DIFFERENTIATION OF RADIOACTIVE GROUND CONTAMINATION WITH TENORM

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1 ABSTRACT

Sites of previous production and/or utilization of radioactive elements often show ground contamination with natural radionuclides. The kind and amount of contaminated soils, sediments and other materials depends on the applied technical procedures applied and on the previous handling of raw materials, products, by-products and wastes.

Hazard assessment and remediation planning have to be based on the knowledge of both inventories and the actual nuclide composition of the contaminated soil. Since, normally, several decades will have elapsed between the development of contamination and the present site investigation the long-lived isotopes are characteristic indicators of the nuclide composition measured today. Among these isotopes, the nuclides Th-230, Pa-231 and Ac-227 are often neglected. However, they may retain valuable information on the processes that proceeded in the past.

2 INTRODUCTION

There are several sites in Germany were where substantial amounts of material enriched with uranium and/or thorium (TENORM) had been used or processed in former times. Besides the uranium processing plants in Sachsen (Saxony) and Thüringen (Thuringia) (Wismut plants of Crossen and Seelingstädt) and Ellweiler in Rheinland-Pfalz (Rhineland Palatinate) there are certain locations where thorium was produced for gas mantle production, radioluminous paints were used for clock production, or rare earth elements were produced; some of these are given in Table 1.
Therefore, soils or old waste dumps at such sites are often contaminated with natural radionuclides. Usually, the radioactive contamination is accompanied by other human- or ecotoxic substances, resulting in a mixed ground contamination.
So far such cases of mixed contamination have not been considered in German radiation protection or in the regularities for soil protection and it is still missing in in the new Radiation Protection Ordinance of July 20th 2001 (StrlSchV 2001).
However, at a sufficient level of contamination the radioactive component has to be taken into account when it becomes to practical site investigations, remediation measures or the disposal of excavated ground. In such cases reasonable and practicable solutions may be required from the involved engineers and regulatory authorities.

Table 1: Locations in Germany where radioactive material had been produced or utilised (examples)

Location (Land)	Material used or produced	Ref.
Ellweiler (Rheinland-Pfalz)	uranium processing plant	(1)
Crossen (Sachsen)	uranium processing plant	(2)
Seelingstädt (Thüringen)	uranium processing plant	(2)
Oranienburg (Brandenburg)	thorium, rare earth elements	(3), (4)
Braunschweig (Niedersachsen)	radium	(8)
Villingen-Schwenningen (Baden-Württemberg)	radium-paint	(5)
Schwarzheide (Brandenburg)	thorium-catalyst	(6)
Schweina (Thüringen)	thorium; gas mantle production	(7)
Gottow (Brandenburg)	uranium	(3), (4)

According to our experience gained in several studies, the complete nuclide composition plays a decisive role when assessing contaminated materials. This aspect will be discussed below.

3 INVESTIGATION OF SITES WITH RADIOACTIVE CONTAMINATION

As a rule, sites suspected of being contaminated are examined for the purpose of preventing or determining a hazard. This requires first of all to ascertain the distribution of harmful substances in soil and groundwater. Usually such site investigation is carried out step by step beginning with a historical investigation, followed by exploratory investigation and finally the detailed investigation. Concerning the radioactive components, certain pecularities have to be observed when planning and implementing these investigations.

- Natural radionuclides form decay series. As a result, there ever occur mixtures of different radionuclides, sometimes in a significant disequilibrium. In many cases the series of both uranium and thorium are important for characterising the TENORM.
- Historical investigation is an essential step for getting information about the materials formerly used. Apart from the materials, the processes involved are very important too. Especially any processes that involved dissolution/ precipitation could result in strong fractionation of radioelements and also in uncontrolled losses leading to ground contamination with mobile

radionuclides. On the other hand the utilisation of radioactive products, e.g. catalysts, usually resulted in a rather uniform composition of the residues.

- Unlike with other trace elements, the existence of radionuclides (except isolated uranium, Pb-210, Th-230) can easily be checked by field measurements, this enabling the mapping contamination close to the surface.
- Measurements of gamma logs in boreholes or simple radiometric counting of the samples in the field often provides qualitative information about radioactive materials in the ground. This information can then be used to select samples with different levels of specific activity.

4 LABORATORY MEASUREMENTS

To characterise the nuclide composition (nuclide vector) of a certain material it is necessary to measure the long-lived radionuclides. Gamma spectrometry as the most common technique is most suitable for measuring a significant part of the long-lived nuclides either directly (by their own gamma lines) or via their short-lived daughters (see Table 2).

