Discharges from NORM industries in Germany: estimate of doses to members of the public

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Abstract. Council Directive 2013/59/Euratom (the European Basic Safety Standards, or BSS) has removed the previous distinction between practices and work activities. Henceforth the dose limits for public exposure shall apply for the sum of annual exposure for members of the public resulting from all authorized practices. From this it follows that exposure levels being significantly smaller than the dose limits have to be considered, where appropriate, and the NORM industries, in particular the discharges of radionuclides from this industries into the environment have to be re-assessed. In transposing the BSS into national law, Art. 23 requires Member States, *inter alia*, to identify classes or types of practice involving NORM that may need to be regulated because they lead to exposures of the public that cannot be disregarded from a radiation protection point of view. Discharges are part of a practice and, therefore, may be subject to regulatory control. A comprehensive investigation of discharges in Germany included the 16 sectors of NORM industries listed in Annex VI of the BSS, and additional sectors that have been known in Germany for potentially being of radiological concern. This paper provides an overview of the approach, methods and results of the research project. A conservative screening tool was developed and applied to select NORM industries that may safely be excluded from a more detailed analysis. NORM industries that passed the screening stage were then analysed in detail with respect to the source term of radioactive discharges. The dispersion of radioactivity in the environment was described using advanced models such as air quality code ARTM, and all relevant pathways were considered for dose estimates. It is important to note that the project deliberately avoided a site-specific analysis but adopted a "generic" approach covering a wide range of sources terms and environmental dispersion conditions encountered in Germany.

KEYWORDS: NORM; discharges; atmospheric dispersion; public dose estimate.

1 INTRODUCTION

Council Directive 2013/59/Euratom (the European Basic Safety Standards, or BSS) [1] have removed the previous distinction between practices and work activities. This follows the concept adopted by the International Commission on Radiation Protection (ICRP) in its Recommendation No. 103 [2], which distinguishes planned, existing and emergency exposure situations. Henceforth handling and processing NORM is covered by the concept of practices.

Henceforth the dose limits for public exposure shall apply for the sum of annual exposure for members of the public resulting from all authorized practices, according to Art. 12 of the BSS. Discharges that are intrinsically part of a practice and may then be subject to regulatory control. Therefore practices involving naturally-occurring material (commonly but not entirely correctly known as "NORM industries") may need to be taken into consideration if exposure of members of the public resulting from discharges cannot be disregarded from a radiation protection point of view. Art. 12 (1) of the European BSS specifies that "the dose limits for public exposure shall apply to the sum of annual exposures of a member of the public resulting from all authorised practices". Hence, exposure of the public due to multiple sources must be taken into consideration. Consequently, in transposing the BSS into national law, Article 23 requires Member States *inter alia* to identify classes or types of practice involving naturally-occurring material that may need to be regulated. As guidance which practices may be relevant, Annex VI provides a list of industrial sectors that shall be taken into account. However, Member States may need to include other industrial sectors over and above the Annex VI of the BSS, if they are suspected of significant contributions to public exposures.

In Germany, the Federal Office of Radiation Protection (BfS) is responsible for investigating such issues and contracted IAF-Radioökologie GmbH (IAF), an accredited radiological laboratory and

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radiation safety consultancy, to implement a research project entitled “Estimation of potential public exposure due to discharges from NORM industries”. IAF investigated the 16 industrial sectors of Annex VI of the BSS, and additional sectors that have been known in Germany for potentially being of radiological concern. In total, water- and airborne discharges from 26 industrial sectors were analysed with respect to the public exposures caused.

A preliminary “threshold of relevance” less than the dose limit for members of the public is required to account for the summation of multiple exposures. Within this project, an annual effective dose of 100 µSv/a received by the representative person as defined in Article 4 (89) of the BSS was used as a threshold for what is considered radiologically relevant (preliminary “relevance threshold” or working hypothesis for the purposes of this study). If doses resulting from multiple sources are added, this relatively low threshold of 100 µSv/a provides a sufficiently conservative framework. It is important to note that the project deliberately avoided a site-specific analysis but rather adopted a “generic” approach, i.e., it covered a wide range of situations that may typically be encountered in Germany, in terms of sources terms and environmental dispersion conditions of air- and waterborne discharges. While the research project covered discharges of dust, radon and water, this paper focuses on dust-borne radioactive discharges. Waterborne radioactive discharges and discharges of radon from the industrial sectors considered within this project have been found to be less relevant.

