PRELIMINARY SOIL CHARACTERIZATION FROM RADIONUCLIDE CONTAMINATED SITES: WHAT FIRST? WHAT LATER?

William S. Richardson, III and Scott S. Hay
Sanford Cohen and Associates, Inc.

Michael C. Eagle and Clinton Cox
U.S. Environmental Protection Agency

ABSTRACT

As part of treatability studies of radionuclide contaminated sites, total characterization to evaluate the feasibility of volume reduction by physical separation is an intricate process, consisting of multi-step protocols designed to identify differences between clean and contaminated particles that can be exploited for remediation. It is possible, however, to select preliminary, specific soil characterization tests that represent the more probable and less costly options.

Selecting a likely option is based on the finding that radionuclide contaminants are often associated with small soil particles (fines). In addition, some contaminated soils contain discrete-sized artifacts. Thus, size separation is a good candidate for volume reduction; and separation, accompanied by radioanalysis of resulting fractions, is a relatively simple and inexpensive procedure. It can be achieved by wet sieving, complemented by vertical-column hydroclassification, a procedure that simulates many plant-scale processes. It has the additional attractive feature of being easier to scale up the results to plant level than many other separation technologies.

Should characterization reveal that particle-size separation might be applicable, the next step depends on the recovery of clean material. If separation produces a large recovery, the second tier of treatability studies, bench-scale testing or process development, would be indicated. If separation produces a marginal recovery or if remediation by particle-size separation is not feasible, further soil characterization would be required to identify other exploitable physical properties for separation; or at this point, a decision should be made to determine if it is economically feasible to continue with soil characterization, that is, are adequate funds available to continue.

For further characterization studies, the question of the next step is based primarily on the source of contamination, knowledge of the sample, and the potential of chemical extraction as a reasonable, alternate remediation candidate to other, more expensive methods. Ore-based (natural) or artificial contamination should be examined by petrographic techniques. Petrographic characterization may reveal the presence of an offending radiomineral with a unique property that might be used for separation. Should the contamination be anthropogenic with sufficient background information to indicate a specific contaminant, identifiable physical properties of soil or contaminant particles should be evaluated before petrographic analysis in an attempt to identify alternate physical processes that might be applicable. The chemical extractability of contaminants could also be evaluated at this time. Select reagents should be considered in scout tests. These chemical tests usually can be performed in a preliminary characterization study in a brief time period at reasonable cost.

INTRODUCTION

As part of site treatability studies, total soil characterization to evaluate the feasibility of volume reduction by physical separation is an intricate process. The approach generally consists of multi-step testing protocols designed to identify those differences in physical properties of clean and contaminated particles that may, in turn, suggest a field-scale volume reduction process and be exploited for site remediation. There are numerous physical properties that may be exploited: size, density, effective shape, magnetism, friability, solubility, surface properties (especially with specific flotation agents), and radionuclide concentration.

In the treatability field, there is often limited funding for obtaining information to be used in planning for site remediation. As a result, the determination of what to do first in soil characterization or what method will most likely be successful in reducing the volume will be important. It is possible, however, to select specific soil characteristic tests, based on typical properties of radionuclide contaminated soils, that represent more probable and less costly options for accomplishing volume reduction by physical separation. Thus, in an effort to obtain the most profitable information early in the treatability study, it is possible to suggest an answer to "what first" when it applies to soil characterization.

MOST PROBABLE SEPARATION OPTION

Basis for Option

Selecting a likely separation option for radionuclide contaminated soils is based on the finding from examination of soils from numerous sites (1,2,3) that radionuclide contaminants, primarily metallic ions, are associated in many instances with small particles, usually less than 300 microns (50 mesh) in diameter. The results from particle-size characterization of a soil sample from the Montclair, New Jersey, Superfund site illustrates this finding. Table I demonstrates the general increase in radium-226 concentration as particle size decreases in a whole-soil sample containing 40 pCi/g radium-226 (4). Table II is another illustration of the same finding provided by the results of size separation of a soil sample from the Maywood, New Jersey, Superfund site.