Table 2: Measurability of long-lived decay series nuclides by gamma spectrometry

Direct measurability (sufficient sensitivity)	**Measurement via short-lived daughter**	**Direct measurablility (low sensitivity)**	**Not detectable**
Ra-226	U-238 (Th-234)	U-234	
Pb-210 (*)		Th-230	Po-210
Th-228	Ra-228 (Ac-228)	Th-232	
U-235 (**)	Ac-227 (Th-227)		
Pa-231 (**)			

(*) Limitations due to self-absorption; (**) Limitations due to background effects

However, it is difficult to determine the nuclides U-234, Th-230 and Th-232 by gamma spectrometry. Po-210 emits no gamma rays and therefore it cannot be detected by gamma spectrometry. As the contaminations considered here were generated many years ago, Po-210 is in equilibrium with Pb-210.
From the three nuclides measurable only with low sensitivity U-234 in natural materials is almost in equilibrium with U-238, and the activity ratio U-234/U-238 is not influenced by any chemical processing. Therefore, the activity ratio U-234/U-238 yields only little information about the materials used before and about the chemical processes applied.
The activity ratio Th-230/Th-232 is not influenced by chemical processes either. However, since Th-232 and Th-230 belong to different decay series, the activity

ratio is determined by the Th-230/Th-232 ratios of the raw materials. Therefore, the activity ratio Th-230/Th-232 is a valuable indicator to disentangle materials of different origin. Often this ratio needs to be determined with high accuracy and requires alpha-spectrometric analysis.

5 DIFFERENTIATION OF MATERIALS

Measuring results describe the contaminated ground by the specific activity of the nuclide vector and its composition. In the following we will discuss the information obtained especially from the nuclide vector.
Despite different specific activities, the nuclide vectors of miscellaneous materials can be compared by using the scaling transformation

$$a_{iN} = a_i / (a_{U\text{-}238}+a_{U\text{-}234}+a_{Th\text{-}230}+a_{Ra\text{-}226}+a_{Pb\text{-}210}+a_{Th\text{-}232}+a_{Ra\text{-}228}+a_{Th\text{-}228}+ 21.7*(a_{U\text{-}235}+a_{Pa\text{-}231}+a_{Ac\text{-}227}))$$

where a_{iN} denotes the scaled activity value of the nuclide “i”. This gives a clue to check the uniformity of different measuring results and to characterise miscellaneous materials.

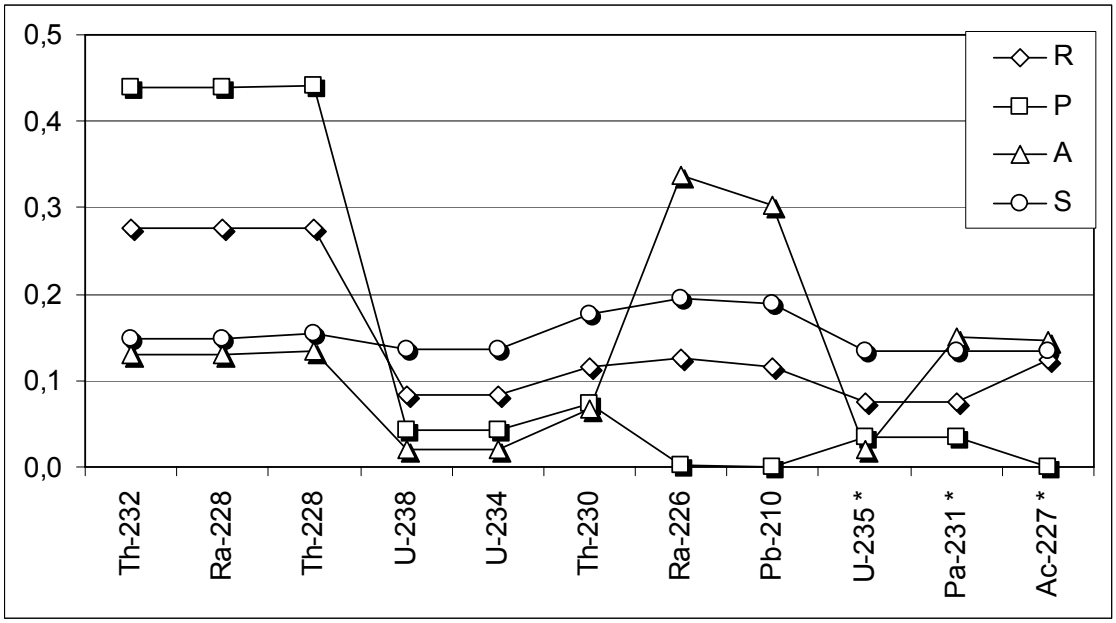

Figure 1 Nuclide vectors of residues at thorium-contaminated sites

Nuclide vectors inferred from measuring results of sites with thorium contamination are shown in Fig. 1. The patterns represent typical groups of materials:

- Residues of the raw material (identifier “R”) show high Th activities and low U activities. Both decay series are roughly in radioactive equilibrium.
- Residues of chemical processing (identifier “A”) show highly depleted Th activities as compared with Ra-226. The example represents an acid extraction in which uranium and also thorium were removed by leaching. Therefore, the residues contain mainly Ra-226 and daughters including Pb-

210 and Po-210 (not shown in the diagram). Compared with U-235, relative enrichments of Ac-227 (Pa-231) were found. This means that Pa-231 and Ac-227 remained together with Ra-226 in the solid material.