This paper provides an overview of the approach, methods and results of the research project. It largely follows the outline of the final report that will be available on the official BfS website [3]. Due to limited space, it provides little information on intermediate steps, and largely omits discussion of peculiarities of certain industrial sectors or processes. Readers who are interested in this in-depth information are referred to the final report of the project [3], or are encouraged to contact the authors.

2 IDENTIFICATION OF “NORM INDUSTRIES” THAT ARE CURRENTLY ACTIVE IN GERMANY

Annex VI of the EU BSS [1] provides some guidance on industrial sectors involving NORM, including research and relevant secondary processes that shall be taken into account when identifying sectors with discharges that may lead to exposure which cannot be disregarded from a radiation protection point of view. Since obviously not all of the sectors listed in Annex VI are currently active in Germany, those that are active had to be identified under the project in a first step. In addition to the above 16 sectors, another 10 sectors known to have existed, or are still existing, in Germany were deemed to be worth considering by BfS, based on historic evidence and experience. These additional sectors include, *inter alia*, the optical industry, the production of abrasives, ceramics and refractories, the use of thoriated welding electrodes, aluminium production including bauxite processing.

Whether these sectors still today are using materials with elevated levels of natural radioactivity, and may therefore have relevant discharges of natural radionuclides, was a question that had to be answered under this project. Apart from the mere existence of active operations in Germany belonging to the above sectors, it also had to be determined whether there are any relevant discharges from the operations. In answering this second question, a broad range of sources were used, including company information available in the public domain and interviews with industry representatives at both company and industry association level, best practice reference (BREF) documents of industrial sectors (e.g., the series of BREF documents released by the European Commission’s Joint Research Centre on various industries), and finally the experience accumulated within the project team from more than two decades of working with NORM industries. It was found that with the exception of extraction of rare earths from monazite, thermal phosphorus production, and phosphoric acid production, all sectors listed in Annex VI of the BSS are active in Germany and are associated with discharges that may, in principle, be radiologically relevant.

In order to identify those sectors that can be safely excluded from a more detailed analysis, a series of steps were taken that are briefly described in the next section.

3 SCREENING OF NORM INDUSTRIES REGARDING THEIR RADIOLOGICAL RELEVANCE

The most relevant exposure pathways and radionuclides were identified that would lead to the highest effective doses in any of the age groups <1, 1-2, 2-7, 7-12, 12-17, and >17 years, respectively [4]. It was concluded that dust settling on agricultural products and the subsequent consumption chain "mother – breast milk – infant" leads to the highest effective doses, and infants (age bracket <1 year) are the representative person as defined by Art. 2 (89) of the BSS [1]. Unsurprisingly the nuclide with the highest contribution is Po-210 due to its high ingestion dose conversion factor. Due to its volatility, Po-210 is also predominantly present in dust from all typical high temperature processes, e.g., from cement kilns, furnaces, or smelters.

A deposition rate of 1 Bq/(m² a) of Po-210 on agricultural products leads to an effective dose of 39 µSv/a in infants (age group "<1 year"). For later reference (see Section 6) it is noted in passing that the same settling rate of 1 Bq/m²/a of Pb-210 on agricultural areas leads to an effective dose of 7 µSv/a in infants. Other exposure pathways such as inhalation of dust, or continued accumulation of radioactive dust on soil, contribute significantly less to the total effective dose and can be dismissed for the purposes of this study. A detailed breakdown of dose contribution from the various exposure pathways can be found in the full study [3] and is omitted here for brevity.

