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Relationship between Dieldrin Uptake in Cucumber and Solvent-Extractable Residue in Soil

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To prevent the distribution of cucumbers with dieldrin contamination exceeding the limit set by the Japanese Food Sanitation Law, the extraction solvent for dieldrin-contaminated soil was selected prior to cultivation so that the dieldrin residue level in cucumber could be predicted. The exhaustive extraction from soil could not explain the dieldrin uptake by cucumber plants. However, significant correlation (R^2 = 0.966, P < 0.001) was observed between dieldrin concentrations in cucumber and dieldrin concentrations extracted with 50% (v/v) methanol−water solution from soils. This was a result of the phytoavailability of dieldrin to the cucumber plants. The extractability of soil dieldrin with the methanol−water solution decreased as the organic carbon content in the soils increased. This suggested that a 50% (v/v) methanol−water solution is the optimal solution for predicting dieldrin concentrations in cucumbers by soil analysis.

KEYWORDS: Prediction; POPs; dieldrin; cucumber; soil

INTRODUCTION

Recently, 1,2,3,4,10,10-hexachloro-6,7-epoxy-1,4,4a,5,6,7,8,8a-octahydro-*endo*,*exo*-1,4:5,8- dimethanonaphthalene (dieldrin) has been detected in cucumbers produced in some Japanese agricultural areas, and its concentrations exceed the residue limit (0.02 ppm) set by the Japanese Food Sanitation Law (*1*, *2*). Therefore, to prevent the distribution of crops having contamination exceeding the limit, a method is required for the prediction of the dieldrin residue level in cucumbers before cultivation. In soil, dieldrin—a member of the group of persistent organic pollutants (POPs)—is known to form from the oxidation of 1,2,3,4,10,10-hexachloro-1,4,4a,5,8,8a-hexahydro-1,4:5,8-dimethanonaphthalene [aldrin (*3*)]. Aldrin and dieldrin were earlier used extensively as agrochemicals on arable land. They were not manufactured in Japan, but were imported over the period 1958−1972. During this period 3300 t of aldrin and 683 t of dieldrin were imported (*4*). The registration of these agrochemicals lapsed in 1973, and they have not been used in Japan over the past 30 years. However, dieldrin is still detected in soils due to extreme persistence (*1*).

In general, POPs are not transferred efficiently or rapidly to the above-ground tissue of plants via the root because they are strongly adsorbed to the soil as a result of their high hydrophobicity, which is apparent from their high *n*-octanol/water partition coefficient (K_{OW}) (*5*,*6*). Direct deposition from airborne compounds is the predominant source of POPs in plants (*7*), and their accumulation depends on the value of the *n*-octanol/air partition coefficient (K_{OA}) of compounds (*8*). However, plants belonging to the family Cucurbitaceae, such as cucumber, squash, pumpkin, and melon, have the ability to readily accumulate POPs such as dioxins in the above-ground tissues by absorbing them from soils via the root (*9*). Despite studies on the long-term fate of POPs in soil (*10*−*13*) and their bioavailability [e.g., to earthworms and microorganisms (*14*−*18*)] having been carried out since the last century, studies on the phytoavailability of POP residues in soil to Cucurbitaceae have been performed only in recent years (*19*−*25*). Otani et al. (*19*) found the differential phytoavailability of POPs among 34 cultivars of *Cucurbita* sp. grown in dieldrin-contaminated soil. By means of grafting tests, it has been shown that the type of plant root influences the uptake of POPs from soil (*20*−*22*). Therefore, soil amendments such as surfactants and low molecular weight organic acids have been tested to increase the phytoavailability of POPs (*22*, *23*). Moreover, the translocation of POPs within plant tissue has been explained on the basis of the analysis of a chlordane enantiomer fraction (*23*−*25*). Although the uptake mechanism of POPs in Cucurbitaceae has been studied, the prediction of the POP residue level has not been investigated.

Most techniques for the exhaustive extraction of soil have been developed from the viewpoint of the determination of total organic contaminant concentrations. In such cases, soils have been exhaustively extracted by very harsh techniques such as Soxhlet extraction. However, even if the total concentration is constant, the bioavailability can decline with time because the organic contaminants become sequestered in the soil (*17*). In addition, in the case of some POPs, the soil properties influence their bioavailability (*16*). Therefore, recently, nonexhaustive extractions have been investigated with the aim of developing a method that corresponds to bioavailability.