- Product residues (identifier “P”) also show high Th activities and low U activities. Compared with the raw material however, the U-238 series is depleted, whereas Th-230 remained.
- Uncontaminated soils (identifier “S”) show roughly comparable U and Th activities. Both decay series are practically in an activity equilibrium.

Measuring results with different nuclide vectors can usually be interpreted as a mixture of these four types of material.

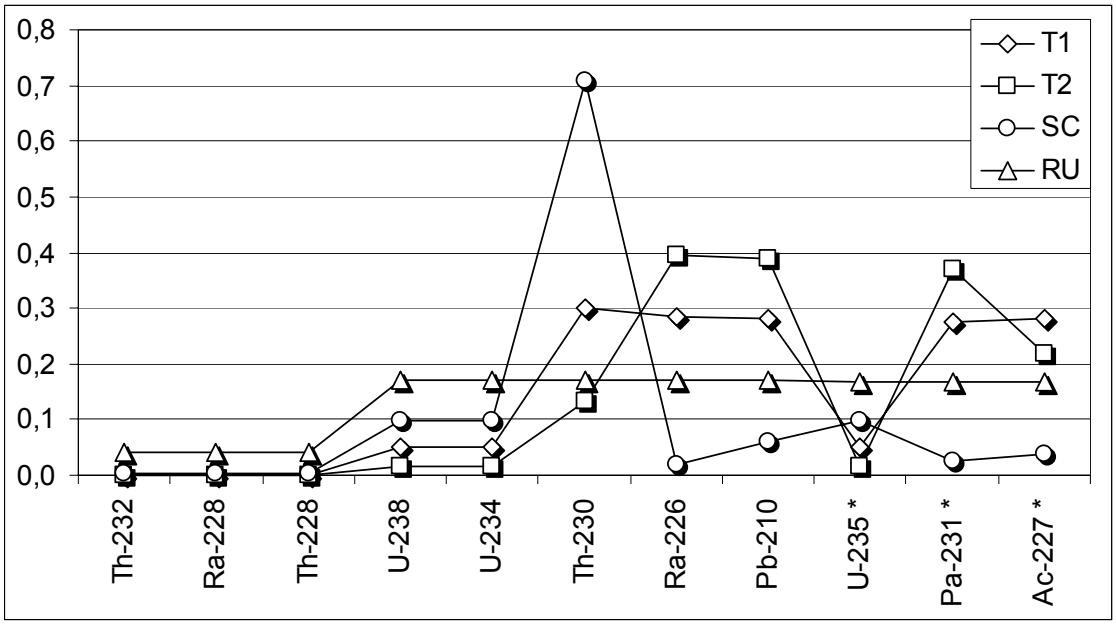

Figure 2: Nuclide vectors of residues found at sites with uranium ore processing

Similar plots of residues from uranium processing are shown in Fig. 2.

- Residues of uranium ore (identifier “RU”) have high U activities and low Th activities. Again both decay series are roughly in activity equilibrium.
- Residues of chemical processing are deposited in tailings ponds. Tailings 1 (identifier “T1”) originate from alcaline leaching, tailings 2 (T2) from acid leaching. As Th is extracted in an acid leaching process, the scaled nuclide vector of T1 differs from T2 especially with regard to Th-230.
- An uncommon type of contamination is plotted as “SC”. Here, the nuclide vector is heavily dominated by Th-230. The source of this contamination could be traced back to a loss of process liquids with high concentrations of uranium and Th-230. Due to the higher mobility of U however, this component was washed out and only Th-230 remained.

6 CONCLUSIONS

Typical groups of materials expected at sites of former production or utilisation of natural radioactive elements include residues of raw material (ores), residues of chemical treatment, product residues (U, Th, Ra concentrates), residues of secondary processes (e.g. water purification) or contaminations from uncontrolled process losses.
Identification of different residues is an important step towards the investigation of contaminated sites. It helps to reconstruct the site history and forms a basis for an individual exposure path analysis.
A scaling method has been proposed for comparing the nuclide vectors of miscellaneous materials irrespective of their specific activities.
Besides the possibility of classifying the contaminated materials, such scaling allows to check the quality of different measuring results on the same footing.
Complete recording and consideration of all long-lived nuclides is very important for characterising contaminations. Especially Th-230 can hold plenty of information about the processes in the past.
Additionally, in some processes this nuclide is enriched to such an extend that an additional separate treatment is required for hazard assessment.
Alpha-spectrometric measurements of thorium should be carried out supplementary to gamma spectrometry in order to get sufficiently accurate values of the Th-230 activity as well as of the Th-232/Th-230 and Th-228/Th-232 activity ratios.
Since results of radiometric measurements, such as dose rates or detector counting rates, strongly depend on the specific nuclide vectors involved, knowledge of the latter facilitates the interpretation of the results of such cheap field measurements on a well-founded basis.

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