

Chemical Fractionation of Some Natural Radionuclides in a Soil Contaminated by Slags

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To investigate the chemical fractionation of ^{238}U , ^{226}Ra , ^{210}Pb , and ^{228}Ra in soils contaminated by slags from coal firing and from pyrite roasting, a sequential extraction method (modified Tessier procedure) has been applied. The following fractions were each extracted: I, easily exchangeable; II, bound to carbonates; III, bound to iron–manganese oxides; IV, bound to organic matter; V, persistently bound; VI, residual. In addition, the extractants were also analyzed for the insoluble matrix elements Al and Fe to provide some information on the effect of each extraction step on the dissolution of the matrix. The results show that the percentage amounts of these radionuclides in fractions I (<5%) and II (<10%) are quite low for all soil/slag mixtures. Significant differences in the chemical fractionation of the four radionuclides were however observed for the subsequent fractions III–VI. Redistribution processes of the radionuclides between the various solid constituents of the soil/slag mixture were examined for each extraction step by applying sequential extraction also to the uncontaminated soil and to the pure slags. In this way, it was possible to demonstrate that in a soil/slag mixture ^{238}U and ^{210}Pb released from the slag by the iron–manganese oxide extractant are subsequently reabsorbed rapidly to a considerable extent by soil minerals and thus do not appear in the iron–manganese oxide fraction III but rather in fractions IV and VI. As a result of such redistribution processes, it will be almost impossible to predict quantitatively the chemical fractionation of radionuclides in contaminated soils by investigating pure slags only.

Introduction

Slags from coal or from pyrite roasting are used to level hollows in the terrain and for the construction of farming and forestry roads. In addition, they are used as filling material during house construction, especially to improve the water drainage and the insulation along the lateral foundations. In rural and semirural areas, we also observed that in vegetable beds close to these houses—probably as a result of repeated spading—this soil is frequently mixed to a considerable extent with the underlying slag.

Because these slags, depending on their origin, can contain rather high levels of natural radionuclides from the uranium

and thorium series, it cannot be excluded that vegetables grown in these soils may take up these contaminants via their roots (1, 2). In addition, radionuclides eluted from the slags may eventually also reach the groundwater table and thus water used for drinking or irrigation. Because, however, these radionuclides are generally incorporated in the slag matrix, only a fraction of them can be expected to take part in ecological processes. Thus, to estimate a possible hazard from these radionuclides in the soil, not only their total concentration in the soil but also their chemical form has to be known. In addition, this information is needed when efficient remediation processes of contaminated soils have to be considered.

Various methods exist to examine the chemical fractionation of metals in the soil (3–11). At present, it seems that—based on investigations with heavy metals—the sequential extraction procedure of Tessier et al. (8) is most widely used for this purpose. With this method, the following fractions, although essentially only operationally defined, are usually obtained: I, easily exchangeable; II, bound to carbonates; III, bound to iron–manganese oxides; IV, bound to soil organic matter; and V, residual. In this way the chemical fractionation was investigated by numerous authors not only for heavy metals (for a review, see ref 11) but more recently also for artificial and natural radionuclides in contaminated soils (12–24), although to a much smaller extent, but not yet for slag-contaminated soils.

The purpose of the present paper was to investigate the chemical fractionation of ^{210}Pb , ^{238}U , ^{226}Ra , and ^{228}Ra from slags in the soil by using the modified Tessier procedure (additional extraction of persistently bound radionuclides with 7 N HNO_3 subsequent to the extraction of the organic fraction IV) (19, 14, 15, 18, 22, 23). For that, a non-contaminated soil was mixed uniformly with two different types of slags at the ratios 1:1 and 3:1. Sequential extraction was then applied to the non-contaminated soil, the slags, and the soil/slag mixtures. In this way, it was also possible to examine for each fraction and for each radionuclide whether its amount extracted from the soil/slag mixture is the sum of the amount extracted from the components (uncontaminated soil and slag). If this additivity is not observed, one has to conclude that a redistribution of this radionuclide during the extraction process has occurred. We demonstrate that in this way rather detailed information is obtained on the redistribution processes of these radionuclides in the soil/slag mixture during their extraction.

