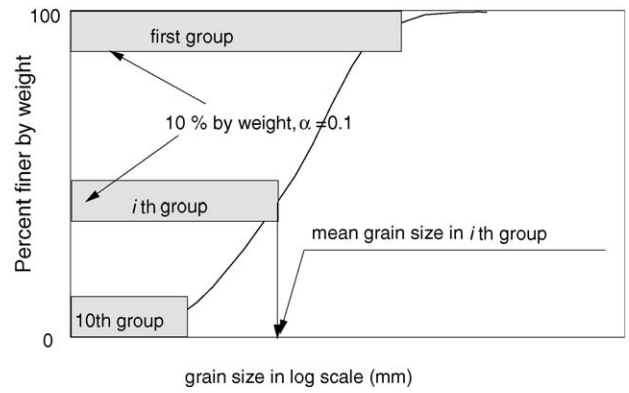


Fig. 5. Method for the determination of the mean grain size of groups.

shown in the figure. The reason why these analyses were used is explained below.

Conventional grain size analysis requires approximately 50 g of sample, but a weight of only 4–6 g is enough for the X-ray method and 0.03–0.06 g for the laser method. When a sediment sample is obtained using a tube, the amount of sample available for grain size analysis is usually within several g. Both test methods are presented later.

3.2. Calculation of SS

For calculation of SS, the sediment particles were assumed to be spherical. If a sediment sample is separated into 10 sequential groups of equal weight, the grain size distribution of the sediment can be obtained, as shown in Fig. 5. In this case, $\alpha_i = 0.1$ in Eq. (3).

In Fig. 5, the specific surface area of each group, SS_i , can be calculated using the mean grain size of each group. In this study, the mean grain size of a group was defined as grain size for average percentage fines. Because the mean grain size defined is based on weight percent, as well as specified grain sizes, i.e., D_{10} and D_{60} used in geotechnical engineering [12], calculation using this value provides an accurate SS_i . For the calculation of SS_i , the density of particles was assumed to be 2.67 g/cm^3 . After all the SS_i values are obtained, SS can be calculated using Eq. (3).

The SS values calculated are given with the fine contents (FC) in Table 1. Table 1 shows that the SS increases with increasing fine contents, where FC is defined as the content of clay-silt fractions smaller than 0.075 mm.

3.3. Ideal specific surface area and background values of heavy metals

Fig. 6 shows relationships between SS and FC, using the grain size distribution curves shown in Fig. 4a and b. Two types of curves based on the experimental and smoothed grain-size distribution curves show different patterns resulting from the minimum grain size. The SS represented by the curve A'–E' corresponds to a smoothed grain size distribution, which is greater than the SS given by the curve A–D, based on the total experimental grain-size distribution. These SS values are examined later.

Table 1
Fine content and ideal specific surface area obtained from various grain size distributions

Sediment	A	A'	B	B'	C	D	C'	D'	E'
Fine content, FC (%)	96.5	95	88	80	69	54.5	43	28	0
SS for experimental curve (m ² /g)	1.465	–	1.212	–	0.906	0.548	–	–	–
SS for smoothed curve (m ² /g)	–	24.98	–	8.93	–	–	1.02	3.77	0.008

An example of calculation of the SS for individual groups is presented in Fig. 7, for which sample A' was used for the calculation. Fig. 7 shows that the group of smallest particles has considerably higher SS in comparison to other groups with larger particles. This means that the SS value of sediments is governed by smaller particles and the SS value for larger particles can be neglected. Therefore, abrupt drops in grain size distribution obtained using the Mie theory give relatively small SS values.

3.4. Theoretical background concentration

The intensity of reactions or phenomena on the surface of sediment particles depends on the area of the boundaries between the solid and seawater. The area of the boundaries can be expressed in terms of the specific surface area. Under a constant charge density and constant concentration of a substance, the amount of adsorption of an element may be assumed to be

proportional to the SS. Then,

$$B_{GP} \propto SS_{act} \quad (4)$$

Introducing a proportionality constant k_p , we can express this as:

$$B_{GP} = k_p SS \quad (5)$$

where k_p ($\mu\text{g}/\text{m}^2$) has dimensions of retained amount per unit area. If we can correlate SS with FC, a relationship between B_{GP} and FC can be obtained.

