
Methods for calculation of radiation exposure in case of re-use and disposal of NORM waste

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Abstract:

While implementing EURATOM guideline 96/29 in the German legislation, in June of 2001 an essential pre-condition was created for re-use or disposal of NORM and TENORM waste. An essential progress has been achieved allowing to re-use NORM and TENORM or to remove it together with other residues and waste, either if their specific activity do not exceed the limits defined in the Radiation Protection Ordinance (StrlSchV) or the dose calculated for a given application lead to a value less than 1 mSv/yr.

While the limits of specific activity according to StrlSchV, Annex XII part B or C were derived from basic environmental pathway models having conservative parameters an excess of the dose limit can be excluded. Otherwise, if the specified limits in terms of concentration are exceeded, than the release from radiological supervision can only be permitted if the dose limit calculated on the basis of less conservative assumptions and parameters is observed.

This paper describes special methods and models which were developed by GRS for the calculation of dose to man when disposing or re-using different NORM wastes in dependence on there physical chemical characteristics, leaching behaviour, nuclide vectors and, if appropriate, radon emanation. Special attention is also given for restrictions or limitations arising from other fields of low, like soil protection or regulations on hazardous waste.

1 INTRODUCTION

The general provisions as well as the single paragraphs of the German Radiation Protection Ordinance (StrlSchV) which are directed towards natural radiation sources are based on recommendations of international organisations, especially according to Title VII of the Directive 96/29 EURATOM, but also according to radiological protection principles developed by the German Commission on Radiological Protection (SSK). Accordingly, radiation protection measures must be taken into account if the existence of natural radiation sources could lead to enhanced radiation exposure. For that purpose regulations were agreed. According to the StrlSchV materials which contain natural radionuclides are divided into residues (§97) and other materials (§ 102). Residues are defined as materials which are resulting from different technological processes listed in Annex XII part A of StrlSchV with specific activity of at least 0.2 Bq/g. This list comprises sludges and scales from the exploitation of natural gas and crude oil and residues and by-products from mining and processing of raw phosphate, of iron and non-iron ores and of minerals. Other materials are considered as radioactive material only if they are a source of enhanced exposure and if countermeasures must be taken into account by the responsible authority.

The defined concentration level for "enhanced" is derived from the annual dose limit for members of the public considering basic exposure scenarios. In Germany and some other EU member states this dose limit is 1 mSv/yr from which a minimum specific activity of 0.2 Bq/g per radionuclide of the U238 decay chain and the Th232 decay chain was derived. Subsequently, natural or processed materials with lower specific activities turn out from radiation protection regulation.

The above described methods for re-use or disposal of NORM and TENORM are only applicable if they are in agreement with the legal frame conditions. That concerns not only the regulations on radiological protection but also those on recycling and storing of common residues and waste as well as on the shipment of dangerous goods. In every case the precondition for the implementation of each practice is the release of the concerning residue from the radiological supervision according to § 98 of StrlSchV in connection with Annex XII, part A (kind of material) part B (maximum permissible specific activity), part C (maximum permissible specific activity of the total material of an landfill) and/or part D (keeping of the dose limit of 1 mSv/yr). The procedure necessary for the release of such materials from radiological supervision is much more complicated than assumed, because the regulations among the different fields of law are not well tuned or contradicting.

When the limits for release of NORM waste from radiological supervision according to Annex XII part B or C of StrlSchV are observed a simplified licensing procedure for release can be applied. The limits given in Annex XII depend on the foreseen way for disposal or re-use. Otherwise, when such a limit is exceeded than it must be proved that the selected option leads to a dose less than the limit of 1 mSv/yr as provided in part D of Annex XII. Nevertheless, part D gives only a common explanation on how the dose calculation should be made without an exact calculation scheme, e.g. according to radioactive discharges from nuclear installations. Recently, only the so called "Procedure for calculation of radiation exposure resulting from handling with mining related materials" is applicable, while this guideline is primarily directed towards the calculation of dose to man by heaped up waste rock materials from former uranium mining activities in Germany.

