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# **Review of NORM occurrence and application of a tailored graded approach** for the radiation protection in geothermal plants

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**Abstract** In the present paper, the general methodological approach developed to manage legislative requirements for NORM involving industries was fitted to the geothermal industrial sector, which is in the indicative list of the European Directive 2013/59/Euratom (EU-BSS). A review of the state of the art about the radiological characterization of NORM in geothermal plants have been performed with the aim to identify matrices and exposure scenarios of radiological concern. From the analysis of collected data, it results that radiological content of NORM residues generally depends on the characteristics of the geothermal fluid as well as on the type of the plants. In several plants, residues (both scales and filtering materials) show generally high activity concentrations, especially for Ra-226 and Ra-228 decay segments, exceeding Exemption Levels of the EU-BSS. Several tables have been presented as tools to support the stakeholders in the application of the legislative requirements regarding radiation protection in the geothermal sector.

#### **1** Introduction

Climate change and depletion of energy sources increasingly require the adoption of new strategies to limit this crisis. In recent years, the European Commission adopted a set of strategic proposals that aims to lead the EU towards a green transition making Europe "the first climate neutral continent in the world" by 2050 [1]. The most recent European Green Deal [2] contains proposals covering many areas, e.g. climate, energy, transport and taxation policies setting as the first objective the reduction of emissions by at least 55% by 2030 compared to 1990 levels.

In this framework, the economic and environmental benefits of geothermal energy are well recognized [3, 4], and a recent work has estimated that geothermal technology could contribute 4–7% to overall power generation in Europe [5]. Unlike wind or solar resources, geothermal energy is not influenced by meteorological fluctuations, and the higher initial cost for the installation and geographical dependence can be offset by using recent research and developments in geothermal technology [6].

However, geothermal energy production plants could have also an impact on environment [7] and on human health due to the presence of some chemical hazards, such as mercury, hydrogen sulphide [8] and arsenic [9]. Moreover, the presence of naturally occurring radioactive materials (NORM) can also be of concern for workers and population living nearby.

For this reason, in the European Union (EU) legislation, geothermal energy production was recently included amongst the industrial sectors involving NORM that can lead to an exposure for workers and/or members of the public, which cannot be disregarded from a radiation protection point of view [10].

Nevertheless, the impact of NORM in geothermal energy sector has received much less attention compared to that in oil and gas production. Indeed, even if the radiological impact of geothermal energy production has been well known for more than 20 years [11], very few scientific papers in peer-reviewed journals have been published on this topic so far. Most of the relevant publications are present in the grey literature (conference proceedings, activity reports of national institutions, etc.), which is generally difficult to access for stakeholders.

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Therefore, in this work, a literature review about the radiological characterization of NORM in geothermal plants have been performed also considering the grey literature with the aim to identify matrices and exposure scenarios of radiological concern. The results will be used to tailor to the geothermal industrial sector, the general graded methodological approach recently developed to characterize the exposure scenarios in NORM involving practices [12].

## 2 Background

#### 2.1 Characteristics of geothermal plants

Geothermal energy, according to the characteristic of the fluid, can be used directly to heat buildings, grow plants in greenhouses or indirectly to produce electricity. There are different approaches to generate electricity from geothermal sources. The simplest one is provided by *dry steam power plants* that use steam from geothermal fluid sent through a turbine which drives a generator that produces electricity. The steam coming from the turbines is mostly reinjected into the underground reservoir after its condensation through cooling towers. However, most reservoirs produce only hot water, or a combination of steam and hot water (two-phase fluid). In the latter case, if the reservoir temperature is above 250 °C, the vapour is generally the dominant phase. Instead, for lower temperatures, the water is the dominant phase [13].

In case of two-phases fluid at temperature higher than about 200 °C, the fluid is drawn into a container (called flash tank) held at a much lower pressure than the fluid, so that, the sudden decrease in pressure causes the liquid water to vaporize ("to flash") into steam that is then used to power the turbine-generator set. This is the operating principle of the *flash steam power plants* [11].

Below 200 °C instead, the fluid is mostly hot water: in this case, the plants are generally based on a *binary cycle* in which hot water is drawn up through a primary set of pipes from which the energy contained in the water is transferred by a heat exchanger to another fluid (working fluid) with a lower boiling point. The working fluid is contained within another closed loop of pipes and the steam produced powers the turbine-generator set.

A simplified scheme of above-mentioned geothermal plants is reported in Table 1.