One extraction method that can be used for examining the bioavailability of organic contaminants in soil is solvent extraction. Kelsey et al. (*18*) compared the extractabilities of

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Table 1. Properties of the Tested Soils

soil sample	texture[a]	particle size analysis (%) sand	silt	clay	classification[b]	OC[c] (g/kg)	C/N[d]
A	clay loam	50	33	17	Typic Dystrochreps	20	12.6
B	clay loam	47	33	20	Typic Dystrochreps	16	10.8
C	sandy loam	69	17	14	Typic Dystrochreps	16	11.9
D	clay loam	61	22	17	Typic Dystrochreps	12	11.2
E	light clay	27	34	39	Typic Hapludants	55	11.2
F	light clay	25	35	41	Typic Hapludants	79	10.3
G	light clay	29	32	39	Typic Hapludants	69	12.3
H	light clay	44	28	28	Typic Epiaquups	30	22.2
I	sand	92	5.2	3.0	Typic Udipsamments	2.0	7.30

[a] According to the International Society of Soil Science (*28*). [b] According to USDA Soil Taxonomy (*29*). [c] Organic carbon content in soil. [d] Ratio of OC content to total nitrogen in soil.

compounds with different hydrophobicities by using a mild solvent to evaluate the bioavailability of the compounds to earthworms and bacteria. Krauss et al. (*14*) assessed the bioavailability of polycyclic aromatic hydrocarbons (PAHs) and polychlorinated biphenyls (PCBs) to earthworms by using exhaustive (*n*-hexane/acetone mixture) and nonexhaustive (methanol–water in the ratio of 1:1 and 0.5 M NaOH) extraction methods. With regard to phytoavailability, Tao et al. (*26*) found that the apparent accumulation of PAHs by wheat roots was correlated with the organic matter content in the soil. In a previous study carried out by the present authors, different phytoavailabilities for cucumbers grown in two different organic carbon (OC) soils were found (*21*). It is therefore important to elucidate the phytoavailability of dieldrin and to select the extraction solvent for examining the phytoavailability; an understanding of the phytoavailability is necessary for the development of a soil analysis method to predict the dieldrin residue level in cucumber before cultivation.

In this study, first, the dieldrin concentrations in cucumbers grown in contaminated soils were measured to assess the phytoavailability of dieldrin; the OC in the soils was also taken into consideration. Subsequently, the dieldrin extractability was determined using solvents (methanol–water solutions, acetone), and the relationship between the phytoavailability and extractability was investigated.

MATERIALS AND METHODS

Soil Samples. Soil samples were collected from the upper layers (0–15 cm depth) of nine cultivated fields in Japan. From every field, approximately 1 kg of soil was collected from five sites, and these samples were then composited (i.e., the representative sample for every field contained approximately 5 kg of soil). Precise records were not available, but these farm areas had received regular application of dieldrin for insect control from the late 1950s to the early 1970s. All collected soil samples were air-dried, passed through a 2 mm sieve, used for plant cultivation, and subjected to soil analysis. Soil particle sizes were analyzed using a soil analysis method (*27*). The soils were classified according to standards developed by the International Society of Soil Science (*28*) and using USDA Soil Taxonomy (*29*). The OC and total nitrogen (T-N) contents were determined using the dry combustion method and analyzed with an NC analyzer (Sumigraph NC analyzer, NC-900, Sumika Chemical Analysis Service Ltd., Osaka, Japan), and the water content was measured according to Japanese Industrial Standard specifications (*30*). The soil properties are given in **Table 1**.

Plant Cultivation. To measure the uptake of dieldrin from contaminated soils, a pot experiment was conducted in a growth chamber. Plastic pots (400 mL) were filled with different soil samples (290–530 g). The soil in each pot was fertilized with a chemical fertilizer [0.12 g of N as $(NH_4)_2SO_4$, 0.052 g of P as $Ca(H_2PO_4)_2H_2O$, and 0.10 g pf K as KCl] as basal dressing. Cucumber (*Cucumis sativus* L. cv. Sharp-1) seeds were germinated in perlite. After 2 weeks of growth, one seedling was transferred to each of the three pots containing different soil samples. The same methodology was used to grow cucumbers in a control soil sample with no detectable dieldrin. The pots were cultivated in an environmentally controlled cultivation room, and the plants were grown with a cycle of 14 h of light and 10 h of darkness at 25 °C. Each pot was set in a plastic tray for bottom-up irrigation with tap water and mulched with a silver plastic film sheet to avoid/minimize dieldrin contamination of the shoots by soil adhesion. During the growing period, an additional amount of fertilizer was applied twice to the soil in each pot as 100 mL of nutrient solution [1–2 times the strength of the Otsuka Chemical A prescription (mg/L): N, 130; P, 26; K, 168; Ca, 82; Mg, 18; Mn, 0.6; B, 0.25; Fe, 1.35; Cu, 0.015; Zn, 0.045; Mo, 0.015] by bottom-up irrigation. Twenty-one days after the transplantation, the shoots, including the leaves and stem, were harvested. The shoot samples were finely chopped and divided into two subsamples: one was dried at 70 °C to measure the moisture content and the other (approximately 10–20 g) was homogenized for 5 min in acetone. The solution was passed through an 0.8 μm glass fiber filter and concentrated to 50 mL for dieldrin analysis.

