

DIFFERENCES IN RADIUM AND URANIUM DISTRIBUTIONS IN QUATERNARY DEPOSITS

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ABSTRACT

Radium distribution in soils is an important control on radon emanation. The aim of this study has been to study the differences and similarities of Ra and U distributions of Quaternary deposits with varying grain-size distributions. A discussion is made of the possible influence of varying sources of the sediments and the time available for soil formation. Clay, sand and till were investigated. Radium and uranium distributions were characterized by sequential extractions. Concentrations of all analyzed elements in surface-bound phases increased with decreasing grain-size, with higher concentration in oxide-bound than in exchangeable phase. Sand was sampled from the B-horizon, thus it should be more affected by soil-forming processes than the till, obtained in the C-horizon. Still results from till and sand samples were similar; showing that re-distribution of elements is also important at deeper levels in the soil. Clay generally had the highest concentrations of surface-bound elements. One conclusion is that surface area available for adsorption is the major control on Ra distribution.

INTRODUCTION

Radon in indoor air is a recognized health-risk, which has led to extensive research in the area of radon emanation. The actual mechanisms that control radon emanation are not totally understood. The distribution of radium in the soil is one important parameter governing radon emanation. Several workers have presented theoretical calculations, showing that the radium distribution is of great importance for radon emanation, e.g. Morawska and Phillips (1993), however few have studied this in practice.

The physical characteristics of a soil, such as grain-size distribution and permeability, are governed by many factors of which some include parent rock material, mode of formation of the minerals, the means and distance of transport, and the depositional environment. These characteristics together with the uranium and radium content of the parent rock, and recent physical and chemical events (i.e. chemical leaching, transport, precipitation/adsorption) can affect the final distribution of radium in the soil (Kemski et al., 1992). Weathering results in the mobilization of U and Ra out from the bedrock. They are subsequently redeposited, primarily through adsorption to ferric oxyhydroxides, organic material and secondary minerals. U and Ra may also be adsorbed or precipitated on the surface of grains or pore space surfaces (Wanty et al., 1991; Ek and Ek, 1996). Radium is less efficiently sorbed onto iron oxides (Ames et al., 1983a) and more efficiently sorbed onto secondary minerals with high cation exchange capacity, than uranium is (Ames et al., 1983b). Since U and Ra do not behave in exactly the same way, a radioactive disequilibrium is common in soils.

The aim of the study has been to study the differences and/or similarities of the Ra and U distribution of different types of Quaternary deposits with respect to their grain-size fractions. Radium and uranium distributions have been characterized by selective sequential extraction. The method has been used for characterization of uranium and radium distributions, e.g. on uranium mine tailings (Landa, 1984) and on natural soils (Greeman and Rose, 1996; Greeman et al., 1999). To be able to make comparisons between the different Quaternary deposits we chose to obtain the samples from glacial or late glacial unconsolidated deposits in a restricted area, ensuring similar age, similar climatological influence after deposition. In this paper, variations and reasons for the variations in Ra distribution in different materials is discussed. The method has previously been described in Edsfeldt (2000).

GEOLOGY

Bedrock and Quaternary deposits

In accordance with the aim, the samples were taken from different Quaternary deposits located in the same drainage basin to insure similarity in the post-depositional environment. The source material for the sediment is assumed to be primarily the local bedrock material as well as bedrock in the up-ice direction. The bedrock (Fig. 1) is Precambrian in age and consists of different types of gneiss with aplite and pegmatite dikes. Red granites, gneiss-granites and sedimentary gneisses predominate the adjacent up-ice area (N-NW of the sampling area) (Stålhös, 1969). The red granite is enriched in uranium, with a U content of 10 to 30 ppm eU. The uranium content of the other rocks is low. Further to the N-NW there are some plagioclase-porphyrines of andesitic-basaltic composition. Darker bands of gneisses occur both in the sampling area and in the up-ice direction.

The bedrock is overlain by Quaternary sediments (Fig. 1) deposited during or after the Late Weichselian Glaciation. The ice retreated from this area about 9000 BP. At that time the sea level was at about 150 m higher than today (Möller & Stålhös, 1964). The shoreline displacement curves for this region (Ambrosiani, 1984; Brunnberg et al., 1984; Risberg et al., 1991) show the time of emergence for different altitudes. During emergence wave washing and reworking of surface sediments occurred.

