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TREATMENT AND DISPOSAL OF *NORM* CONTAMINATED WASTES

Rainer Gellermann; Jürgen Gerler
HGN Hydrogeologie GmbH; Holzwiese 6; D-38179 Schwülper

Introduction
In the past, in several industrial processes substances with enhanced contents of natural radionuclides have been used or generated, sometimes without sophisticated control of the radiological aspects. Consequently, there remained wastes with significant activities of radium, thorium, uranium or lead-210. In many cases the materials are also contaminated by non-radioactive toxic components.

Once such materials have been ascertained, the further handling requires:
- assessment of radiological exposures to analyse the real hazards to workers as well as to the public,
- concepts for site remediation including suitable treatment methods,
- suitable ways for safe disposal and/or recycling.

In the following some aspects of these problems will be discussed on the basis of experience gained from different projects.

Assessment and classification of materials
All materials contaminated by decay series radionuclides contain a mixture of different radionuclides. In Table 1 four examples of nuclide vectors are given. The activities are referred to the first member of the most important decay chain.

The materials selected represent mine dumps of uranium mining, radium scale of oil-gas production and soils contaminated by former chemical treatment of thorium compounds.

An important aspect of the formal classification concerns the origin of material from a practise under the regulations of the Radiation Protection Act or other kinds of practise (for instance mining). In the first case wastes have to classified as radioactive down to very low exemption levels (see Table 1). In the latter case the same material can be handled without licensing or registration up to total activities of 500 Bq/g. However, the uncertainties of the Rad. Prot. Act causes some differences between different federal authorities concerning the number of radionuclides to be involved in calculation of total activities. In some cases all physically existing radionuclides are taken into account, in other cases only the long-lived isotopes (half-life > 100 days) are included.

Although materials from outside the radiation protection law with total specific activities below 500 Bq/g should be considered as common wastes their disposal on waste deposits frequently fails due to the very restrictive acceptance of radioactivty. This practise is supported by recommendations of the German Commission on Radiological Protection (SSK) which contain very low levels for restricted as well as unrestricted exemption (see Table 2). These recently published levels fall below those of the Radiation Protection Act if the mixture of nuclides is taken into account. The unrestricted exemption level is significantly lower than the average natural concentration of the radioelements considered!

A further problem is the classification of materials contaminated before the establishment of any radiation protection. Such cases exist by different industrial applications of radioelements since the second half of the 19th century.

Besides the formal classification the assessment of radiological risks is required for derivation and justification of measures as well as limitation of exposure situations. Especially in the case of contaminated soils with partly unknown history the exact determination of the nuclide vector is highly important. The Th-Ra material characterised in the last column of Table 1 was found in some thorium contaminated soils. It is characterised by an uncommon thorium isotopes signature with ^{227}Th/^{230}Th up to above 1 (usually this ratio is 0,046). Obviously it is a residue of an uranium ore leached with sulfuric acid and mixed with a thorium contaminated soil. The isotopes of the ^{235}U-series play an important role in the dose calculations for this material and cannot neglected (see Table 3).

Table 1: **Radionuclide composition of different materials. L - long-lived isotopes according German Radiation Protection Act (half-life > 100 days); LT- additional long-lived isotopes according transport regulations (GGVS, ADR), half-life > 10 days**

		U dump material	Ra scale	soil, Th contaminated	soil, Th-Ra contaminated
^{238}U	L	1,0			
^{234}Th	LT	1,0			
^{234}Pa		1,0			
^{234}U	L	1,0			
^{230}Th	L	1,0		0,15	0,25
^{226}Ra	L	1,0	1,0		7,0
^{222}Rn		1,0	1,0		7,0
^{218}Po - ^{214}Po (4)*)		1,0	1,0		7,0
^{210}Pb	L	1,0	1,0		7,0
^{210}Bi		1,0	1,0		7,0
^{210}Po	L	1,0	1,0		7,0
^{235}U	L	0,046			
^{231}Th		0,046			
^{231}Pa	L	0,046			0,22
^{227}Ac	L	0,046			0,22
^{227}Th	LT	0,046			0,22
^{223}Ra	LT	0,046			0,22
^{219}Rn - ^{207}Tl (5)		0,046			0,22
^{232}Th	L	0,10		1,0	1,0
^{228}Ra	L	0,10	0,6	1,0	1,0
^{228}Ac		0,10	0,6	1,0	1,0
^{228}Th	L	0,10	0,6	1,0	1,0
^{224}Ra		0,10	0,6	1,0	1,0
^{220}Rn - ^{208}Tl (5)		0,10	0,6	1,0	1,0