2 BASIC PRINCIPLES

The development or adaptation of methods or parameters describing the release and spreading of natural radionuclides from the different NORM or TENORM are based on existing models and parameters with special emphasis on uranium mining and milling legacies, e.g. the report [1] where the limits for release of NORM waste from radiological supervision according to Annex XII part B and C were derived.

Furthermore, the "Guideline for radiological investigation and evaluation of mining related legacies" [2, 3] as well as the results from actual national and international literature [4, 5, 6] were taken into consideration.

Annex XII part D of StrlSchV provides generic principles and assumptions for the estimation of radiation exposure, namely:

- application of realistic exposure pathways and frame conditions for the calculation of dose to members of the public with special emphasis on the principles of Annex VII part B and C of StrlSchV;
- Inclusion of all exposures of single persons of the public which may appear for all relevant cases of re-use or disposal of other residues with enhanced concentrations of natural radionuclides;
- Inclusion of all exposures of single persons of the public which may appear in case of disposal by corresponding measures as waste treatment, temporary storage or when performing the disposal work.

3 MODELS AND PARAMETERS FOR CALCULATION OF RADIONUCLIDE RELEASE AND SPREADING

Existing models and model parameters for the calculation of radionuclide release and dispersion via air, groundwater and surface water (e.g. [1, 2, 3, 7, 8, 9]) were evaluated with respect to their applicability for re-use or disposal of the different kinds of NORM and TENORM.

3.1 Essential parameters for radionuclide release and dispersion

While the NORM wastes in Annex XII part A of StrISchV are grouped according to their origin and industrial branche our approach is primarily directed towards a grouping according to their physical chemical properties arising from the underlying technological process (table 1).

Table 1: Grouping of residues according to their physical chemical features

Group	Kind of residue	Assignment to Annex XII part A StrISchV
A	Residues from mining and (mechanically) milling	
A1	Heaped-up materials and related residuals	3a): Surrounding rocks, sands, 3b): Minerals
A2	Sludges und dusts	2: <i>Dusts (no residuals according to Annex XII)</i> 3a): (elutriated) dusts
B	Residues from chemical processes	
B1	Coates, scales	1: Scales
B2	Sludges und dusts	1: Drilling sludge 2: Phosphogypsum and processing sludge 3a): Tailings
B3	Sludge and solid residues	Residues from water treatment
C	Residues from thermal processes	
C1	Sludge und dusts (elutriated dusts)	2: Air filter dust 3a): Dusts from melting, blast furnace dust 4: Dust, sludge and fly ash from flue gas treatment
C2	Slag	2: Silicate slag 3a): Slag from melting b): cast-, sinter- and press molded parts
C3	<i>(bottom)ash</i>	<i>not considered</i>

3.2 Common physical chemical parameters

3.2.1 The radioactive equilibrium within the natural decay chains

A radioactive steady state is considered for the residues of group A1 and A2 (see table 1). These heaped-up materials are of mining origin or resulting from mechanical ore processing. The residues resulting from chemical processes (group B) are characterised by non-equilibrium conditions reflecting the chemical property of each chemical element, i.e. it is equal for all isotopes of one and the same element. These NORM and TENORM are either the result of natural chemical processes as for scales by co-precipitation of radium isotopes with barium to barium-radium-sulfate (baryte) or of technological processes as phosphogypsum from acidic treatment of raw phosphate.

The nuclide relations in residues of group C result from the volatility of each element, i.e. with enrichment of the isotopes of the high volatile elements lead and polonium in dust or fly ash. The following table 2 contains an overview on the different equilibrium and non-equilibrium relations of NORM wastes as a result of the technological process of their appearance. While for the air path and the path of direct ingestion the actual nuclide relationship within the natural decay chains will be applied, for the water path the temporal changes of the nuclide relations must be taken into account when considering a long observation period of some hundred years.