Exhaustive and Nonexhaustive Extraction Using Various Concentrations of Methanol. To determine the total dieldrin residue in the soil, 5 g of each air-dried soil sample was extracted with a Soxhlet apparatus and 130 mL of acetone for 16 h. This method is referred to as exhaustive extraction in this study. For nonexhaustive extraction, the dieldrin present in 8 g of triplicate soil samples was extracted by shaking with 40 mL of methanol–water solutions using a glass test tube. The test tube was placed on a horizontal shaker operating at 120 reciprocations per minute at a constant temperature. The effects of the extraction duration on the dieldrin extraction efficiency were first studied with 50% (v/v) methanol–water solution at a shaking temperature of 25 °C; the extraction contact time was varied: the initial duration was 10 min, and the subsequent durations were 1, 2, 3, 4, 6, 8, 10, 12, 18, 24, and 48 h for four samples (**Table 1**, samples A, C, E, and I). To examine the effect of the concentration of the methanol–water solution, various percentages [0, 10, 25, 50, and 100% (v/v)] of methanol were used for the shaking extraction at 25 °C for 24 h. The solutions were centrifuged at 1400*g* for 10 min and filtered through 0.8 μm filters.

Analysis of Plant and Soil Extracts. Analytical methods (purification and measurement) and quality control for dieldrin in soil and plant samples have been described previously (*20*). Briefly, portions of plant (10%) and soil (10–100%) extracts were taken and spiked with 100 ng of $^{13}C_{12}$-labeled dieldrin as internal standards. The extracts were transferred with 25 mL of *n*-hexane into a separation funnel and washed with water; excess water was removed with Na_2SO_4. The extract was concentrated by using a rotary evaporator and was purified with Florisil and a graphite carbon column. The sample was then spiked with $^{13}C_{12}$-labeled 2,2′,4,4′,5,5′-hexachlorobiphenyl, which was used as a syringe spike, and further concentrated to 50 μL under a gentle stream of nitrogen. The purified samples were analyzed using a gas chromatograph–mass spectrometer (GC-MS) (HP6890-5973N, Agilent Technologies, Santa Clara, CA) equipped with an ENV-8MS capillary column (30 m × 0.25 mm i.d.; 0.25 μm film thickness; Kanto Kagaku, Tokyo, Japan). The limits of quantitation (LOQs) were calculated according to the Japan Industrial Standard (JIS K 0312) (*31*). The LOQ values for dieldrin in the soils ranged from 0.15 to 2.68 ng/g of dry weight (dw). The LOQ values

Table 2. Mean Dieldrin Concentration in Soil and in Cucumber (n = 3)

soil sample	fw of shoot (g)	dieldrin concentration soil[a] (ng/g of dw)	dieldrin concentration cucumber[b] (ng/g of fw)	BCF[c] of dieldrin
A	37	363	141	0.39
B	35	89	47	0.53
C	42	31	21	0.68
D	37	46	17	0.37
E	36	460	102	0.22
F	42	278	77	0.28
G	39	325	68	0.21
H	37	90	49	0.54
I	21	43	48	1.12

[a] Exhaustive concentration in soil extracted with the Soxhlet apparatus using acetone. [b] Dieldrin concentration in young cucumber grown in each sample of soil. [c] Ratio of dieldrin concentration in cucumber to that in soil [(ng g^{-1} of fw in tissue)/ng g^{-1} of dw in soil].

for dieldrin in shoot tissue ranged from 0.53 to 1.23 ng/g of fresh weight (fw). The reference material for dieldrin in soil (JSAC 0441) obtained from the Japan Society for Analytical Chemistry was analyzed in triplicate for quality assurance. The value obtained was 73.0 ± 2.5 ng/g of dw (coefficient of variance = 3.54%), which was within the range of specified values (76 ± 14 ng/g of dw). The average recoveries of $^{13}C_{12}$-labeled dieldrin for the soil (n = 266) and plant (n = 30) samples were 83 ± 12 and 73 ± 9.1%, respectively.