Recently, Clark et al. (17) have shown that valuable information can be obtained for the interpretation of the extracted phases if not only the contaminants but also naturally-occurring elements are analyzed in the extracts. For this reason, we determined in all fractions Al and Fe as typical insoluble matrix elements in the slags and in the soil.

Materials and Methods

Soil. The soil was a typical garden soil as used frequently for the horticulture of home-grown vegetables or flowers. Several physicochemical properties of this soil are also given Table 1.

Slags. Two types of slags were used: red slag, an iron rich residue from pyrite roasting during the process of cellulose production, and black slag, a residue from coal firing. Both slags were actually used for many years as refilling material. Their chemical compositions and the specific activities of ^{210}Pb , ^{238}U , ^{226}Ra , and ^{228}Ra are given Table 1. In order not to change the surface properties of the material with respect to their sorption/desorption properties, both

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TABLE 1. Chemical Characterization of Two Slags, Physicochemical Properties of the Soil, and Activity Concentrations of Natural Radionuclides in Slags and Soil^a

	black slag	red slag	soil
Al (%)	7.4 ± 0.7	2.3 ± 0.2	1.6 ± 0.1
Fe (%)	7.9 ± 0.7	62 ± 6	2.1 ± 0.2
Si (%)	16.5 ± 1	7.3 ± 0.7	
Mg (%)	1.3 ± 0.1	0.4 ± 0.04	
Ca (%)	7.1 ± 0.7	0.9 ± 0.1	
pH (KCl)			6.1 ± 0.1
organic matter (%)			1.9 ± 0.2
total N (%)			0.13 ± 0.01
clay %			9.6 ± 2
silt %			42 ± 2
sand %			49 ± 2
CEC (mol _c kg ⁻¹)			48 ± 5
²¹⁰ Pb (Bq kg ⁻¹)	440 ± 20	1510 ± 40	50 ± 20
²³⁸ U (Bq kg ⁻¹)	470 ± 140	1480 ± 200	40 ± 20
²²⁶ Ra (Bq kg ⁻¹)	500 ± 60	1770 ± 180	53 ± 4
²²⁸ Ra (Bq kg ⁻¹)	26 ± 14	45 ± 14	34 ± 4

^a The errors given correspond to one standard deviation.

slags were deliberately not crushed or sieved, but only the small fraction of particles > 5 mm was removed by hand. To prepare soil/slag mixtures, soil and either red or black slag were mixed in the ratios 1:1 and 3:1 according to weight and well homogenized.

Sequential Extraction. The sequential extraction of the radionuclides and the stable elements from the soil, the slags, and the soil/slag mixtures were performed according to Tessier et al. (8) with the following modification: Subsequent to fraction IV (bound to organic matter), the residue is extracted with concentrated nitric acid (7 M). This fraction will be called persistently bound (19, 14, 15, 18, 22, 23). The amount of material used was 100 g each (soil, slag, or soil/slag mixture). A total of 1 M MgCl₂ at pH 7 was used for the extraction of the first fraction. Separation of the liquid from the solid after each extraction step was achieved by passing the suspension first through a paper filter (Schleicher & Schuell, 589 Black, coarse grade, 9 cm in diameter) and finally through a membrane filter (1.2 μm).

Determination of the Radionuclides by γ-Ray Spectrometry. The total activity concentrations of the radionuclides in the solid samples and in the extracts were determined by direct γ-spectrometry. A total of 100–130 g dry solids was filled into cylindrical polyethylene vessel (75 mm in diameter, 30 mm height). Similarly, 500 mL of the extracts was filled in Marinelli beakers. The vessels were sealed gas-tight using insulating tape and stored for at least 2 weeks to allow radioactive equilibrium of the ²²⁶Ra series. After that time, the samples were measured for at least 1200 min, using a well-shielded low-level γ-ray spectrometer with a high-purity Ge-detector (33.9% relative efficiency at 1.33 MeV). Quality control over the accuracy of the data was assured by analyzing standard reference samples (e.g., NBS Coal SRM 1632a).