4. Estimation of background concentrations

4.1. Specific surface area and plasticity index

If the sorption capacity is proportional to the SS of particles, interactions between particles must be a function of SS. Fig. 8 shows correlation between SS and the plasticity index, I_p , for various marine sediments. In general, I_p varies with the interactions between particles, and is used for the classification of soils in geotechnical engineering. Although some scattering is evident in Fig. 8, it is apparent that I_p increases with increasing SS. Thus, SS can reflect variations in the interactions between particles.

4.2. Measurements

4.2.1. Measurement of metal concentrations

Various sediment samples from Seto Inland Sea and Osaka Bay, Japan, were obtained using a gravity tube corer and a grab sampler. The samples were dried at 110 °C. Dry sediment samples were digested according to the US EPA 3051 guideline: 0.1 g of sediment were digested in 2 mL of 65% HNO₃ and 0.6 mL of 48% HF in Teflon bombs using a microwave oven.

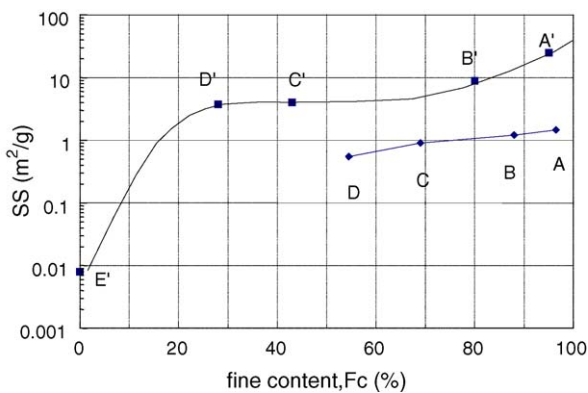


Fig. 6. Relationship between specific surface area and fines content.

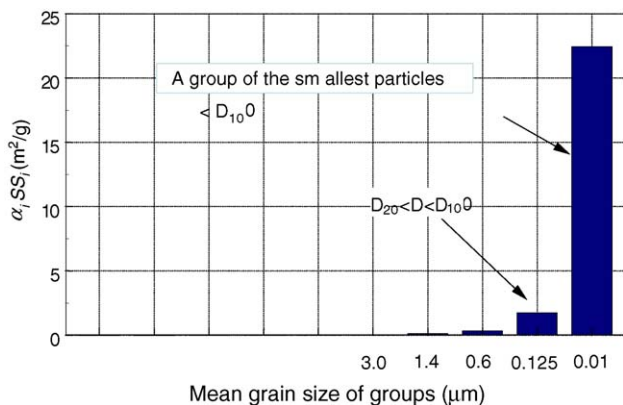


Fig. 7. Specific surface area in relation to the total weight of sample and mean grain size.

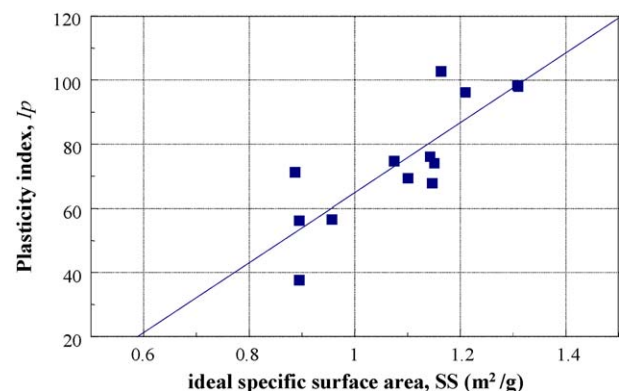


Fig. 8. Relationship between plasticity and ideal specific surface area.