**) number of short lived nuclides, which have to considered in calculation of total activities*

Table 2: **Derived exemption levels for materials characterised in Table 1. The activities are referred to the nuclide given in brackets**

	Bq(^{238}U)/g	Bq(^{226}Ra)/g	Bq(^{232}Th)/g	Bq(^{232}Th)/g
practise /2/;				
• only long-lived isotopes	78	119	159	20,3
• all isotopes included	32,1	34,7	49,3	0,67
transport regulations /3,4/	9,3	16,7	22,2	2,8
radioactive waste /2/	0,096 (*)	0,12	0,40 (*)	0,022 (*)
restricted ~~exemption~~ clearence for waste deposits /5/ (**)	0,057	0,092	0,42	0,010
unrestricted ~~exemption~~ clearence level /5/ (**)	0,007 (!)	0,009 (!)	0,016	0,0013 (!)

(*) calculated with U_{nat}, Th_{nat} and long-lived daughters (without ^{234}U, ^{228}Th)
(**) secular equlibria considered

Table 3: **Most relevant radionuclides for internal exposure of adult persons with materials given in Table 1. The figures represent the quota of radionuclides to the total exposure according /6, 7/.**

	U dump material	Ra scale	soil, Th contaminated	soil, Th-Ra contaminated
Ingestion; Germany	^{210}Pb: 48 %	^{210}Pb: 55 %	^{232}Th: 53%	^{210}Pb: 48 %
	^{210}Po: 16%	^{210}Po: 19%	^{228}Ra: 27%	^{210}Po: 16%
Ingestion, EU	^{210}Po: 45 %	^{210}Po: 45 %	^{228}Ra: 62%	^{210}Po: 50 %
	^{210}Pb: 26%	^{210}Pb: 26%	^{232}Th: 21%	^{210}Pb: 29%
Inhalation, Germany	^{230}Th: 27%	^{228}Th: 82%	^{232}Th: 80%	^{232}Th: 40%
	^{227}Ac: 26%	^{210}Pb: 5 %	^{228}Th: 17%	^{227}Ac: 36%
Inhalation, EU	^{230}Th: 54%	^{228}Th: 50%	^{232}Th: 65%	^{227}Ac: 26%
	^{227}Ac: 26%	^{226}Ra: 20%	^{228}Th: 23%	^{232}Th: 24%

Site remediation and waste disposal

Generally, there must be a demonstrable public danger or a environmental hazard in order to initiate a site remediation. This can be a result of the radioactive contamination but also the result of toxic chemical substances. Frequently, contaminated material becomes a problem if contaminated areas are included in developing measures.

In all such cases the bottlenecking have been proved as an effective solution for disposal. The significance of this solution becomes evident in view of

- the costs for deposition in a final repository of more than DM 20 000 per ton (in 1997),
- the lack of available repositories in Germany (the single operating repository ERA Morsleben accepts only weak alpha activities and practically no thorium).

The main options which we have checked are:

- extraction of radionuclides and production of saleable metals or chemical compounds
- volume reduction by soil washing or other separation methods
- use of contaminated materials
- on site reduction of releases by isolation or immobilisation
- removal and off-site deposition

The extraction of radionuclides and production of saleable metals or chemical compounds seems to be the most favourable option in order to avoid the deposition of radioactive contaminated materials. The only radioactive materials which are technically used are uranium and thorium. From these elements the demand of thorium is very restricted and the reprocessing of low-grade material contaminated with other toxic components will not be accepted by the producers.