The most predominant Quaternary sediments in the area are till, glaciofluvial material, glacial clay, postglacial clay, and wave-washed sand. The till has been deposited directly from the glacial ice. It consists of a large variety of grain sizes from clay to large boulders. The relatively high boulder content, loose packing, and location of the till suggests that it is a lee-side till. The larger grains are expected to be predominately from local bedrock sources. In contrast the fine grains reflect a higher degree of mechanical breakdown and thus are expected to contain a greater percent of grains that originate from more distal sources (Sugden and John, 1976). The glaciofluvial esker sand and gravel have been deposited within cavities or crevasses in the glacier by meltwater streams. The source of the sediment is expected to be similar to that of the till. Large clasts are commonly from local sources. Fine grains are often from more distal sources. Clay size particles are often lacking in esker deposits, these are often washed out of the glacier and deposited in proglacial lakes. The glacial clay has been deposited in the Baltic Basin during the deglaciation. The clay is transported by meltwater streams, out from the glacier into the basin where it is laid down in thin layers over the basin floor. Thus the clay does not represent one source but a mixture of

several often very distal sources. The post-glacial clay differs from glacial clay with respect to source area. It originates from fluvial erosion of the surrounding landmass. Thus the source is restricted to the bedrock and Quaternary deposits in the drainage basin and the post-glacial clay reflects a more local source than does glacial clay. The post-glacial wave-reworked sand was formed as the land emerged from the sea. The source of this sediment is primarily the pre-existing glacial deposits, the till and glaciofluvial material.

SAMPLES

Samples were collected in the beginning of November 1996 from the drainage area of Edsviken, an inlet of the Baltic Sea, close to Stockholm. In this area the Stockholm esker trends S/N, parallel to and on the W side of the Edsviken Inlet. Till samples were collected from the eastern shore of Edsviken Inlet (in Danderyd). The sand and clay were sampled from the western shore area of Edsviken (in Ulriksdal). Sampling locations can be seen in Figure 1. High concentrations of radon in soil air have been measured in the Danderyd area; 70000 Bq/m³ in till, and 150000 Bq/m³. Gamma spectrometry measurements at the same location gave a soil uranium content of 5-6 ppm, and a thorium content of 24 ppm. In the Ulriksdal area the soil radon concentrations clay-silt material were somewhat lower, 35000-39000 Bq/m³. At the sampling sites the mean annual temperature is approx. 5.5° C (coldest month February, mean January temperature -4.5° C, warmest month July, mean temperature 16.5°C). Mean annual precipitation is 600 mm.

Two till samples (Till 1 and Till 2) were collected from different depths at the same site (1.5 m apart), on a slope at about 20 m above MSL (mean sea level). Till 1 was sampled from 0.40-1 m depth. It had a sandy matrix and was rich in cobbles and boulders. Till 2 was sampled from 1.40-1.80 m depth, it had a silty-sandy matrix, and contained somewhat fewer cobbles and boulders than Till 1. Judging from the sampling depth, the till samples would belong to a C-horizon, however there were no evident soil horizons at the site. The sand was sampled from 0.5 m depth in a gravel pit in the Stockholm Esker at a about 15 m above MSL. It was medium to coarse-grained and had a reddish color. The sample was obtained at a depth shallower than would normally correspond to the C-horizon, and the reddish color suggests that the sand sample might belong to a B-horizon. Postglacial clay was sampled adjacent to the esker, about 3 m above MSL, from a depth of about 1.5 m. The postglacial clay was overlain by a superficial, anthropogenically influenced layer 0-0.5 m, and underlain by glacial clay. Small nodules of iron oxide were dispersed in the clay. The depth and the impermeable material suggest that the sample should be little affected by soil-formation processes.

Shorelevel displacement and age of samples

Soil formation first begins as the deposits are lifted up out of the sea into a land environment. The length of time available for this process to act on the different samples studied differs. Based upon shore displacement curves from the Södertörn area, located about 60 km to the south of the study area, land at 20 m above sea level would have emerged about 3400 to 2800 years BP and land at 15 meters above sea level would have emerged at about 2900 to 2500 years BP (Tab. 1). The dates would be roughly applicable to the study area since little variation in emergence is expected over such a short distance (Risberg, pers. comm., 2000). The dates are based on ¹⁴C dates; Risberg et al. (1991) dates are of lake sediments, while Brunberg et al. (1984) are of peat overlying the lake sediments. The latter will give numbers

a bit too young, since roots and other recent material penetrates down into the dated levels. The lake sediment dating might be too old, due to reworking of the sediments. However, they are in agreement with archaeological evidence, and are thus assumed to be the more exact of the two.