Chemical analyses of lead (Pb), copper (Cu) and zinc (Zn) were carried out using inductively coupled plasma-atomic emission spectrometry (ICP-AES) after digestion.

4.2.2. Grain size analyses

For samples obtained using a tube sampler, the grain size distributions of samples were determined using laser and X-ray methods [14,15] for the reasons stated earlier. The X-ray extinction method for grain size analysis utilizes Stokes' law, as well as the conventional method. Therefore, the density of particles is required for the calculation of grain size and percentage fines. The settling velocity of sediment particles in a small cell (6 cm × 4 cm × 0.2 cm) filled with distilled water is measured using X-rays instead of light for the photoextinction method. The sample weight required is approximately 5 g. The percentage fines of the sample is calculated using the Lambert–Beer law for the relationship between the concentration of suspended solids and the intensity of X-ray extinction. However, this method cannot be used for carbon black, graphite and high-molecular-weight compounds. The measurement instrument can be automatically controlled.

On the other hand, the laser-diffraction scattering method utilizes the Fraunhofer diffraction theory for particle sizes greater than 0.001 mm, with the diffraction of particle reflection expressed as a function of grain size. A laser is used to irradiate a small cell (5 cm × 3 cm × 0.3 cm) filled with sample solution mixed with sediment particles and distilled water. The weight of sample required is approximately 0.05 g. For particles smaller than 0.001 mm, the Mie scattering theory is used. According to this theory, the scattering of light is a function of the index of refraction of particles. The index of refraction for soil particles is approximately 1.08.

4.3. Metal adsorption and background concentrations

Bay sediments often consist of a silty soil, which usually has FC greater than 80%. Fig. 9 shows typical metal concentration profiles for two cores obtained from two site locations in Osaka Bay, Japan. The FC of the sediment samples is 95 and 98% through from the top surface to the bottom portion. Therefore, we usually assume that the minimum concentration of Zn for these sediment cores is approximately 130 mg/kg, and that an approximate thickness of 50 cm from the top surface is contaminated with Zn. This implies that the deeper sediments are not contaminated. This minimum value obtained can be used as the background value of zinc at these site locations. Similar background concentrations of Zn were obtained in Tokyo Bay [16] and on the coast of Galicia (northwest Spain) [3]. On the other hand, the background values of Zn for sandy sediments were 55 ± 11 mg/kg [4]. The sand content of the sediments varied approximately from 50 to 80%.

In sediments, clay minerals are among the most important sorbents for metal cations. The adsorption of metals to clay particles can be explained by the Langmuir isotherm. According to the theory and experimental results [11,17], the adsorption of metals can proceed until the saturation of sorption occurs. This is one of the reasons why sediments can be contaminated by dis-

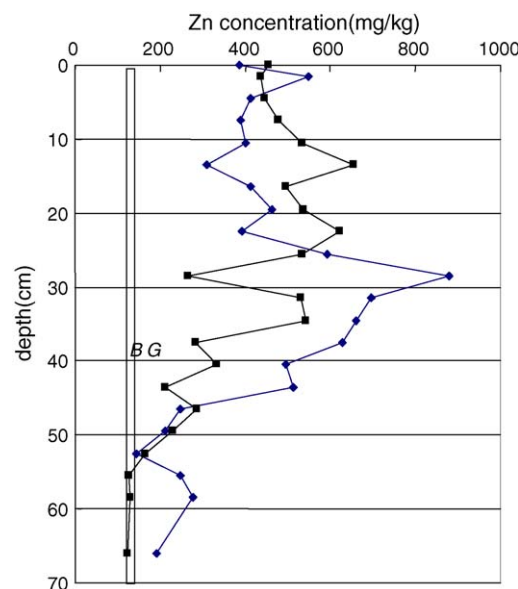


Fig. 9. Profiles of Zn concentration for fine sediments from two locations in Osaka Bay.

charge of metals through human activities. In fact, contaminated fine sediments can still show adsorption capacity if they are put in highly concentrated metal solutions [18].