Soil washing procedures can be used for volume reduction of contaminated materials. The ideal procedures should be characterised by the effective reduction of radioelement concentration in the largest part of the mass treated. Because of mass balance this is accompanied by production of a highly enriched concentrate, which have to be handled and disposed as a significant radioactive material. Following, soil washing methods are preferably to recommend if no alternatives remain. In such a case the different chemical behaviour of the radioelements (especially Ra - Th - U) makes single step chemical extraction usually less efficient. More efficiency could be expected from physical methods like high pressure soil washing. However, the results of several tests made by different groups and evaluated by us demonstrated clearly that the yield of washing procedures strongly depends on the individual character of soil and the kind contamination.

Carefully separation of different contaminated materials during dredge by accompanying radiological measurements, eventually followed by a mechanical sorting of macroparticles like bricks and stones can effective reduce the volume without to left behind highly enriched concentrates.

The use of radioactive contaminated materials is possible under controlled conditions or after restricted exemptions. Controlled conditions exist in industrial ranges licensed under the Radiation Protection Act. We could not find any sensible application for contaminated soils in this field. The utilisation in other fields requires a permission of the competent authority. Such permission is usually based on the demonstration that the planned use will not exceed critical doses for workers and/or persons of the general population. Typical kinds of use are the refilling of boreholes or the fulfilling of mine caverns. These possibilities are restricted on materials generated under the regulations of the Federal Act on Mining /8/. The use of contaminated material outside of mining frequently runs against objections. Nevertheless, a use of material contaminated by Th - Co - V and organics for the grounding of an industry building /9/ demonstrates the general feasibility of this option. One of the requirements of this measure was the immobilisation of the pollutants by special cementation.

The most favourable way of waste disposal seems to us a deposition on dump sites. It guarantees the compliance of some conditions which can difficulty be fulfilled at other surface environments. Some of the very important conditions in this regard are the usually well known natural conditions of dump sites which enable detailed exposure analysis and the possibility to realise optimal techniques of deposition. Additionally, the area of a dump site can be assumed to be restricted for use over long time.

The licence for deposition of materials containing enhanced levels of natural radionuclides requires detailed exposure analysis. By such analysis the limitation of doses on the 10 µSv level (de minimis) must be shown in a form acceptable to the competent authorities. The radioecological models and basic scenarios for such calculations are described in /9/. The acceptable concentrations increase considerably if site specific parameters are included in the calculations.

Aspects of de minimis calculations

Sensitive pathways concern the inhalation of dust and external radiation by workers. These pathways can be effectively reduced by packages and/or solidification of material.

A crucial part of de minimis calculation regards is the long-term assessment of prognostic public exposures via groundwater. If this pathway has to be considered for decay series isotopes, some special aspects must be taken into account.

Assuming any location with a material enriched in natural radionuclides ("source"), the additional exposure by drinking or other applications of water depends on the dissolved radionuclide concentration in such water. Migration processes are the typical processes which influence this concentration.

- The radioecological standard model considers this migration as the advective transport of a reversible sorbed single tracer substance. The characteristic parameter of this model is the K_d-value or the retardation factor R. This "isolated migration" model is applicable in the case of decay series nuclides, if the travel time remains lower than the half-life of the isotopes, i.e. for long-lived nuclides.
- Radioactive decay permanently generates short-lived daughter nuclides. In this way short-lived nuclides can, effectively, reach springs or wells far away from a source, if a long-lived precursor exists. However, the dissolved concentrations of the short-lived nuclides do not correspond to the radioactive equilibrium. Rather, they are determined by the water-rock interaction. For a homogeneous aquifer an analytical solution of the "coupled migration" was derived and is used in our radioecological models /10/.

This concept, however, requires corresponding site investigations and parameter determinations. The naturally occurring radioactive disequilibrium may be used to estimate the migration parameters for the real hydrochemical system. This method can favourably be integrated in the investigation program.

References

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