Determination of Stable Elements. Al and Fe in the extracts were determined by high-resolution ICP spectrometry. To determine these elements in the solid residue, they were first leached from the samples with aqua regia. Reagent grade chemicals were used throughout. In addition, any contacts of the soil or slag during sampling, storage, or processing the samples with materials which could release these elements were avoided.

Results and Discussion

Total Amounts of ²³⁸U, ²²⁶Ra, ²¹⁰Pb, and ²²⁸Ra in Slags and Soil. The total amounts of ²¹⁰Pb, ²³⁸U, ²²⁶Ra, and ²²⁸Ra in the two types of slags (red slag from pyrite roasting and black

slag from coal firing) are given Table 1. The largest activity concentrations for ²³⁸U, ²²⁶Ra, and ²¹⁰Pb from the ²³⁸U series were in the red slag (1480–1770 Bq kg⁻¹), followed by those in black slag (450–500 Bq kg⁻¹). Compared to that, the corresponding values in the garden soil were lower by a factor of about 30 for red slag, and of 9 for black slag. For each of the three types of samples, radioactive equilibrium between these three radionuclides was essentially attained. The activity concentrations of ²²⁸Ra (a decay product of the ²³²Th series) was quite low in all samples (26–40 Bq kg⁻¹).

Chemical Fractionation. The chemical fractionation of the four natural radionuclides, as obtained from the sequential extraction experiments, is shown in Figure 1 separately for soil, red slag, black slag, mixture red slag/soil 1:1, and mixture black slag/soil 1:1.

²³⁸U, ²²⁶Ra, and ²¹⁰Pb. These three radionuclides from the uranium decay series show a rather similar behavior in several, though not, all respects. In fraction I (easily exchangeable), the percentage amounts of these radionuclides are quite low (less than 5%) for all five sample types. The observation of low values for a given radionuclide in fraction I does, however, not necessarily mean that it is not available for root uptake. One should always keep in mind that fraction I yields only information on the quantity of ions which are "easily exchangeable" versus the counterion of the extracting solution. Thus, even when this amount is low, it cannot be excluded that special biological mechanisms (such as the mobilization of ions by root exudates, microbial activity, or mycorrhizal symbiosis) are able to increase the soil to plant transfer of an element.

The percentage amount of the four radionuclides in fraction II (bound to carbonates) are less than 10% and fairly similar for the five sample types.

Significant differences in the amounts extracted from the various samples types were however observed for fraction III (bound to oxides). In this case rather high values are found for ²¹⁰Pb and ²³⁸U but not for ²²⁶Ra extracted from red slag and black slag (30–40%). In contrast to that, the corresponding values for ²¹⁰Pb and ²³⁸U extracted from soil and from the two soil/slag mixtures were much lower (around 10%).

In fraction IV (bound to organic matter), low values for the amounts extracted were found for all three radionuclides in the case of red slag (<10%). These values increased to about 20% in the case of black slag. Because the two slags did not contain any organic matter, this demonstrates that an extraction using acidic hydrogen peroxide obviously is able to extract these radionuclides to some extent also from inorganic material, especially from black slag. The amount of ²¹⁰Pb in fraction IV extracted from the soil is rather high (about 40%) and thus significantly larger than the corresponding values for ²³⁸U and ²²⁶Ra (about 20%). This suggests that, in the uncontaminated soil, Pb is associated to a considerable extent with soil organic matter, probably as a fairly strong complex with humic materials. For ²³⁸U and ²²⁶Ra, these complexes are obviously less strong. Rather high values (30–40%) were observed in fraction IV when these three radionuclides were extracted from the mixture soil/black slag and from the mixture soil/red slag (about 20%).