METHOD

Samples of till and sand were dried at room temperature and dry-sieved to obtain the fine fraction, consisting of silt and clay particles (<0.063 mm). The coarser fractions of the samples were washed to remove remaining clay and silt particles, then dried at room temperature and sieved. The clay was not sieved. Sedimentation analysis of clay and fine material (silt and clay) of till and sand was performed.

The procedure for sequential extraction and chemical analyses has been developed for this project based on other existing research methods. A brief description of these follows. A more elaborate description of both the sequential extraction methods and analyses methods is presented in Edsfeldt (2000).

The usual sequential extraction scheme includes extraction agents for exchangeable cations, carbonates, organic material, oxides (of Fe, Mn and Al), and residual (Förstner, 1993). Since the source material for the sampled soils was mainly granitic and the content of organic material was low, separate extractants for carbonates and organic matter were excluded. Dissolved elements were extracted with deionised water from the un-washed fine-fractions (<0.063 mm). Exchangeable elements were extracted with 1M NH_4Cl (ammonium chloride), and oxide bound elements were extracted with 1 M HCl (hydrochloric acid). The residual soil was mixed with lithium metaborate (LiBO_3), and fused by heating at 1000 °C for 45 minutes; resulting in a "glass" bead that was dissolved in 5 % HNO_3 during shaking.

Radium in extractant solutions and digested samples was analyzed with α -spectrometry by means of liquid scintillation counting (LSC) (Suomela, 1993). The concentrations of Ra in the residual were under the detection limit, thus the exact Ra content is not known. The residual Ra was estimated from the total concentration of Ra subtracted by the total concentration of Ra in surface-bound fractions. Uranium was analyzed by mass spectrometry (ICP-MS). Iron, manganese, calcium and barium were analyzed with atomic emission spectrometry (ICP-AES). Total radium content was determined by gamma spectrometry (van der Graaf et al., 1998a), and radon exhalation measurements were performed at the KVI, Netherlands, according to van der Graaf et al. (1998b).

RESULTS AND DISCUSSION

The compositions of the samples are shown in Table 2. The grain-size distributions of the samples can be found in Figure 2. Element distributions are shown in Figure 3A for the exchangeable + dissolved phase, Figure 3B for the oxide bound phase, and Figure 3C, residual. Generally, concentrations of Fe, Mn, Ca, Ba, U and Ra in all surface-bound soil phases increased with decreasing grain-size. The maximum concentration was most often attained in the clay sample, which also had the largest surface area (largest portion of clay-sized grains). With very few exceptions, elemental distributions in the two till samples and

the sand sample were similar. The sand generally had somewhat lower element concentrations in the grain-size fractions $>0,125$ mm and >1 mm, and higher concentrations in the silt-clay fraction, compared to the till samples. The silt-clay fraction of sand sometimes had higher element concentrations in soil phases than did the clay, e.g. it had higher total Ra concentration. However, the fine sand had lower Rn exhalation than the other fines. In general, Ra correlated better with Ca than did U; and U correlated better with Fe than did Ra.

Soil phase

The main portion of all elements, except Fe, was found in the residual. For Fe the residual and the oxide-bound fraction were of equal importance. The main part of surface-bound elements was also found in oxide-bound fraction, where the good correlation between Fe suggests that these other elements were associated with Fe oxides in these soils. Ca was the predominating element extracted in the exchangeable fraction, where the correlation between Ca, Ba and Ra was good. Since their geochemistries are similar, this correlation could be expected. In Figure 2, the dissolved elements have been added to the exchangeable elements (for grain-sizes <0.063 mm). The dissolved concentrations were negligible, with the exception of clay, in which dissolved Fe and U were important. Noticeable is that Fe and U had negligible concentrations in the exchangeable phase.

Soil age and climate

Soil formation processes that are active are dependent on climate. The temperature at the time of the Weichselian termination was approximately similar to today's. It rose to a maximum, about two degrees C higher than present, at about 5000-6000 years BP, then slowly decreasing again until that of today. Deviating from this main trend, there have been oscillations (Eronen, et al., 1999). At the time of emergence of the sampled soils from the postglacial sea the mean annual temperature might have been about 1° C higher than today. This difference is probably not enough to dramatically change the active soil formation processes from those of today.