The amount of metal adsorption is strongly dependent on the adsorption capacity of the sediment particles, the concentration of the metal solution and the pH value of the solution [11,17]. The pH of ordinary seawater and marine sediments is within a narrow range, i.e., between pH 6.5 and 8.0. If the concentration of a metal solution is high, the amount of adsorption is also high. Therefore, the background level is determined by the natural metal concentration of soil water or seawater.

Fig. 10 shows the general features of the amount of metal adsorbed under a wide range of equilibrium concentrations of metal solution based on the discussion above. In Fig. 10a, the amount of adsorption increases with increasing equilibrium concentration. Many studies have examined the characteristics of this type of adsorption theoretically and experimentally [11]. Under natural conditions, the equilibrium concentration of elements is relatively low, and more or less constant, indicated as a shaded zone in Fig. 10a. In this case, the higher the adsorption capacity, the higher is the amount of adsorption. However, if the equilibrium concentration is relatively low, the amount of adsorption for clayey soil may be constrained by the equilibrium concentration, as indicated by the shaded column in Fig. 10a, even if the soil has a high sorption capacity.

If adsorption capacity is relatively low, this can constrain the amount of adsorption, as shown in Fig. 10a. The state can be described as “saturation of adsorption”. For coarse sediments under a certain equilibrium concentration, the amount of adsorption is relatively low because of the small specific surface area. Thus, the background level of metal for sediments can be interpreted from two aspects, as follows:

- If a sediment has a relatively high adsorption capacity, B_G is constrained by the metal concentration (equilibrium concentration) in water.

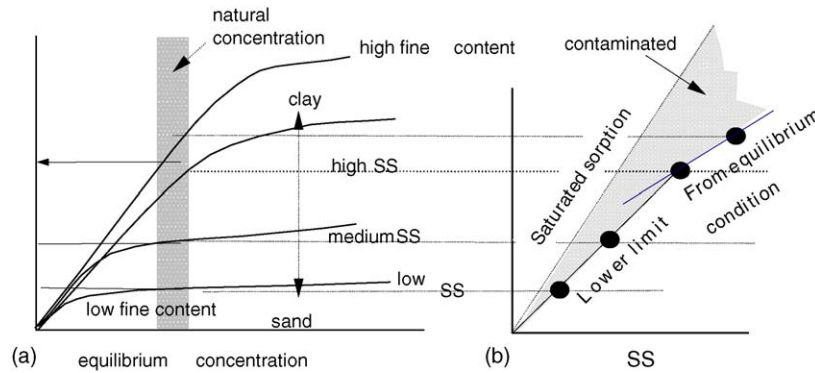


Fig. 10. Interpretation of background values based on the relationship between adsorbed metal concentration and equilibrium concentration in terms of sediment type. (a) Sorption characteristics of particles under a narrow range of natural equilibrium condition. (b) B_G -SS relation.

(b) If a sediment has a relatively low adsorption capacity, the equilibrium condition is nearly saturated with respect to adsorption. Therefore, B_G is strongly influenced by the adsorption capacity, which can be a strong function of grain size and its distribution. The grain size characteristics can be expressed by SS or FC.

These can be interpreted by the two different curves shown in Fig. 10b.

4.4. Concentration and FC diagram

In this study, FC is conveniently used instead of SS. This is because FC is usually obtained from the grain size analysis and because the concept of FC is familiar to scientists and engineers. For fine sediments, there are other factors more important than the grain size characteristics, as previously explained. Furthermore, if the background level is completely characterized, no further analysis is required.

Zn concentrations and FC are shown in Fig. 11, with data for both contaminated and non-contaminated sediments. Since the background value should theoretically be a minimum value, evaluation of the background level is achieved by obtaining the lower limit of the scattering data. In this study, a theoretical approach was taken, using the relationship between B_G and SS given by Eq. (5) and the relationship between FC and SS. The proportionality constant is determined from the best fit of the lower limit. If we use the curve A'-E' in Fig. 6, a proportionality constant k_p of $7.8 \mu\text{g}/\text{m}^2$ is obtained. On the other hand, if the curve A-D in Fig. 6 is used, the best fit gives a k_p value of $62 \mu\text{g}/\text{m}^2$.