Note the high concentration of radium-226 in the fraction smaller than 37 microns (400 mesh): the concentration is over twice that of the whole soil. These results, complemented by

TABLE I
Particle Size and Radionuclide Distribution of a Montclair Sample

Particle Size (mm)	Weight Percent (%)	Ra-226 Activity[1] (pCi/g dry)
+6.35	12.2	7.0
-6.35/+1.19	6.3	19.2
-1.19/+0.297	11.6	8.1
-0.297/+0.149	12.2	12.3
-0.149/+0.074	14.1	12.9
-0.074/+0.037	12.9	21.8
-0.037	30.7	85.8

[1] Activity of whole soil is 39.9 pCi/g.

bench-scale testing and process development, resulted in the design and testing of a pilot plant that successfully separated from the Montclair test soil a small-size particle fraction, less than 76 microns (200 mesh), from the cleaner fraction that had an average radium-226 concentration of less than 11 pCi/g, a value within the 5/15 pCi/g clean-up criteria established by the Record of Decision (ROD). The cleaned fraction represented over 55 percent of the whole soil.

The inverse relationship between particle size and radioactive concentration is primarily the result of two characteristics. One is the attraction of radionuclide cations to the surface of clay particles, small soil particles (< 2 microns) with negatively charged surfaces that provide a site for the exchange of radionuclides for native cations (5). The other characteristic is the increasing surface-to-volume ratio as the size of particles decreases, resulting in more surface area on which absorption may occur. In addition to these two characteristics, some contaminated soils contain discrete-sized artifacts from fission, activation, or ore-processing. Examples of artifacts are weapons-fired plutonium, radiobarite, or radium buttons that may be found in a particular soil fraction, large- or small-sized. Particularly, many precipitates or co-precipitates, such as radiobarite, will usually be located in the small-sized fractions, since they form as a fine solid. Thus, particle-size separation is a very good candidate for remediation by volume reduction, permitting the isolation and collection of the highly active fraction or fractions. Also, size characterization, accompanied by radionuclide analysis of the resulting fractions, is a simple and relatively inexpensive laboratory procedure.

SEPARATION PROCESS

Soil characterization for particle-size analysis may be achieved by wet sieving or vertical-column hydroclassification. Before separation by either process, it is important to mix the soil with water in a manner that will ensure effective particle liberation. Liberation removes small particles from larger ones and breaks apart aggregates, often releasing smaller, trapped soil particles or artifacts. Effective liberation before separation is especially important with radionuclide-contaminated soils, since the small particles usually contain a higher concentration of contaminants. The method used for liberation must be chosen to assure release of most of these offending particles from the larger, usually contaminated ones. Alternately, it is important during liberation not to generate excessive small material (fines) that will diminish the recovery of larger, less-contaminated product and concomitantly contribute low activity particles to the small, non-remediated material. Vigorous washing, a process that rapidly mixes the soil in a suspension of water at about 25 percent solids, has been very effective in particle liberation while producing minimum fines (1).

Wet sieving separates the washed soil particles by size. The aqueous process is more effective than dry sieving and has the added advantage of eliminating the suspension of small, radioactive soil particles in the air. This could maximize the applicability of any proposed treatment process by reducing the potential for airborne exposure to on- and off-site personnel. The fractions produced are dried, weighed, and analyzed for appropriate radionuclides. The particle-size and

TABLE II
Particle Size and Radionuclide Distribution of a Maywood Soil Sample

		Activity[1]		
Particle Size (mm)	Weight Percent (%)	U-238 (pCi/g dry)	Ra-226 (pCi/g dry)	Th-232 (pCi/g dry)
+6.35	2.4	2.0	4.5	1.7
-6.35/+1.19	6.6	5.7	6.9	12.5
-1.19/+0.60	4.0	4.1	5.0	10.4
-0.60/+0.25	10.0	1.2	1.9	2.4
-0.25/+0.15	4.9	1.8	8.8	5.9
-0.15/+0.106	8.3	1.8	3.4	5.6
-0.106/+0.075	9.3	1.8	2.7	6.1
-0.075/+0.053	10.4	2.1	3.3	6.9
-0.053/+0.045	3.5	4.1	4.5	12.1
-0.045/+0.020	13.2	4.3	9.4	14.6
-0.020/+0.010	11.8	7.3	13.0	27.9
-0.010/+0.005	9.4	13.5	22.0	54.7
-0.005/+0.002	5.3	21.1	16.9	98.0
-0.002	1.2	26.6	37.5	132.0

[1] Activity of whole soil is 5.4 pCi/g dry U-238, 9.5 pCi/g dry Ra-226, and 19.6 pCi/g dry Th-232.