Protz et al. (1984) have references on rate of podzolization. The time for a mature podzol profile to form vary from place to place due to the physical environment; just over 1000 years (N Norway) and about 5000 (carbonate rich soils in N Ontario). These sites both have a mean annual temperature of less than 0°C (32°F). The mean annual temperature of the sites investigated in the present study is somewhat higher (5.5°C). It is expected that this difference would result in a faster decomposition of organic material, and hence, less relocation of elements by complexation with organic acids (Schaetzl and Isard, 1996). Soils in this area are generally brown earths, characterized by having no pale eluvial E-horizon (due to reworking of organisms). There might be an illuvial B-horizon, or a red-brown illuvial containing organic material and weathering products such as Fe and Al oxides. The relocation of elements between different depths in the soil horizon is generally rather small. Relocation of elements weathered from the crystal lattice to grain surfaces will still occur in the upper soil layers, as well as transport of weathered salts down the soil profile.

The till samples, although they show no visual signs of soil formation, are clearly affected by soil formation processes. This is shown by the results of the sequential extractions. Not only extractable elements are found here, but also Fe oxide bound elements. The till samples, obtained at different depths, do have very similar element distributions. The shallower till does have somewhat higher extractable concentrations of the analyzed elements, which could be expected since it should be slightly more weathered than the deeper till sample.

Soil type and grain size

The aim of the study has been to study the differences and/or similarities of the Ra and U composition of different types of Quaternary deposits with respect to their grain-size fractions. Where the coarse material in the till would be very local material, the finer fraction would be less predominated by local material. The postglacial clay would reflect the entire drainage basin area at the time of its formation. The sand would reflect the glacial drainage basin. Thus – it could be expected that there are differences in element distributions between the soil types. However, the mineral composition in the different grain-size fractions of sand and till does not differ much (Tab. 2). The rock fragments (1-2 mm) in the sand were slightly platy, indicating a portion originating from gneissic material, whereas the same till fraction consisted mainly of granitic material. The fine fraction of the sand (<0.063 mm) was more coarse-grained than the fine-fraction of the till samples.

Element concentrations in the coarse grain-size fractions of the sand sample are generally slightly smaller than corresponding till concentrations. This applies to both extractable elements and residual. The lower residual concentrations are due to the well-sorted character of this sediment, predominated by quartz and potassium feldspar. The silt-clay fraction of the sand differs by having much higher concentrations of Fe, Mn and U in both exchangeable, oxide bound and residual phases. However, Ra concentrations are almost the same as for the silt-clay fraction of the till. It thus seems that the silt-clay fraction of the sand contained Fe-Mn oxides, to which U was associated, but not Ra. This concurs with the results of Ames et al. (1983a) that U is more efficiently adsorbed to Fe oxides than is Ra. The reason for this deviating behavior of the fine sand compared to the till and the coarser sand can be that the fine material was washed down from more superficial soil layers.

The element distributions in the different grain-size fraction of sand and till are similar, in spite of their dissimilar genetic origin and position in the soil horizon. Both till and sand are inferred to be derived from a more local source than would the clay. Still, for these samples, the primary control on element distribution seems to be grain size / surface area, which governs the extent of adsorption and co-precipitation onto the grains.

Although the clay had a very short exposure to a weathering environment, less than 700 years, it had the largest portion of extractable elements. Especially the concentrations of Ca, Ba and Ra in the exchangeable fraction were very high compared to the silt-clay fractions of till and sand. This extensive deviation from the other soil types could be expected due to the many special qualities of clay. Not only does clay have a superior specific surface area, but also a negative charge on particles that promotes adsorption of ions. Also, clay usually contains minerals that are altered and weathered, which often increases the cation exchange capacity. A first assumption, based on the large sampling depth, would be that the sampled clay material would consist mainly of crushed rock, and not secondary clay minerals formed by soil formation processes. However, the XRD analysis of the clay sample showed that 47 % of the material was clay minerals (including kaolinite and micas, Tab. 2).

VandenBygaart et al. (1999) notes that for soils containing clay material, the clay content seems to be more important in determining background radioactivity than soil mineralogy. They found that for many radionuclides, radioactivity was linearly correlated with clay content. This study again points out that not only clay content, but grain-size in general, is a very important control on radioactivity of soils.