For airborne discharges, a simple, but sufficiently conservative, Gaussian plume dispersion model from [5] was used to calculate upper bounds of the dust settling rate. The model contains several parameters such as height above ground level of the point of emission, dust particle settling velocity that is closely correlated with the dust particle size, and angular distribution of wind direction. In order to span a sufficiently wide but conservative range of dispersion conditions, chimney heights of 20 and 100 meters, respectively, were used for the initial screening step. For dose estimates, maximum dust deposition rates were used, which for chimney heights of 20 and 100 m, respectively, occur at distances of 100 and 600 m from the source, respectively. It was also conservatively assumed that dust was only transported within a single angular element of 30 degrees, and a particle settling velocity of 0.01 m/s was used, which is at the upper end of settling velocities used in the literature for dust particles in the industry sectors in question, and in applicable technical standards [6].

The maximum sum of dry fall-out and wet wash-out of Po-210 from a standard source strength of 1 GBq/a is of the order of 4 Bq/(m² a) and 200 Bq/(m² a), respectively, for the above chimney heights of 100 and 20 meters, respectively. This allows, in turn, to define conservative screening thresholds for airborne dust discharges: A discharge of 15 MBq/a of Po-210 from a chimney of a height of 20 m, or 700 MBq/a of Po-210 from a chimney of a height of 100 m, respectively, lead to an annual effective dose of approximately 100 µSv in infants (representative person).

4 ESTIMATE OF DISCHARGES FROM POTENTIALLY RELEVANT INDUSTRY SECTORS

4.1 Data sources

In order to *quantitatively* estimate radioactive discharges, a broad range of information sources were used, including but not limited to environmental permits of operations including permit conditions that limit airborne and/or waterborne discharges, emission data from the Pollutant Release and Transfer Register (PRTR) set up under European Directive 2006/166/EC [7], information provided by plant operators or industry associations, process technology textbooks and good practice handbooks of some industrial sectors both in Europe and overseas, data on the specific activity of raw materials, wastes, and discharges, that can be found, *inter alia*, in articles and presentations of NORM and IAEA conferences.

Information on non-radiological parameters of discharges such as dust streams (tons per year, particle size distribution) or wastewater streams (m³/h, concentration of total suspended solids), is publicly available to a limited extent for some but not all industries. However, information on radiological parameters such as specific activity of dust particles or dissolved and/or particulate-bound radionuclides in waste water was extremely difficult to come by for some industries. Estimates using analogues and simplified radioactivity balances of the industrial process in question based on assumed radiological properties of raw materials were needed to close data gaps. If no data on airborne discharges (source term expressed in activity per unit time) of natural radionuclides was forthcoming, the discharged activity was estimated using one, or a combination, of the following methods depending on the data available:

- Maximum mass flow of dust particles (kg/h) allowed under the environmental permit of an operation, or obtained from published data such as the PRTR [7], multiplied by the specific activity of the dust (Bq/g). The specific activity of the dust may be estimated using the specific activity of the raw material and the typical mass reduction factor of the process.
- Reverse modelling of (non-radioactive) dust deposition data (g/m^2a) available from environmental monitoring programs to obtain the mass flow (t/a), e.g., environmental monitoring data of conventional lead deposition around a lead smelter. The radioactive discharge was then obtained from multiplying the mass flow by the specific activity of the dust particles (Bq/g),
- Consumption rate of raw materials (t/a) were multiplied by the specific activity of the raw material (Bq/g), resulting in an activity flow entering the operation. It is then conservatively assumed that the nuclides of volatile elements such as Po-210 and Pb-210 leave the process attached to dust particles. An industry-specific retention rate of dust filters (see Section 4.3) is applied to estimate the actual discharge through the chimney.

Discharges from some sectors are constrained by general environmental protection or emission control regulations, and/or best environmental protection practice adopted by most industrial operators in Germany. Such considerations that are not related to radioactivity in the first place are very useful in determining upper bounds to emissions in general. For example, according to the Technical Instructions on Clean Air [8], the limit of the average annual concentration of PM10 in ambient air is 40 $\mu g/m^3$, and total dust emissions (including the PM10 fraction) must not exceed either a mass stream of 200 g/h or a concentration of 20 mg/m^3. As it was assumed in the study that industry operators comply with these limits as part of their environmental permits, these constraints indirectly put upper limits on radioactive discharges. In turn, this fact can be used to determine the radiological relevance of an industry sector.