Statistical Analysis. Linear regression and logarithmic regression analysis were carried out using SAS ver. 9.2 (SAS Institute Japan Ltd., Tokyo, Japan).

RESULTS AND DISCUSSION

Dieldrin Concentration in Soil and Cucumber Plants. The exhaustive extraction concentrations of dieldrin in the soil samples and the concentration of dieldrin in cucumber plants grown in the contaminated soils are shown in **Table 2**. The dieldrin concentration in the soils ranged from 31 to 460 ng/g of dw. Dieldrin was detected in all cucumber plants grown in the contaminated soils. The dieldrin concentration in the shoot ranged from 17 to 140 ng/g of fw, and the fw of the shoot ranged from 21 to 42 g (SD = 6.2, CV = 17%) for the 27 samples. For the plant grown on dieldrin-free soil in the same cultivation room, the dieldrin concentration in the shoot, if any, was below the LOQ. Therefore, differences in the dieldrin concentration among the plant samples depended upon the ability of the plants to take up dieldrin from the contaminated soils. Soil sample E showed the highest dieldrin concentration. On the other hand, the dieldrin concentration in the shoot grown in soil sample A was the highest. This confirms that the concentration of dieldrin in the cucumber plant did not correlate with the concentration in the soil determined by exhaustive extraction.

The bioconcentration factor (BCF) was calculated as the dieldrin concentration in the shoot divided by the exhaustive soil concentration (**Table 2**). Soil OC is known to reduce the bioavailability of hydrophobic organic contaminants in soil (*14*). **Figure 1** shows the relationship between the BCF and soil OC. This result indicated that soils with high OC (**Table 1**, samples E, F, and G) held dieldrin very strongly and restricted its transferability to the cucumber shoot even at high concentrations of dieldrin. Thus, the phytoavailability of dieldrin to cucumber plants cannot be explained by the exhaustive extraction of soil with different OC values. Therefore, a nonexhaustive extraction method for obtaining the extractability corresponding to the OC content was considered to be necessary for the evaluation of the phytoavailability through soils with different OC contents. Some studies have indicated that nonexhaustive extraction by an organic solvent can explain the bioavailability of organic contaminants in soil. Tang and Alexander (*32*) showed that the uptake of aged and unaged anthracene, fluoranthene, and pyrene by earthworms

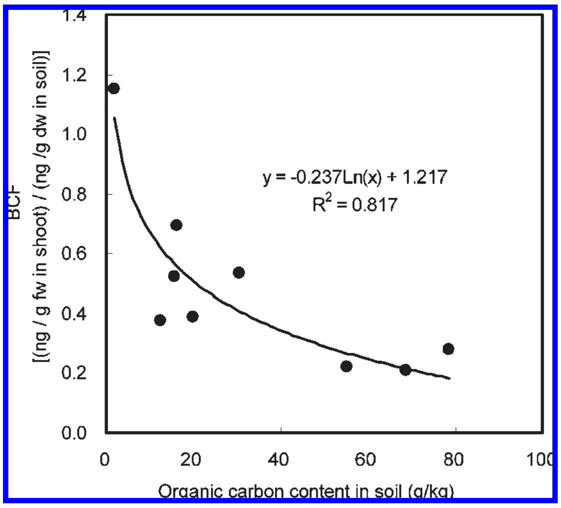

Figure 1. Relationship between bioconcentration factor (BCF) and organic carbon (OC) content. The BCF was calculated as the dieldrin concentration in the shoot divided by the exhaustive soil concentration.

was correlated with the amounts recovered from soil by extraction with n-butanol, propanol, and ethyl acetate. Kelsey et al. (*33*) used a variety of organic solvents and solvent combinations to extract atrazine and phenanthrene from soil to compare the proportions of these compounds that could be either mineralized by bacteria or accumulated by earthworms. Their findings suggest that atrazine bioavailability to bacteria and earthworms could be best predicted using 50% methanol−water (v/v) and 10% methanol−water (v/v) solutions, respectively. In this study, methanol−water solutions were focused upon for the nonexhaustive extraction of dieldrin from soils for phytoavailability.