The amounts of these radionuclides found in fraction V (persistently bound) depend strongly on the sample type. While only small amounts were extractable with concentrated nitric acid from the uncontaminated soil (about 10%), much larger amounts were released from the two slags and the two soil/slag mixtures (30–40%). Quite high values of these radionuclides were also present in fraction VI (residue), especially for the uncontaminated soil (up to 60% for ²²⁶Ra). The corresponding values for red slag and black slag were smaller (about 30% and 20%, respectively). The values for

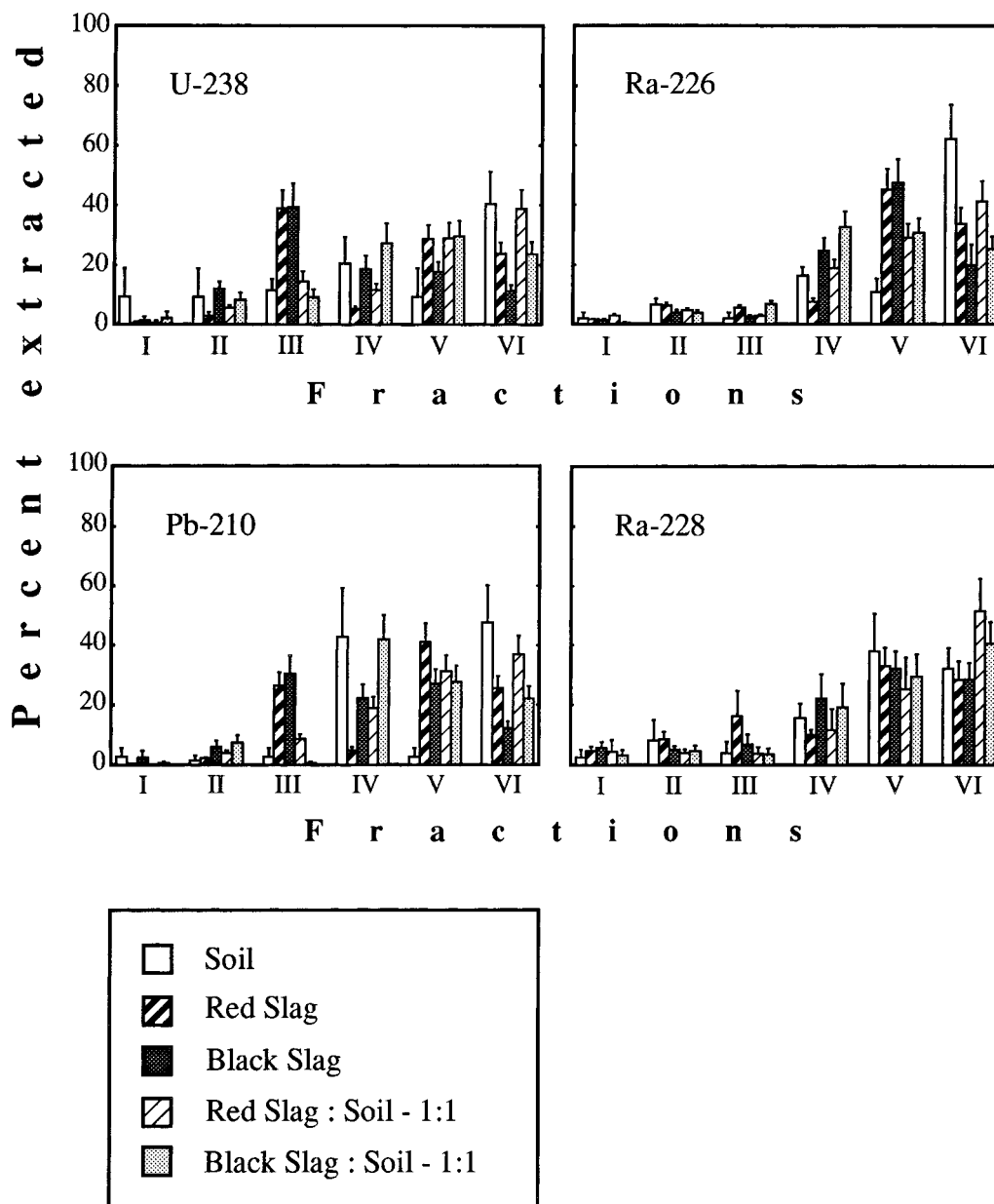


FIGURE 1. Percent distribution of natural radionuclides among the fractions: I, easily exchangeable; II, bound to carbonates; III, bound to iron–manganese oxides; IV, bound to organic matter; V, persistently bound; and VI, residual for uncontaminated soil, red slag, black slag, red slag/soil mixture 1:1, and black slag/soil mixture 1:1.

mixtures soil/red slag and soil/black slag were in between for those of the uncontaminated soil and the corresponding slags.

The above values for the uncontaminated soil may be compared with investigations by Lima and Penna-Franca (16), who determined the chemical fractionation of ^{226}Ra in Brazilian soils in areas with a high natural background activity by sequential extraction. Their results yielded quite similar values as observed by us and demonstrate also that this radionuclide is predominantly found in the residual fraction.