The B_G values calculated using the k_p and SS values in Table 1 are indicated as solid and open squares in Fig. 11, corresponding to k_p of 7.8 and $62 \mu\text{g}/\text{m}^2$, respectively.

As seen in Fig. 11, the lines passing through the solid squares and open squares express almost the lower limit of plots, except for fines content higher than 90%. The lines obtained are called "background baseline (BB)" in this study. The BB of Zn is very small for sand or gravel and increases with increasing FC. The BB of Zn for FC between 20 and 60% is approximately $35 \text{ mg}/\text{kg}$. This range is slightly lower than the background

level for sandy sediments from the Galician Ria (NW Spain) [4].

Beyond a FC of 80%, the two curves start to part, as shown in Fig. 11. It seems that both lines deviate from the lower limit. This deviation may be due to the following reasons:

- (a) there might be an error in measurements of concentration or in calculation of the BB using the assumptions made in this study;
- (b) B_G is constrained by adsorption under equilibrium concentrations, which can be explained by Langmuir adsorption; and
- (c) the Mie theory may overestimate the grain size of sediments.

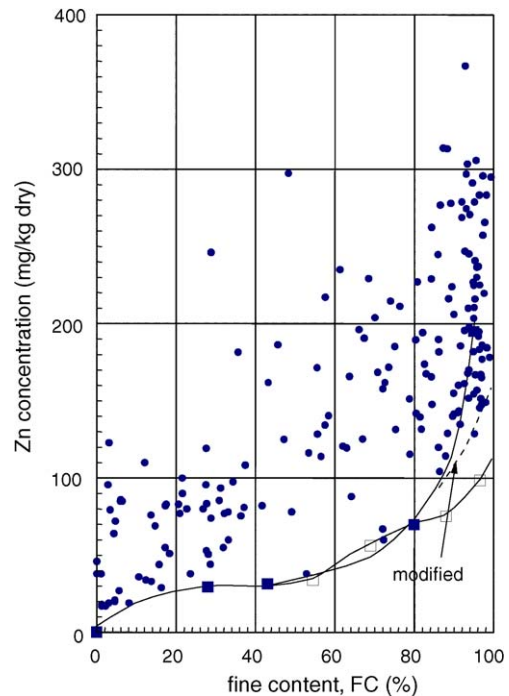


Fig. 11. Deviation of Zn concentration and background level for various marine sediments. Solid squares: k_p of $7.8 \mu\text{g}/\text{m}^2$ using the smoothed grain size distributions, Open squares: k_p of $62 \mu\text{g}/\text{m}^2$ using the grain size distributions from the Mie theory.

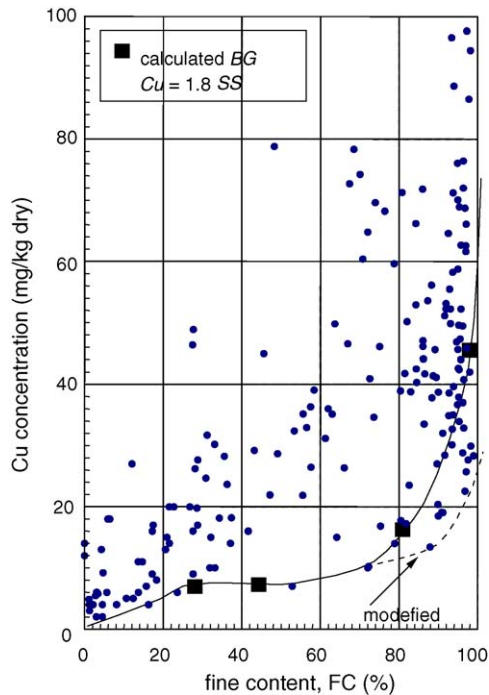


Fig. 12. Deviation of Cu concentration and background level for various marine sediments.