4.2 Industry sectors that are subject to more detailed investigation

A combination of all information sources and approaches described in Section 4.1 have enabled the study team to draw relatively robust and plausible conclusions on the discharges from most industries listed in Table 1, expressed in GBq/a, at least up to the order of magnitude. Details of the discharges from each industry sector can be found in the study [3] and are not reproduced here for limitations on space. Upon application of the screening method described in Section 3, Table 1 provides an overview of those industry sectors with potentially relevant discharges that exceed the conservative screening threshold and are therefore subject to more detailed investigation. Note that the data in Table 1 describe a base case that is complemented by a sensitivity analysis in Section 4.3.

Table 1: Industry sectors that exceed the screening threshold of radioactive discharges and therefore are subject to more detailed investigation, base case of production figures

Potentially relevant sectors	**Potentially relevant processes and associated discharges**	**Typical production figures**	**Typical discharges of relevant radionuclides (GBq/a)**
Cement production	Dust from clinker kilns	0.7 Mt/a	Pb-210: 0.014 Po-210: 0.28
Coal-fired power plants (hard coal)	Dust from coal combustion	1 GW_{el}	Pb-210: 0.67 Po-210: 1.35
Primary iron production	Sinter and smelter dust	5 Mt/a	Pb-210: 8.5 Po-210: 8.5
Lead smelting	Dust from smelting of ore concentrates	0.1 Mt/a	Pb-210: 0.3 Po-210: 0.3

4.3 Parameter variations for sensitivity analyses

It must be emphasised that the data collected in the study only reflect orders of magnitude of an industry as a whole covering typical ranges of operating facilities. In order to arrive at a realistic picture of the radiological impact of discharges from a NORM industry *sector* on public exposure, a careful sensitivity analysis is required. While Table 1 contains a "base case" scenario for each industry sector with regard to production figures that reflect the size of a *typical* plant in Germany, parameters for conservative scenarios were developed for the purposes of a sensitivity analysis, as shown in Table 2. Over and above the variations of the parameters shown in Table 2, the atmospheric dispersion conditions are subject to uncertainties that must be taken into consideration in a sensitivity analysis, too. Variations of atmospheric dispersion parameters are considered in Section 5.4 below.

Table 2: Variations of emission parameters for industry sectors from Table 1

Industry sector	**Conservative production figures**	**Discharges of relevant radionuclides (GBq/a)**	**Variation factor to account for uncertainties of the specific activity of dust particles**	**Dust filter retention rate (%)[a]**
Cement production	2 Mt/a	Pb-210: 0.04 Po-210: 0.8	2	95-**99**
Coal-fired power plants (hard coal)	2 GW_{el}	Pb-210: 1.3 Po-210: 2.7		95-**98**
Primary iron production	10 Mt/a	Pb-210: 17 Po-210: 17		97-**99**
Lead smelting	0.15 Mt/a	Pb-210: 0.45 Po-210: 0.45		95-**99**

(a) Base case retention rates on which figures in Table 1 are based are printed bold face

5 DETERMINATION OF GENERIC DISPERSION CONDITIONS FOR DUST

5.1 Topographical and meteorological parameters

As stated in the Introduction (Section 1), the objective of this study is *not* a site-specific calculation of public exposure due to discharges from a particular facility, but an investigation of which NORM industries would warrant regulatory control because discharges of a facility of "typical" size and technological parameters under "typical" meteorological and topographical conditions may lead to significant public exposure. Following this objective the challenge of this study was to develop a range of characteristic dispersion conditions that are typical for a sufficiently large number of NORM industry facilities operating in Germany. This task has therefore been tackled from various directions, as outlined in the following:

- *Geographical distribution of facilities:* The geographical distribution of facilities in the potentially relevant industrial sectors listed in Table 1 is analysed in order to determine whether there are regions in which these facilities are concentrated. If this is the case, the meteorological and topographical conditions of those regions should be considered "typical".
- *Meteorological parameters:* For geographical regions in which a large number of NORM facilities with discharges are concentrated, the statistical distributions of meteorological parameters such as wind speed and direction, turbulence (stability) class, and precipitation patterns must be obtained. However, even more important than the statistical data for each region is the question how sensitive the atmospheric dispersion depends on the meteorological conditions. In relation to this question it is important to remember that the source terms are known only up to an order of magnitude, so that the requirements on the precision of the dispersion modelling are limited. The German Meteorological Service (DWD) provides so-called TRY (Test Reference Year) model datasets that represent average weather conditions over the course of one year in one-hour time steps [9]. They were available for 15 characteristic regions[1]. From an evaluation of the geographical concentration of the locations

[1] It should be noted that from 2018 the datasets for 15 TRY regions that were used for the study, have been replaced by datasets for each square kilometre of Germany.

of NORM facilities the following can be concluded that 70% of all sites of potentially relevant NORM industries listed in Table 1 are located in only 4 TRY regions (Nos. 3, 5, 6, and 12). TRY regions no. 3, 5, and 6 are very similar with respect to key meteorological parameters such as wind speed and direction, and have been treated with one common meteorological dataset (that of TRY region no. 6). TRY region no. 12 is characterised by a bimodal angular distribution of wind directions. Therefore two distinct datasets were used to cover a wide range of meteorological conditions typical for NORM facilities in Germany. Between the various locations, precipitation varies typically between 500 and 1000 mm/a.

- *Topographical relief:* The overwhelming majority of sites of potentially relevant NORM industries listed in Table 1 are located in lowlands and rolling country with relatively flat hills. This fact significantly simplifies the atmospheric dispersion modelling.

For the numeric modelling of atmospheric transport of dust and radon the code ARTM (Atmospheric Radionuclide Transport Model [10]) was used. ARTM is based on Lagrange trajectories of unit air volume elements. It includes dry and wet deposition and takes into account radioactive decay along the trajectory.

5.2 Other relevant parameters, and parameter variations

To test the sensitivity of the deposition rates (and hence, the effective dose) with respect to variations of the atmospheric dispersion conditions, the following sensitivity analyses were carried out:

- The parameter that is ultimately relevant for the dose calculations under this project is the deposition rate of dust-borne radioactivity on agricultural areas. While the wet deposition rate depends on the precipitation rate and its temporal distribution (short, intense rainfall events vs. long periods of drizzle), the dry deposition rate depends on the atmospheric stability class and on the deposition velocity of the dust particles, which in turn depends on the particle size distribution.
- Dispersion modelling started with a complete initial parameter set for a reference site; the parameter sets for precipitation and stability class distribution were then varied to test the sensitivity of the model results with respect to these parameters.
- Different settling velocities were tested in the dust dispersion model.
- In order to test the sensitivity of the radionuclide deposition rate with respect to variations the atmospheric stability class, variable statistics were used, ranging from stable highly stable to unstable, and the resulting deposition rates compared.
- Atmospheric dispersion patterns also strongly depend on the height of the source. A survey carried out under this project has identified typical ranges of chimney heights in each of the industry sector of interest.
- Another important parameter for the dust deposition and dose model is the lateral distance between source and point of impact. Based on empirical evaluation (using Google Earth) of randomly selected sites of the first four industries listed in Table 1 it was found that typically the lateral distance between the point of emission and areas of agricultural use is at least four times the chimney height. As a, possibly oversimplified, working hypothesis it can be assumed that plants with large emissions (and higher chimneys) are part of industrial zones that are generally farther away from agricultural areas than smaller plants. The model base case, therefore, assumes that the point of impact is four times the chimney height, whereas in a conservative variation this distance is reduced to twice the chimney height.
- As was stated in Section 5.1, most facilities of the industry sectors in question are located in areas of a relatively flat topography or in mildly rolling hills. In order to test the model sensitivity with respect to special topographical conditions, dust dispersion calculations were also carried out using the digital surface model of a relatively deep and narrow valley.