^{228}Ra . Even though one would expect that ^{228}Ra should exhibit a similar chemical fractionation as ^{226}Ra , this is not always the case. Appreciable differences are found for these radionuclides in fraction V (persistently bound) of the uncontaminated soil, which indicates that ^{228}Ra is four times more extractable with nitric acid from soil than ^{226}Ra (40% versus 10%). As a consequence, ^{228}Ra in fraction VI (residual) of the uncontaminated soil is much smaller than that observed for ^{226}Ra (30% versus 60%). This suggests that ^{232}Th ,

the parent radionuclide of ^{228}Ra , is in this soil in a form that is more susceptible to nitric acid attack and thus present in different minerals than ^{238}U , the first member of the decay chain of which ^{226}Ra is a member. The opposite behavior is observed for the two slags where in fraction V more ^{226}Ra than ^{228}Ra is found. This is, however, not surprising because these materials are also not homogeneous and acid attack can, therefore, release different amounts of these isotopes. In contrast to these differences observed for the pure soils and slags, no significant differences were present for these two Ra isotopes in fraction V of the two soil/slag mixtures (Figure 1, right side). At present, however, it cannot be decided whether this is generally the case or observable only for the materials investigated here.

Deviations from Additivity. From the amount of a radionuclide found after extracting it from the pure slag and from the uncontaminated soil, it should be possible to calculate the corresponding amount for each fraction of a soil/slag mixture. For that, we assume as a first approxima-