Item (c) is more realistic, because divergence of the grain size distributions is apparent between the two theories used. Therefore, the smoothed grain size distributions were used for the following analyses.

In this study, an empirical background level was defined for fine sediments with FC greater than $\sim 80\%$, as indicated by the modified line in Fig. 11. The modification is reasonable, because from the interpretation made previously, the increase in B_G with FC in this region should be lowered.

If we accept the background level of Zn, the degree of pollution can be obtained using Eq. (1).

Following a procedure similar to the case for Zn, background levels of Cu and Pb were determined, as shown in Figs. 12 and 13. Modified lines based on the empirical results obtained from Osaka Bay, Seto Inland Sea and Tokyo Bay [16] are also given in Figs. 12 and 13.

The results from Figs. 12 and 13 are summarized as follows:

- (1) Background levels based on the SS show good correlation with the actual data.
- (2) For fine sediments, modification of B_G for Cu and Pb is needed, based on the many reasons discussed.
- (3) The k_p value for both Cu and Pb is $1.8 \mu\text{g}/\text{m}^2$.

4.5. Background values

From Figs. 11–13, the background characteristics of sediments can be divided into four stages, as presented in Table 2. The sediments with B_G values are divided into four types, i.e., coarse, coarse–medium, medium–fine, and fine sediments. The boundaries between them are based on the BB shape. Note that the B_G values presented in Table 2 are approximated.

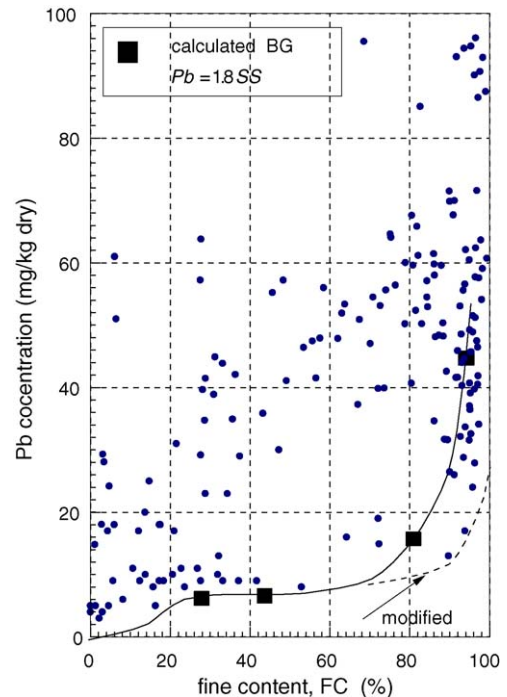


Fig. 13. Deviation of Pb concentration and background level for various marine sediments.

5. Practical application

5.1. Sediments from Seto Inland Sea

A contaminated sediment core was obtained from Kasaoka Bay in the Seto Inland Sea in Japan. A 140-cm-long core was taken by driving a stainless steel core tube into sediments. The tube has a length of 2.0 m and a sectional area of 100 cm^2 [19]. The top 30 cm of the core was cut into 3-cm sections, and each section was kept in a clean glass container until testing. In the laboratory, the grain size distribution and heavy metal concentrations of each section were determined. The grain size distribution of the samples was obtained using the techniques described above. Elemental analyses were performed by ICP spectrometry. The sample digestion method has already been described.