Nonexhaustive Extraction of Soil Dieldrin by Methanol−Water Solutions. The difference in extraction efficiencies among soil samples and the effect of the extraction durations for 50% methanol−water (v/v) solution were examined by considering four soil samples with different properties. The result is shown in **Figure 2**. The y-axis shows the extraction efficiency, indicated by the extractability; it is calculated as the dieldrin concentration with 50% methanol−water (v/v) extraction divided by the acetone exhaustive extraction. The extractability for dieldrin was different for each soil sample. The difference was observed to be correlated to the soil OC. The extractability increased gradually when the contact time was increased to 6 h. The plateau observed for all samples at extraction durations above 12 h can be attributed to the equilibrium in the distribution of dieldrin between the liquid and soil phases. Therefore, for comparison of the phytoavailability and nonexhaustive extraction, an extraction

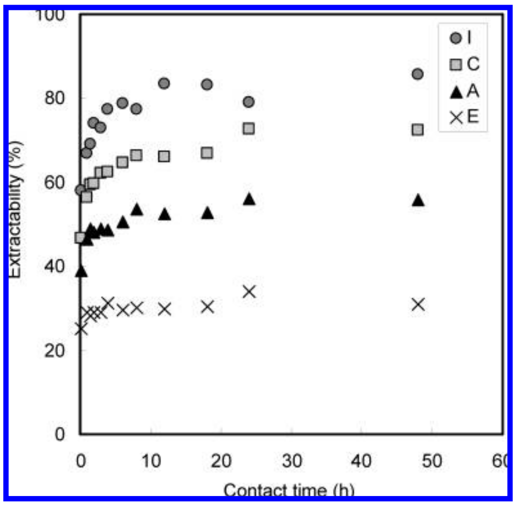

Figure 2. Time trend of the extractability of dieldrin in soil using 50% (v/v) methanol−water solution. The extractability of dieldrin was calculated as the concentration extractable with 50% (v/v) of methanol−water solution divided by the exhaustive soil concentration extracted with the Soxhlet apparatus using acetone. Soil sample (A, E, C, and I) information is provided in **Table 1**.

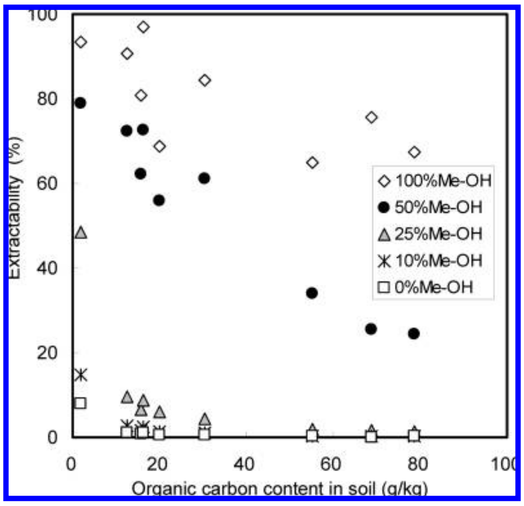

Figure 3. Relationship between OC content and extractability of dieldrin for various concentrations (v/v) of methanol−water solution. The extractability of dieldrin was calculated as the concentration extractable with the methanol−water solution divided by the exhaustive concentration of the soil extracted with the Soxhlet apparatus using acetone.

duration of 24 h was selected to avoid the effects of differences in soil property.

To examine the effect of the mixing ratio of methanol and water, the various solutions used for extraction were tested by shaking them for 24 h at 25 °C. The relationships between the extractability and OC content for each soil are shown in **Figure 3**. The extractability for all methanol−water solutions correlated with the OC content, as extractability decreased with increasing OC. The extractability for a 50% methanol−water (v/v) solution ranged from 24 to 79% in the soils. Differences in the extractability, namely, differences in the partitioning of dieldrin between the soil and liquid phases, were assumed to be caused by the differences in mixing ratio of methanol and water in the solution and differences in the OC content in the soils.

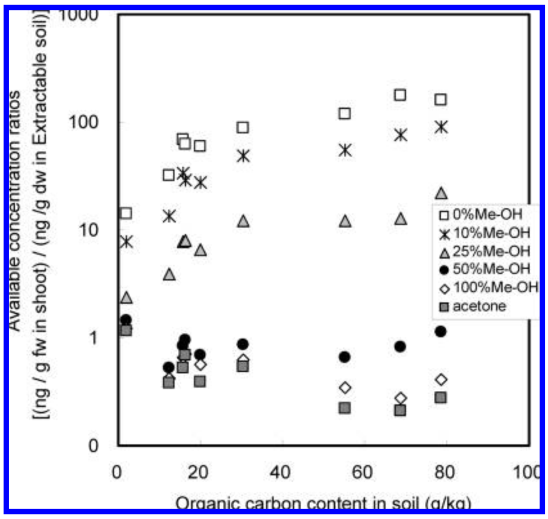

Figure 4. Relationships between OC content and the ratio of dieldrin concentration in cucumber shoots to the extractable soil concentration for various concentrations of methanol−water.