CONCLUSIONS

In general uranium is more closely associated with iron and manganese, probably adsorbed to or co-precipitated with oxides of Fe, Mn and Al. Radium on the other is more related to Ca and Ba in its behavior. These conclusions are in accordance with the results of Ames et al. (1983a and 1983b), U is more efficiently adsorbed to Fe oxides than is Ra, and Ra is more efficiently sorbed onto secondary minerals with high cation exchange capacity, than U is. This is one of the processes responsible for the radioactive disequilibrium between Ra and U in soils.

The effects of soil age on soil formation is not evident from these samples. The general method to quantify such effects is to study a soil chronosequence (Protz et al., 1984). However, the investigated till, sampled at a depth of approx. 1 m, with no visible signs of soil formation, is clearly affected by soil formation processes. Not only extractable elements from the downward transport of weathered salts are found here, but also Fe oxide bound elements occur. The shallower till sample does have somewhat higher extractable concentrations of the analyzed elements, which could be expected since it should be slightly more weathered than the deeper till sample.

In spite of their dissimilar genetic origin and position in the soil horizon, the Ra and U distributions in sand and till are very similar. For the till and sand samples, the primary control on element distribution appears to be grain size / surface area, which governs the extent of adsorption and co-precipitation onto the grains.

The very young clay sample (700 years) had very high exchangeable concentrations of Ra, Ca, and Ba, compared to the silt-clay fractions of till and sand. The suggestion by VandenBygaert et al. (1999) that for soils containing clay material, the clay content is more important in determining background radioactivity than soil mineralogy, is not contested by the results of this study.

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Table 1. Time of emergence of soil samples from the post-glacial sea.

Sample	height above M.S.L. (m)	Time of emergence (years B. P)		
		acc. to Risberg et al. (1991)	acc. to Brunberg et al. (1984)	acc. to Ambrosiani (1981)
Till 1	20	3400	2800	
Till 2	20	3400	2800	
Sand	15	2900	2500	
Clay	3		750	700

Table 2. Major mineral and rock types in samples. Minerals in till and sand determined by ocular inspection. Constituents of clay determined by XRD analysis.

	GRAIN-SIZE [mm]	MAIN MINERALS/ROCK TYPES
Till 1	1-2	Mostly rock fragments: 2/3 with granitic material, 1/3 greenstones ¹⁾ . Very few quartz particles.
	0.25-0.5	Equal portions of quartz, potassium feldspar and greenstone ¹⁾ grains.
	0.125-0.5	Potassium feldspar and amphibole in equal portions, somewhat more of quartz. Some muscovite and biotite.
	<0.063	Very fine-grained. Quartz and some muscovite grains recognizable.
Till 2	1-2	Corresponds to Till 1 (1-2 mm)
	0.25-0.5	Somewhat less greenstones ¹⁾ , than in Till 1 (0,25-0,5 mm). Some muscovite.
	0.125-0.25	Corresponds to Till 1 (0,125-0,25 mm)
	<0.063	Very fine-grained. Quartz grains recognizable.
Sand	1-2	Mostly rock fragments, particles are somewhat more platy than in the till materials. Granitic material, greenstone ¹⁾ material and ~10% mica schist. Quartz and feldspar grains.
	0.125-0.25	Quartz grains predominate. Potassium feldspar and amphiboles. Some biotite and muscovite.
	<0.063	Less fine-grained than Till 1 and Till 2 (<0,063 mm). Quartz, muscovite, biotite and amphibole grains.
Clay	<0.063	66 % clay-sized material and negligible amounts of grains >0.06 mm. Quartz, 23 %, cristobalite, 8 %, potassium feldspar, 6 %, plagioclase, 12 %, "mixed- calcite", 2 %, pyrite, 2 %, clay minerals, 47 %. In "clay minerals" micas and kaolinite, as well as smectites etc, is included. The amount of smectites is believed to be rather small.

1) "Greenstones" refers to aggregates of dark minerals, consisting mainly of amphiboles.