The sediments consisted of silty clay to clayey sand. The corresponding FC values varied from 95.3 to 55.5%. The water content of silty clay, i.e., the top sediments, was 183% and the ignition loss was 9.9%. The specific gravity of particles was 2.58. Sediments in this depth range are usually contaminated,

Table 2
Background levels estimated in this study

Name	FC (%)	B_G (mg/kg)		
		Zn	Cu	Pb
Coarse	<20	<35	<8	<8
Coarse to medium	20–60	35	8	8
Medium to fine	60–90	35–100	8–14	8–13
Fine	>85	100–150	10–30	10–30

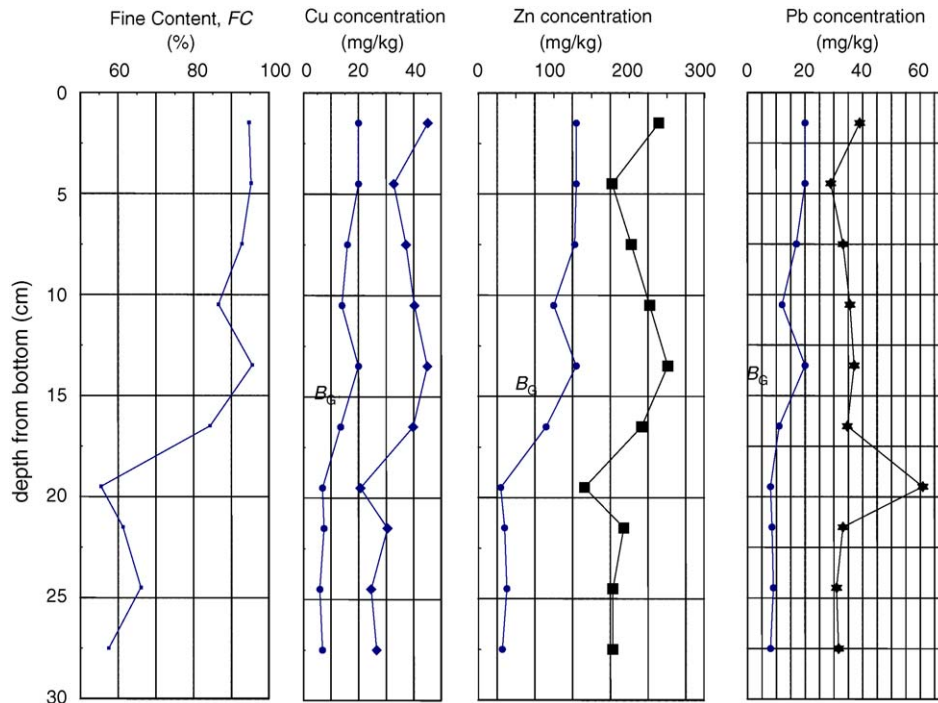


Fig. 14. Estimation of background values for a sediment profile in Seto Inland Sea.

but the degree of pollution cannot be evaluated without the corresponding background concentration.

Evaluations of the contamination are presented in Fig. 14. The background values for Zn, Cu and Pb were estimated using the fine contents and the relationships between FC and B_G shown in Figs. 11–13, respectively. The B_G value for Cu varied from 7 to 30 mg/kg, which is similar to values for Pb. Comparison of current concentrations and B_G values shows that all the sediment samples were contaminated, as shown in Fig. 14.

The lowest degree of pollution, defined previously, is 0.4 for both Zn and Pb, while the highest degree of pollution is 6.6 for Pb at a depth of 19.5 cm.

5.2. Clayey sediments

In this section, clay content is examined as an index property for the contamination of fine sediments. A sediment core sample obtained from Osaka Bay showed a very high FC, i.e., 98%, but the clay content varied with depth, as shown in Fig. 15; the clay content varied from approximately 30 to 70% with depth, and the lower sediments contain more clay fractions. Therefore, it might be expected that the lower sediments would contain more heavy metals. However, the results obtained show the reverse trend, as demonstrated in Figs. 16 and 17. This is because of contamination of the shallower sediments.

Fig. 16 shows the deviation of Cu concentration with clay content. If we take a constant and lower limit, a B_G value of approximately 30 mg/kg can be obtained, as indicated in Fig. 16. This value is comparable to that for FC of 98% in Fig. 12. It is important to note that this B_G value is provided for a FC of almost 100%.