radionuclide distribution are used to assess the potential effectiveness of volume reduction by size separation. Wet sieving is complemented by vertical-column hydroclassification. Hydroclassification is accomplished by allowing the soil to fall through a column of water flowing at a select velocity in the opposite direction. The process follows the principles of Stoke's Law in which particles settle and separate to a degree governed primarily by size and secondarily by density. Setting velocity depends on the second power of the particle size and the first power of the density and soil particle densities are usually similar throughout the size fractions. However, large density differences between clean and contaminated particles do provide a potential alternate separation process, especially if high density artifacts are present. Vertical-column hydroclassification is particularly attractive as a laboratory separation procedure in characterization since it takes much less time to perform than wet sieving and is a process that simulates many plant-scale operations. It has the additional attractive feature of being easier to scale up to plant level than many other plant technologies since the components are relatively simple when compared to other technologies and are readily available in the mining industry.

NEXT STEP

After determining the particle-size and radionuclide distribution of the soil samples, what is the next step in the treatability study? Basically, there are three alternatives: either the next tier of treatability studies could follow, bench-scale testing and/or process development could begin, or soil characterization and treatability studies could be terminated, primarily if funding is limited.

Should soil characterization reveal that volume reduction by particle-size separation might be applicable to site remediation, the next step in the study depends on the percent recovery of clean (remediated) material. If size separation produces a large recovery that would, alone, result in significant monetary savings when compared to alternate remediation proposals, the second tier of treatability studies, bench-scale testing and/or process development, would be indicated. Additional basic soil characterization would not likely be required. It is typically not essential, for example, to determine the actual substance(s) contributing to the activity in these cases; time and funding is usually better spent at this stage on the next tier. If separation produces a marginal recovery, as determined by comparison of remediation cost using particle size-separation to those employing alternate remediation methods, or if remediation by particle-size separation is not feasible, further soil characterization would be necessary. The additional effort is to identify other soil properties that could be adopted to supplement the marginal recovery or accomplish volume reduction where particle-size separation was unsuccessful. For example, in some soils the radionuclide activity might be primarily in the fines, but the larger soil particles might also contain sufficient activity such that they would not meet the clean-up criteria. Another soil may contain significant radionuclide activity distributed essentially throughout all soil particle sizes. In the former case, additional methods may be discovered that would move the activity to the smaller-sized fraction or remove it from the particles altogether. In the latter case, an alternate physical separation method based on a different physical property such as density or magnetic properties might isolate the activity. At this point in the study, it should be determined if proceeding with soil characterization and treatability would be economically feasible. With the exceptional savings that are often possible using soil treatment instead of disposal (6), further characterization will likely be worthwhile.

ADDITION CHARACTERISTIC STUDIES

If additional characteristic studies are indicated, selection of the next step in the process is based primarily on the source of contamination, knowledge of historical or available analytical information, and the potential of chemical extraction. Should the contamination be from fission or other man-made products, for example, and if there is sufficient information to indicate the presence of a likely contaminant with a unique physical property, a separation study of the material based on the unique property might well be indicated. The presence of weapons-related plutonium particles might be readily isolated by a density separation, for example, particularly if the particles do not have a uniform, unique size that would allow particle-size separation.

Ore-based contaminated soils, primarily those containing naturally occurring minerals with radionuclides from the uranium or thorium series, and anthropogenetically contaminated soils, other than those described in the preceding paragraph, should be examined by petrographic techniques that can detect the presence of radionuclide contaminants. Petrographic characterization, performed on soil fractions prepared during size characterization, might reveal the presence of an offending radiomineral with a unique property, such as high density, ferromagnetic or electromagnetic susceptibility, or specific-collector association for frothing, that might be used for separation.

At this time in the study, chemical extractability of contaminants from all or specific size fractions of the soil should be considered. Chemical extraction might be economically competitive with certain physical-separation techniques or with complete disposal of the whole soil, especially when extraction is coupled with physical separation. A soil, for example, might be fractionated by size-separation, producing a clean, gravel-to-sand size fraction that requires no further treatment, a sand-sized fraction that could be rendered clean by simple chemical extraction, and a silt-and clay-sized fraction with such high activity that further physical or chemical treatment would not be economically or technically feasible. Preliminary scout tests using dilute mineral acids typically employed for metal extraction, detergents, and hot water should be considered at this time. The results of petrographic studies might suggest a specific reagent for a radiomineral found in the soil. A sequential extraction protocol (3) that increasingly exposes the soil matrix to greater chemical dissolution power combined with concomitant examination of radionuclide released activity should also be considered. Both these chemical tests can be performed as part of a preliminary characterization study in a relative brief time period at a reasonable cost.

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