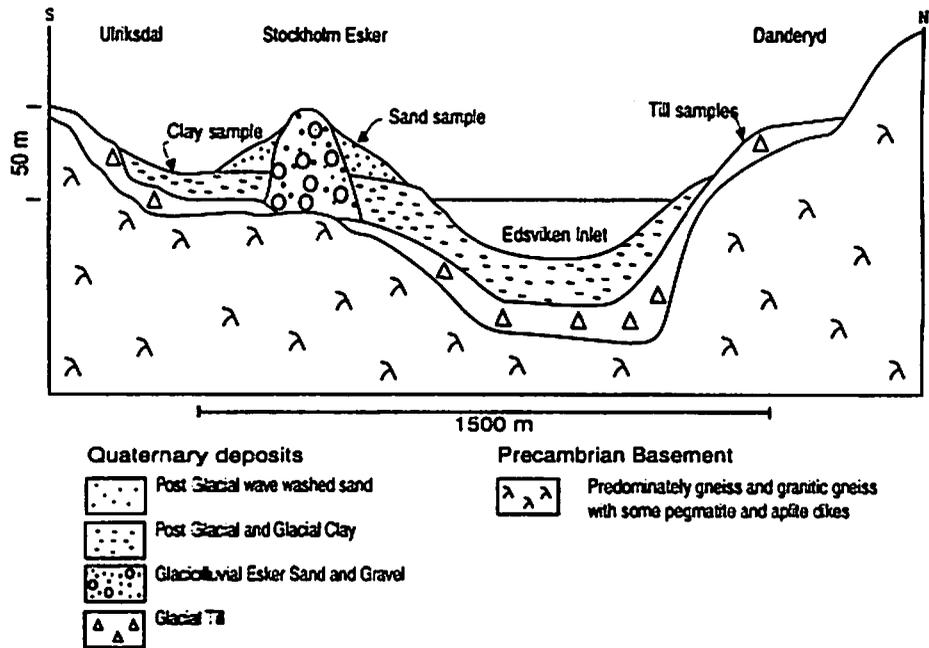


Figure 1. Geology of the sampling area.

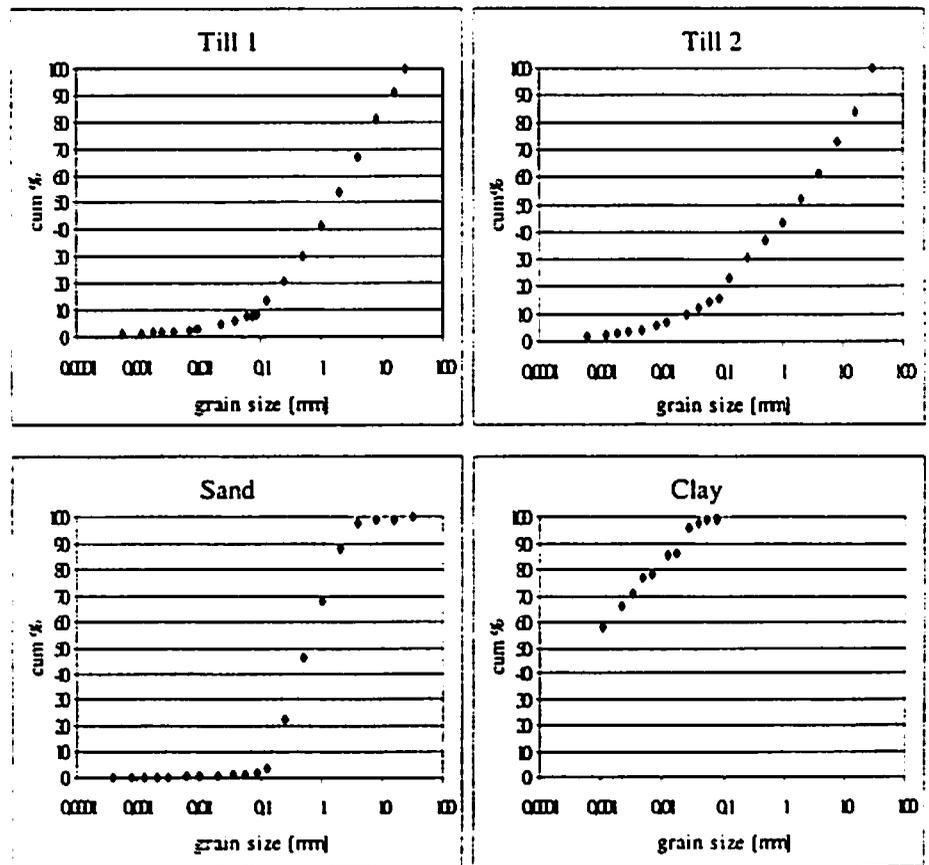


Figure 2. Grain-size distributions of the samples.