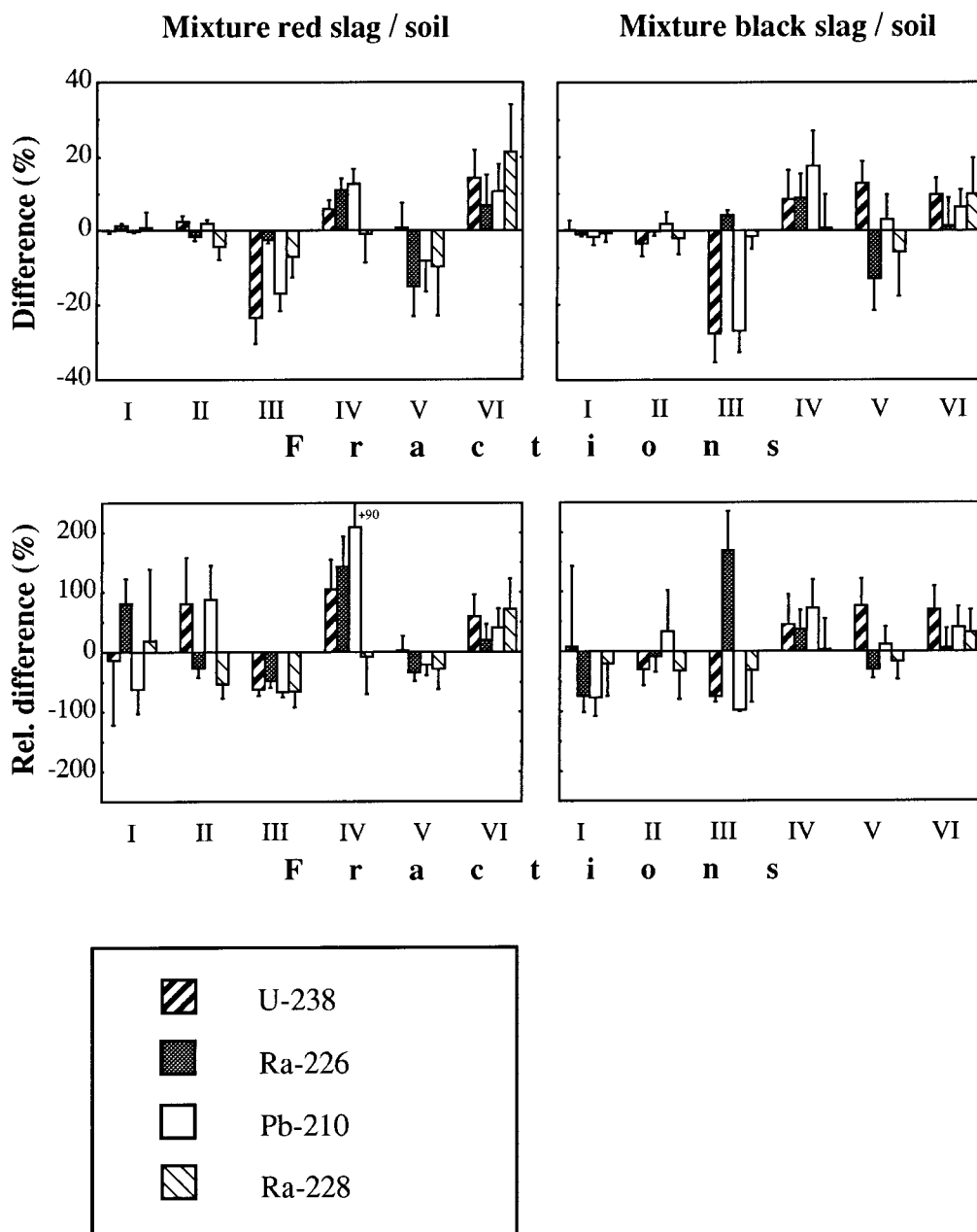


FIGURE 2. Comparison of percentage amounts extracted of the radionuclides from slag/soil mixtures with the corresponding values calculated by adding the amounts extracted observed for pure slag and uncontaminated soil. (Top) Difference of *observed* percentage amounts extracted minus *calculated* percentage amounts extracted (see eq 1). (Bottom) As above, but with respect to the calculated percentage amount extracted into each fraction (see eq 2). Notation of fractions I–VI as given in Figure 1.

tion, additivity, i.e., the amount of a radionuclide extracted into a given fraction is the sum of the corresponding values observed for the two components (i.e., pure slag and uncontaminated soil, respectively). This assumption is however only justified if no redistribution occurs, i.e., a radionuclide extracted from slag into a given fraction is not partially reabsorbed by one or more of the soil constituents (or vice versa) and will consequently not appear entirely in this particular extract. Thus, by comparing for a mixture the observed amount of a radionuclide in a given fraction with the corresponding calculated value obtained by assuming additivity, it is possible to obtain information on the presence and the extent of redistribution processes. For this reason, we calculated for the 1:1 soil/slag mixture *for each fraction* I–VI the difference (D) between the observed and the calculated value of the radionuclide according to