Table 2: Nuclide relations for different NORM and TENORM

Group	Nuclide relations
A1 A2	U238sec → same specific activity of all nuclides of U-Ra decay chain For U235 decay chain: U235 : U238 = 1 : 20 Th232sec → same specific activity of all nuclides of Th decay chain
B1	Sulfate scales: Ra226+ → same specific activity of Ra226, Rn222 and short lived RnDP; „regrowing“ of Pb/Po210 Ra228+ → same specific activity of Ra228 and Ac228; „regrowing“ of Th228 by “dynamic equilibrium” Lead scales: Pb210++ → same specific activity of Pb210, Bi210 and Po210 Carbonate-, silicate- and fluoride scales: like sulphate scales
B2	1.: Ra226+ → same specific activity of Ra226, Rn222 and short lived RnDP; „regrowing“ of Pb/Po210 Ra228+ → same specific activity of Ra228 and Ac228; „regrowing“ of Th228 by „dynamic equilibrium” 2.: Acidic leaching of phosphate: Ra226++ → same specific activity from Ra226 to Po-210 U238+ → same specific activity of U238 and U234 with initially 20% of Ra226 activity 3.: red mud: U238sec → same specific activity of all nuclides of U-Ra decay chain
B3 (water treatment sludge)	Depending on the applied separation method U238+ and/or Ra226+ and/or Ra226++, and/or Pb210++
C1	Pb210++ → same specific activity of Pb210, Bi210 and Po210; for fly ash: U238++ → initially 20% of Pb210
C2	U238++ → same specific activity from U238 to Ra226; „regrowing“ of Pb/Po210 Th232sec → same specific activity of all nuclides of Th decay chain

3.2.2 Homogeneity of radioactivity within the residual

A homogeneous distribution of radioactivity can be assumed for all kinds of slag and ash but also for all materials which are no waste rock. Besides the residues of group B (chemical processes) and C (thermal processes) it is also valid for sludge and dust from mechanical treatment of ores and minerals (group A2).

3.3 Model parameters for the air path

3.3.1 The concentration factor A_{Inh}

The concentration factor A_{Inh} is defined in [1] as a dimensionless relationship between the specific activity of a radionuclide in the dust fraction of a waste rock sample (≤ 0.02 mm or ≤ 0.063 mm) and the specific activity of the same nuclide in the gross sample. Based upon numerous analyses a conservative value of 4 for the concentration factor A_{Inh} for the dust fraction was recommended in [10] if no site specific measurements are available.

The concentration factor A_{Inh} according to the above mentioned definition does not consider the percentage of the dust fraction in the gross sample. Its application is therefore confined to heaped up surrounding rock having a diversified grain size spectrum. The problems arising from the application of A_{Inh} according to [1] are in detail discussed in [11].

For all other TENORM emerging from chemical or thermal treatment as well as for dust from mechanical ore processing (milling) the radioactivity is homogeneous distributed within the residual, i.e. the concentration factor A_{Inh} is equal to 1.

3.3.2 The emanating fraction

According to [1] the emanating fraction was assumed as 1% for all kinds of slag and of 20% for all other residues. These values must be considered in more detail both for slags and for dusts, sludges and scales.

For scales first results are available in [12] which lead to values between the limits of 1% and 20% in dependence on their texture and chemical properties. A wide range of the radon emanation fraction was observed for slags. According to [13] values between less than 1% and 30% were measured. This high variability is based on the applied technological processes which result to vitreous slag (emanation fraction much less than 1%) from copper melting or to highly porous blast furnace slag (emanation fraction up to 30%).

3.3.3 Dust deposition velocity

The application of the value of dust deposition velocity v_g according to [10] must be corrected especially for fine dispersed residues. For some residues with a lower AMAD than in [10] a lower v_g -value should be applied. Recommended values in [14] in dependence on the aerodynamic particle diameter (AED) contains table 3. If measuring values on the grain size distribution are available the deposition velocity can be taken from [14].

Table 3: Deposition and sedimentation velocity in dependence on the AED [14]

Class	AED in μm	TAL [14], Annex 3, No 4	
		Deposition velocity v_g in m/s	Sedimentation velocity v_s in m/s
1	< 2,5	0.001	0
2	2,5 to 10	0.01	0
3	10 to 50	0.05	0.04
4	> 50	0.2	0.15

3.4 Model parameters for the water path

For the applied models and model parameters to estimate the radionuclide drainage from landfills and run-off via water paths the following conditions were derived.

3.4.1 Release via seepage

In [1] it is assumed that for waste rock materials the nuclides from Th230 to Pb/Po210 are available at 10 % related to U238 and U234 in seepage water under oxidizing conditions. For reducing conditions the uranium release is lower, nevertheless this conservative assumption covers all possible environmental conditions.