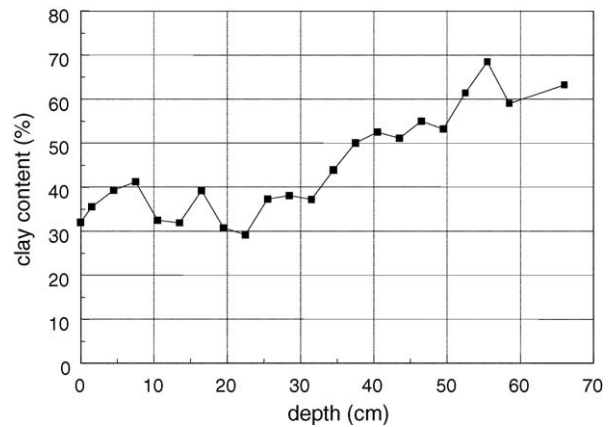


Fig. 15. Clay content with depth for a core sample obtained from Osaka Bay.

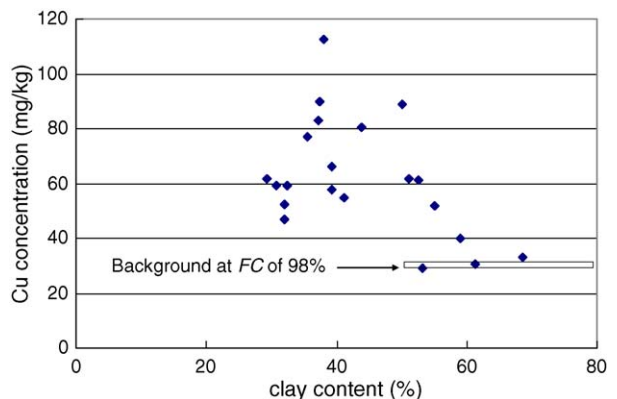


Fig. 16. Concentration of Cu according to clay content for Osaka Bay sediments.

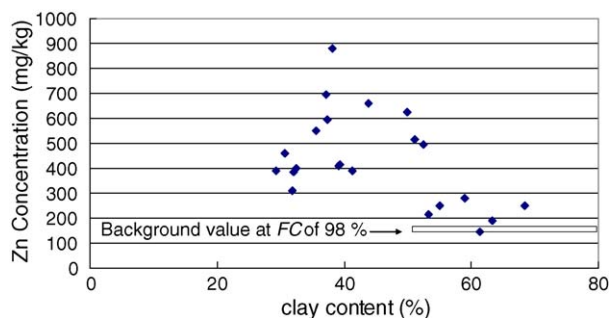


Fig. 17. Concentration of Zn according to clay content for Osaka Bay.

A similar situation can be cited for the case of Zn, as shown in Fig. 17. The Zn B_G value for this fine sediment was assumed to be 150 mg/kg, which is also obtained from Fig. 11. Therefore, the results from fine sediment core samples indicate that the B_G value near FC of 100% is considerably lower than that expected from the BB value, which was calculated using the specific surface area. This probably results from constraint by the equilibrium concentration, as explained above.

6. Concluding remarks

In evaluating the contamination of sediments, background values of metals have often been considered to be constant. On the other hand, normalization techniques using different elements have been used to estimate the background value.

In this study, a technique for estimating background values of some heavy metals in marine sediments is proposed using the grain size characteristics, i.e., fine content and specific surface area.

The examination shows that the background value, B_G , is expressed as a function of the ideal specific surface area, SS, defined in this study. The SS is correlated to fine content, FC. Finally, B_G values of Zn, Cu and Pb are defined in terms of FC, dividing the sediments into four types, i.e., coarse, coarse-medium, medium-fine, and fine sediments.

The B_G of Zn varied from almost zero to 150 mg/kg with FC. The B_G values for Cu and Pb varied from almost zero to approximately 30 mg/kg with FC.

The background values of other metals, such as cadmium, arsenic and chromium, can be determined with procedures similar to those proposed in this study.

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