$$D(\%) = p_{\text{obs,mixture}}(\%) - p_{\text{calc,mixture}}(\%) \quad (1)$$

and

$$D_{\text{relative}}(\%) = D(\%) / p_{\text{calc,mixture}}(\%) \times 100 \quad (2)$$

where $p_{\text{calc,mixture}} = (a_{\text{soil}}W_{\text{soil}}p_{\text{soil}} + a_{\text{slag}}W_{\text{slag}}p_{\text{slag}}) / (a_{\text{soil}}W_{\text{soil}} + a_{\text{slag}}W_{\text{slag}})$ when a is the specific activity of the sample (soil or slag) (in Bq/kg), w is the weight of the sample (soil or slag) in the mixture (in kg), and p is the percentage amount extracted for each fraction.

The value of D , as obtained from eq 1, reveals to which extent the percentage amount of a radionuclide extracted from the mixture deviates for each fraction from ideal additive behavior (i.e., no redistribution) and is useful to examine the *amount of the radionuclide transferred by redistribution processes* from one fraction to another. Because no

radionuclides are lost or created by redistribution processes, the sum of the D values over all six fractions always has to be zero. Thus, when a negative deviation from additivity is observed for a radionuclide in one fraction, one or more corresponding positive deviations have to be present in the other fractions.

The value of D_{relative} shows to which extent this redistribution modified the amount extracted *with respect to ideal additive behavior for that particular fraction*. To examine in this way the extent and the effect of redistribution for each radionuclide during the extraction process, both quantities, D and D_{relative} , as well as the corresponding 1σ standard deviations, are given in Figure 2.

²³⁸U, and ²¹⁰Pb. Figure 2, top, shows that negative values of D are observed for ²¹⁰Pb and ²³⁸U in fraction III (bound to Iron–manganese oxides) of the mixtures red slag/soil and black slag/soil. This demonstrates that these radionuclides are readsorbed after their extraction with hydroxylamine to a considerable extent by other soil constituents still present in the mixture (i.e., constituents of the fractions IV–VI). ²¹⁰Pb and ²³⁸U released from fraction III of the mixture and readsorbed by other fractions originate in this case most likely from the slags and not from the soil, because these radionuclides are originally present essentially in the slags only (see Table 1). Next, the positive values observed in subsequent steps of the sequential extraction for D in the case of ²³⁸U and ²¹⁰Pb (Figure 2, top) reveal that these radionuclides were readsorbed by fractions IV and VI; ²³⁸U in the mixture black slag/soil also by fraction V. Similarly, a small amount of ²¹⁰Pb extracted from the mixture red slag/soil with nitric acid (fraction V) was obviously readsorbed by fraction VI (negative value for D in Figure 2, top left, and corresponding positive value in fraction VI).

Even though the extent of radionuclide redistribution, as characterized by the positive and negative values of D shown in Figure 2, top, is usually less than 20%, this effect produces much larger deviations from the ideal behavior of a mixture when its consequence with respect to the amount of a radionuclide extracted is considered. This quantity, D_{relative} , as obtained from eq 2, is shown in Figure 2, bottom. It illustrates, for example, that D_{relative} for ²¹⁰Pb is not only quite large (about 100%) in fraction III (where D is also large) but also in fractions I and II, because in these fractions the total amount of Pb is rather low and thus sensitive to small redistribution effects.

²²⁶Ra. In contrast to the redistribution behavior of ²³⁸U and ²¹⁰Pb, the radionuclide ²²⁶Ra exhibits a somewhat different pattern. Figure 2, top, shows that for both soil/slag mixtures ²²⁶Ra shows first positive D values for fraction IV and subsequently negative D values for fraction V. This cannot be explained by redistribution processes (because negative D values were not observed for the preceding fractions), but indicates that the peroxide treatment used to extract fraction IV releases from both soil/slag mixtures more ²²⁶Ra than expected from the extraction of the individual components slag and soil. This could be explained by realizing that during the extraction ²²⁶Ra from uncontaminated soil somewhat more hydrogen peroxide is added than actually required to disintegrate all soil organic matter, while during the extraction of this radionuclide from pure slag all hydrogen peroxide added will be consumed to attack the slag and release some ²²⁶Ra. Thus, in the soil/slag mixture the excess of hydrogen peroxide not used for the disintegration of soil organic matter is now available for an additional disintegration of the slag. As a consequence, the release of additional ²²⁶Ra from the mixture seems possible.