The above given percentage is not applicable for phosphogypsum, water treatment sludge and all kind of sludge from wet chemical processes. For such residuals and for those from thermal processes further investigations are needed.

3.4.2 Leachability

The assumption in [1] of a leaching rate of 0.1% uranium per year from landfills or waste rock heaps is probably only valid for surrounding rocks and minerals. For phosphogypsum and water treatment sludge the leaching of uranium and of nuclides of the Th decay chain is negligible due to accumulation in the fertilizer or immobilisation in the treatment sludge.

As for uranium the leaching rates given in [1] for other nuclides of 0.01% per year are also only valid for surrounding rocks and minerals while for sludge from wet chemical processing the Ra and Pb isotopes are accumulated in the residual itself.

The leaching rate from slag of 0.01% per year according to [1] is only applicable for a limited number of slags. The values are higher for porous slags and much lower for inert slags from copper melting (between 0.001% and 0.0001% per year for all long lived radionuclides).

4 EXPOSURE PATHWAYS

The following exposure pathways will be considered in dependence on the corresponding exposure scenario.

- External exposure by gamma-radiation

The external exposure will be considered for workers at all scenarios of re-use and disposal except for residues with low content of gamma emitters (dusts with enrichment of lead and polonium). For members of the public this path will only be considered for house construction and landscape construction works.

- Dust inhalation

The dust inhalation is not applied to workers in case of underground storage (waste in closed containers). For members of the public it is not relevant for house and road construction scenarios. In case of disposal options the dust inhalation is only applied if the deposition velocity v_g is less than 0.01 m/s.

- Inhalation of radon and radon decay products

This path is relevant for all exposure scenarios of workers. For members of the public it is not relevant for road construction and for all disposal scenarios.

- Direct ingestion of contaminated particles

The direct ingestion of soil particles will only be considered for disposal scenarios for workers during dumping of low hazardous materials at landfills. For members of the public it is only considered for the re-use option on landscape works.

- Drinking water consumption

This path is only taken into account for members of the public in case of road and landscape construction scenarios. It is not relevant for all disposal scenarios due to the strong requests for geological barriers according to the technical norms on toxic waste disposal [14].

- Ingestion of vegetables via irrigation

The same assumptions as for drinking water consumption were met.

- Ingestion of vegetables via dust deposition

This is only taken into account for members of the public in case of landscape construction works and for waste disposal at landfills if v_g is less than 0.01 m/s.

5 EXPOSURE SCENARIOS

5.1 Re-use

The limits for radiological supervision of $C = 0.5$ Bq/g according to Annex XII part B No 2. and of $C = 1$ Bq/g according to Annex XII part B No 1 of StrlSchV were derived from scenarios described in [1] for road and house construction. For further scenarios of re-use the conditions of the corresponding disposal scenarios were applied.

5.1.1 Above ground mine backfilling and landfill construction

These re-use options result from Annex XII part B of StrlSchV with the limits for radiological supervision of $C = 1$ Bq/g or $C = 0,5$ Bq/g if more than 5,000 t/yr will be utilized in the drainage area of an usable aquifer. These limits for radiological supervision were derived in [1] from comparable disposal scenarios, namely:

- Working on landfill or waste rock heap where residues with enhanced natural radioactivity are disposed,
and
- Living in the vicinity of such a landfill or waste rock heap, stay in the backyard next to it, cultivation and irrigation of vegetables as well as use of the surrounding area as meadow land and stay on top of the uncovered heap or landfill.

5.1.2 Building materials and additives

These re-use options are based on Annex XII part B No 1. and No 2. StrlSchV with the limits for radiological supervision of $C = 1$ Bq/g or $C = 0,5$ Bq/g. On principle usable as building materials or concrete additives are the following residues due to their geomechanical and physical properties:

- Sludge and gypsum from wet processing of raw phosphate
- Slag and probably dust from thermal processes
- Surrounding rocks, sands and minerals

For the derivation of the limits for radiological supervision of $C = 1$ Bq/g or $C = 0.5$ Bq/g in [1] scenarios on the utilization of NORM wastes for house construction and for road and landscape construction works were considered.