Relationships between Plant Concentration and Various Soil Extraction Concentrations. Both BCF calculated from exhaustive soil concentrations and extractability for all concentrations of methanol−water solution were correlated with the OC content in the soil; they decreased as the OC content increased (**Figures 1** and **3**). Therefore, some relationship was believed to exist between the cucumber shoot concentration and nonexhaustive concentration, which would make predicting phytoavailability possible in the case of nonexhaustive extraction. To identify the optimum concentration (v/v) of methanol−water solution for dieldrin extraction, the extractable soil concentration for each methanol−water solution and cucumber shoot concentration was calculated and compared with the soil OC (**Figure 4**). The *y*-axis plot was calculated as the "available concentration ratios" in the cucumber shoots divided by the extractable concentration in soil for 0, 10, 25, 50, and 100% (v/v) methanol−water solutions. The available concentration ratio for 0, 10, and 25% (v/v) methanol−water solutions and that for the 100% (v/v) methanol solution and acetone were >1 and <1, irrespective of the OC content of the soils, respectively. In the case of a 50% (v/v) methanol−water solution, the plot gives the most uniform assessment across soils with different OC contents as compared to the plots for the other solutions. It is implied that the phytoavailability of dieldrin to cucumber can be explained by soil extraction with a 50% (v/v) methanol−water solution.

Linear regression analysis was used to determine the correlation between the dieldrin concentration of the cucumber shoot and the dieldrin concentration in soil extractable by solvents. The regression lines were assumed to pass through the origin because if the extractable concentration is zero, the shoot concentration must also be zero (**Figure 5**). The extractable concentration when a 50% (v/v) methanol−water solution was used for 24 h at 25 °C was confirmed to be significantly correlated with the dieldrin concentration in the cucumber and is thus appropriate for estimating the uptake of dieldrin.

In this study, the correlation between the phytoavailability of dieldrin to early growth cucumber shoots and its extractability by methanol−water solutions was examined. Likewise, it was verified that soil extraction by a 50% (v/v) methanol−water solution is convenient and useful for accurately assessing

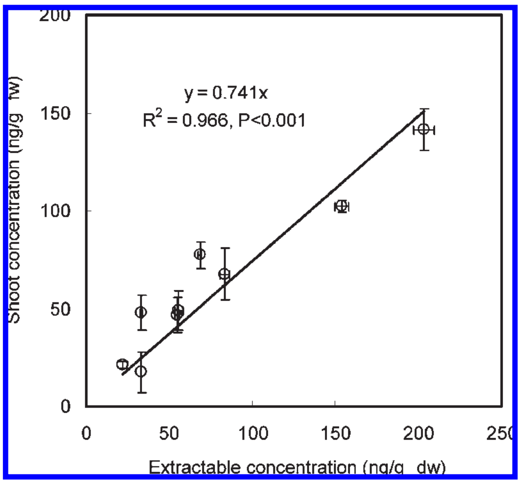

Figure 5. Relationship between dieldrin concentration in the cucumber shoot and the extractable concentration for a 50% (v/v) water−methanol solution. The points represent average concentrations ($n = 3$), and the error bars are the standard deviations.

the phytoavailability of dieldrin in soil. In previous studies, the authors reported on dieldrin concentrations in early growth cucumbers (*20*) and the shoots of mature plants and cucumber fruits (*21*) of grafted plants. The effect of different rootstocks on the dieldrin concentration in cucumber fruits was investigated. The result showed that the dieldrin concentration in the leaf and stem parts of mature cucumber plants was similar to the concentration in the fruits. However, Mattina et al. (*25*) indicated that chlordane accumulation in Cucurbitaceae fruits was a result of xylem transport. Furthermore, it was confirmed that PCB concentrations in pumpkin leaves and stems decreased with increasing distance above the root (*34*). Therefore, it is necessary to confirm the relationship between dieldrin concentration in soil extractable by a 50% (v/v) methanol−water solution and that in cucumber fruits to establish a method for the prediction of a contaminant residue level in cucumbers before cultivation. Such a prediction will help prevent the distribution of cucumbers exceeding the limit of contamination set by the Japanese Food Sanitation Law.

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