5.3 Results of dust dispersion and deposition rate modelling, base case

For the base case parameters of atmospheric dispersion conditions the model results of the deposition rate for Pb-210 and Po-210 are summarised in Table 3. The deposition rates are normalised to a source strength of 1 GBq/a. Deposition rates that were obtained using the ARTM code for TRY regions 6 and

12 and for two sites with high (1000 mm/a) and low (559 mm/a) precipitation, respectively, were averaged and the result rounded up to the next full integer of the base. It is worth noting that wet deposition dominates total deposition in the near field of distances up to around 10 times the chimney height.

Table 3: Deposition rates of Pb-210 and Po-210, normalised to a source strength of 1 GBq/a, at a lateral distance from the source of four times the chimney height

Chimney height (m)	Average total (wet and dry) deposition rate (Bq/m²s), rounded up to full integer of base	Standard deviation of modelling results used for averaging (% of average)
50	1E-7	46%
100	3E-8	27%
200	1E-8	19%

5.4 Sensitivity analyses of the dust deposition rate modelling

Sensitivity analyses were carried out to cover a sufficiently broad range of dispersion conditions. Apart from the dispersion conditions, the source strength of discharges may vary depending on the size and technical parameters of the industrial facility, as shown in Table 2. A complete account of the model results obtained for a large number of scenarios would go beyond the limited space available in this paper but can be found in the original study [3]. An important conclusion is that combining only a few parameter variations from Table 2 and Section 5.2 may lead to deposition rates and effective doses that are higher by a factor of 10 and more compared to those of the base case.

6 ESTIMATE OF EFFECTIVE DOSES DUE TO THE DISCHARGE OF DUST

Using the base case scenario for both source strength of dust discharge and atmospheric dispersion parameters, and the relationship between deposition rate of Pb-210 and Po-210 and the effective dose incurred by the most sensitive age group (infants) established in Section 3, the following results have been obtained (doses exceeding the threshold of 100 µSv/a are printed bold face).

Table 4: Effective annual dose in infants (µSv/a) due to dust discharges from potentially relevant industry sectors

Industry sector, base case	Chimney height (m)		
	50	100	200
Cement production	3.3E+01	9.9E+00	3.3E+00
Coal-fired power plants (hard coal)	n.a.	5.2E+01	1.7E+01
Primary iron production	n.a.	**3.5E+02**	**1.2E+02**
Lead smelting	4.2E+01	1.2E+01	n.a.

In the base case, only primary iron production leads to effective doses in infants that exceed the threshold of relevance of 100 µSv/a. However, as was mentioned in Section 5.3, variations of a combination of only a few parameters within a sensitivity analysis may easily lead to significant increases of the activity deposition rate by a factor of 10 and more, and hence of the effective dose. Therefore, it is obvious that discharges of virtually all four industries listed in Table 4 may be radiologically relevant in the sense of the “relevance threshold” used in this study. It must be emphasised again that these considerations are based on a generic approach; the discharge from each site must be evaluated individually.

7 SUMMARY OF RESULTS

From the 16 industry sectors enumerated in Annex VI of the European Basic Safety Standards [1] and the 10 additional sectors that were investigated within the scope of this study, only a relatively small number has turned out to be potentially relevant from a radiation protection point of view. More specifically, only dust discharges from primary iron production are likely to exceed the threshold of

100 µSv/a in the most vulnerable age group (infants). Within an extensive sensitivity analysis it was found that under more conservative assumptions regarding the source strength of the discharges and atmospheric dispersion conditions, cement production, hard coal fired power plants, and lead smelting may also lead to effective doses of more than 100 µSv/a in infants, if several conservative parameter variations are combined.

One of the reasons for the relatively low effective doses resulting from discharges of dust is the industry's compliance to strict regulatory limits on air- and water-borne discharges that have been introduced independent of radio-ecological considerations in the first place.

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