5.1.3 Backfilling materials in underground mines

This option of re-use is related to Annex XII part B No 3 of StrISchV with a limit for radiological supervision of $C = 5$ Bq/g.

This limit was derived according to [15] by means of a scenario for waste disposal in an underground mine. In that case the water path is not considered due to the high long term safety standards which are regulated by law for an underground waste disposal site. The modelling of the long term release of pollutants is part of the safety case studies for the concerning mine which must be delivered in frame of the planning approval procedure.

5.2 Disposal

The following disposal options will be considered.

5.2.1 Mono-landfill

This disposal option must be considered in relation to Annex XII part B No 1 of StrISchV with the limit for radiological supervision of $C = 1$ Bq/g.

The limit of $C = 1$ Bq/g is based upon the same assumptions as described for the re-use as above ground mine backfilling material and landfill construction material, i.e. working at landfills or waste rock heaps and living in the vicinity of a landfill or heap.

According to the German regulations for landfills for toxic wastes no catchment area may be affected and the minimum distance to the next residential area must be more than 300 m. The stay on the uncovered surface of the landfill is not considered because there is no trespassing.

5.2.2 Hazardous waste site

This option is basing upon Annex XII part C of StrISchV with the mean activity values of $C^M = 0.05$ Bq/g, $C^M = 0.1$ Bq/g or $C^M = 1$ Bq/g.

The C^M values are defined as weighted means of specific activity of waste without enhanced radioactivity (toxic waste) and that with enhanced radioactivity. The above mentioned values of C^M are also based upon the scenarios described for the re-use as above ground mine backfilling material and landfill construction material.

Further aspects for the determination of the different values of C^M concern the size of the landfill and the potential influence of the waste to an aquifer. As already explained for mono-landfills the stay on the landfill is not considered.

5.2.3 Underground waste storage

This disposal option corresponds to Annex XII part C of StrISchV with the weighted mean of $C^M = 5$ Bq/g.

In this case the same scenario as for re-use of NORM and TENORM as backfilling material is considered.

The basic assumption for modelling the pollutant dispersion in an underground mine used for storage of toxic wastes is that after closure of the mine a water penetration takes place. On this basis the expected spreading of a couple of pollutants into the biosphere is calculated for geological periods. Contrary to a radwaste repository the period under consideration is not fixed for an underground disposal site of toxic waste due to the assumption of retrievability of the wastes. The primary leaching behaviour of the solid waste has to meet given physical chemical conditions according to the German regulations on waste storage. Therefore no limitation of the concentration of a pollutant exist in case of an underground storage site in contrast to the above ground storage of toxic waste. Also no immobilisation measures must be taken into account if the given values for leachability are met.

6 SUMMARY AND CONCLUSIONS

The most of NORM and TENORM can be released from radiological supervision according to § 98 of StrISchV in connection with the limits of specific activity given in Annex XII part B and C by means of an simplified procedure. Even if these limits are exceeded the compliance of the corresponding dose limit of 1 mSv/yr must be proved by means of a dose model according to Annex XII part D of StrISchV.

Further problems for the licensing process arise from the superposition of different legal aspects which have to be taken into account.

Because the provisions of Annex XII part D contain only basic assumptions on how the dose should be calculated and hint to the application of model parameters described in Annex VII of StrISchV there was a need to develop or improve models for calculation of dose to workers and to members of the public using more realistic assumptions and parameters for each option of re-use and disposal of NORM and TENORM.

For the air path the radon emanation fraction is the crucial point for the relevance of radon inhalation dose to the total dose. According to new measurements the radon emanation fraction shows a wide range between much less than 1% up to 30%.

The most relevant parameter for the water path is the leaching rate of the pollutant. While for underground storage no limitation of the concentration or immobilisation measures are needed if the maximum leaching values given by law are met for a above ground storage or for re-use options the leachability must be in many cases reduced to fulfill the given legal provisions, both for radiological and for waste management reasons.

When applying the proposed dose calculation methods the predominant part of NORM and TENORM in Germany can be released from radiological supervision and only a small part must remain under the scope of StrISchV.

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