²²⁸Ra. According to the observed values for D in Figure 2, top left, this radionuclide from the ²³²Th decay series is readsorbed to some extent by fractions VI after it has been

TABLE 2. Percentage Fractions of Fe and Al in fractions I–VI Extracted from Uncontaminated Soil, Red Slag and Black Slag^a

sample	I	II	III	IV	V	VI
Soil						
Fe	<0.1	0.1 ± 0.05	14 ± 0.7	9.6 ± 0.5	66 ± 1.3	9.9 ± 0.5
Al	<0.1	0.5 ± 0.1	2.4 ± 0.1	16 ± 0.8	55 ± 1.2	26 ± 1.0
Red Slag						
	<0.1	0.1 ± 0.05	5.1 ± 0.2	0.2 ± 0.08	36 ± 1.0	58 ± 1.3
Fe	<0.1	0.5 ± 0.1	14 ± 0.7	3.7 ± 0.2	74 ± 1.5	7.6 ± 0.4
Black Slag						
Fe	<0.1	0.2 ± 0.08	14 ± 0.7	2.6 ± 0.1	43 ± 1.1	40 ± 1.1
Al	<0.1	0.1 ± 0.05	20 ± 0.8	10 ± 0.5	49 ± 1.1	20 ± 0.8

^a The errors given correspond to one standard deviation.

released by extraction of fraction III and V from the red slag/soil mixture.

Soil/Slag Mixtures with Other Ratios. As mentioned, the sequential extraction of the radionuclides was not only performed for 1:1 but also for 3:1 soil/slag mixtures. These results confirmed essentially all the above conclusions obtained already from the 1:1 mixtures discussed above and are therefore not reproduced here.

Comparison with Matrix Elements. As demonstrated recently by Clark et al., analyzing the extracts not only for the contaminants but also for naturally occurring stable elements in the matrix can provide valuable information on the extraction process. For this reason, we determined in the extracts of the two slags and the soil also Fe and Al, because these elements were the predominant metal constituents (see Table 1). In this way it should be possible to reveal to which extent the matrix of the slags and the soil is attacked by the various extractions of the modified Tessier procedure. Table 2, where these data are given, shows that in both slags Al and Fe are essentially found in the extracts of fractions V and VI, which should be characteristic for insoluble elements. Some Al and Fe however were present also in fraction III (5–20%) of both slags, and some Al (10%) was present in fraction IV of the black slag. Compared to that, considerably higher values are found in fraction III for ²¹⁰Pb and especially for ²³⁸U (up to 40%, see Figure 1). This suggests that these two radionuclides are under the reducing conditions of the iron–manganese oxide extractant more labile than the highly insoluble matrix elements. For ²²⁶Ra, this seems not to be the case.

In the extracts of the uncontaminated soil, Al and Fe are also predominantly in fractions V and VI, with small amounts of Al and Fe in fraction IV (10–16%) and in fraction III (14% Fe, 2% Al). These data can again be compared with the corresponding quantities found for the four natural radionuclides. As evident from Figure 1, the percentage amounts of the radionuclides, especially of ²¹⁰Pb in fraction IV, is considerably higher. This indicates that by acidified peroxide ²¹⁰Pb is not released by chemical attack of the soil matrix but rather from soil organic matter.

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Received for review May 22, 1997. Revised manuscript received October 21, 1997. Accepted October 22, 1997.[®]

ES970446O

[®] Abstract published in *Advance ACS Abstracts*, December 15, 1997.