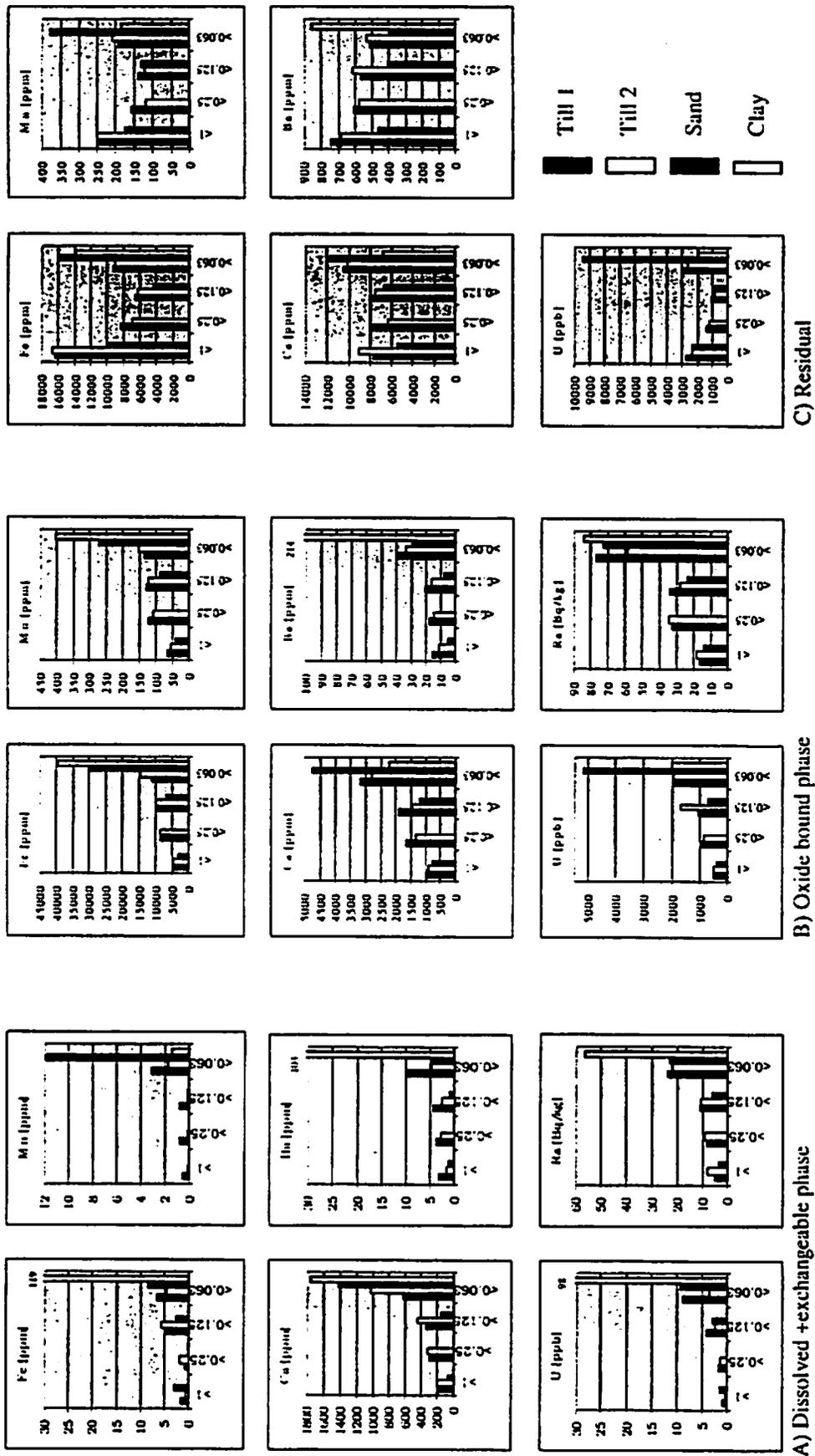


Figure 3. Element distributions in the A) dissolved + exchangeable phase; B) the oxide bound phase; and C) the residual. Legend for all phases is found in part C.