It is worth mentioning that a classification of the geothermal fluid can be done using different approaches i.e. by temperature, use, type and status, accessibility, electric power generation, enthalpy (energy contained in water or steam, in kJ/kg).

## 2.2 NORM occurrence in geothermal plants

#### 2.2.1 Geothermal fluids

Besides water, the main constituents of geothermal fluids are the Total Dissolved Solids (TDS) and non-condensable gases (NCG), which can potentially affect the plant operations as well as be a source of NOR materials [14] (Fig. 1).

The Total Dissolved Solids (TDS) is a measure of the total ions in solution, which are typically in the form of salts, so TDS are generally a proxy of the salinity of the geothermal fluid [14]. The typical dissolved solids found in the fluid are Na<sup>+</sup>, K<sup>+</sup>, Ca<sup>2+</sup>, Mg<sup>2+</sup>, Ba<sup>2+</sup>, Sr<sup>2+</sup> SO<sub>4</sub><sup>2-</sup>, Cl<sup>-</sup>, HCO<sub>3</sub><sup>-</sup>, CO<sub>3</sub><sup>2-</sup> which amounts vary considerably, from about 100 g/tonne (for some fields in Iceland) to about 250,000 g/tonne (Salton Sea) [15].

Isotopes of radium (Ra-224, Ra-226 and Ra-228) and lead (Pb-210) are also present as TDS. Ra isotopes are produced by alphadecay, and recoil damage to the host mineral is generally regarded as the mechanism by which Ra is released [16]. Several authors [17, 18] investigated the effectiveness of alpha-decay product removal from different kind of rocks reporting that recoil ejection from grains and release by natural etching of alpha-recoil tracks are the two principal mechanisms by which alpha-decay products enter the pore water system. In case of presence of U and Th on the mineral grain surface, direct Ra ejection to pore water could occur. Because of the nature of the radionuclides and their solubility and/or mobility and their ability to precipitate as a mineral phase, several situations may occur. In particular, similarly to oil and gas sector [19], radionuclides can be present in:

- 1. scales;
- 2. sludge;
- 3. gaseous effluents (radon);
- 4. water discharges;
- 5. residues from secondary processes (e.g. sandblasting).

## 2.2.2 Scales

During exploitation, the geothermal fluid is submitted to temperature and pressure changes and brought to the surface where heat is extracted in two different ways:

- Heat is transferred to a second working fluid (a binary system) in a heat exchanger;
- Steam is extracted to become the working fluid.

**Table 1** Characteristics of thedifferent types of geothermalplants (adapted from [13, 20])

| Fluid type             | Reservoir temperature  | Production enthalpy | Typical plant process  |
|------------------------|--|---------------------|------------------------|
| Warm water             | T<125 °C   | <600 kJ/kg          | Direct heat use        |
| Hot water              | T<220 °C   | <943 kJ/kg          | Binary cycle           |
| Two-phase, liquid domi | nated:   |                     |                        |
| Low enthalpy           | 220 °C <t<250 td="" °c<=""><td>943–1100 kJ/kg</td><td>Flash steam</td></t<250>             | 943–1100 kJ/kg      | Flash steam            |
| Medium enthalpy        | 250 °C <t<300 td="" °c<=""><td>1100–1500 kJ/kg</td><td>Flash steam</td></t<300>            | 1100–1500 kJ/kg     | Flash steam            |
| High enthalpy          | 250 °C <t<330 td="" °c<=""><td>1500–2600 kJ/kg</td><td>Flash steam</td></t<330>            | 1500–2600 kJ/kg     | Flash steam            |
| Two-phase, vapour don  | ninated:   |                     |                        |
| High enthalpy          | 250 °C <t<350 td="" °c<=""><td>2600–2800 kJ/kg</td><td>Dry steam/Direct cycle</td></t<350> | 2600–2800 kJ/kg     | Dry steam/Direct cycle |

**Fig. 1** Flow-chart reporting how the TDS and NCG of the geothermal fluid affect the plant operations and can lead to NORM occurrence



In the first case, the geothermal fluid is conductively cooled and there is the chance that some of the dissolved species will deposit since the solubility of most compounds decreases with decreasing temperature. In the second case, the geothermal fluid, will be concentrated by removing some of the water as steam: in this case, there is also the possibility of depositing some mineral species. In both cases, as the fluid rises from the well, the physical and chemical properties can change and lead to deposition of scales.

Scales can be found on the internal surfaces of drilling and production equipment (e.g. steam turbines, heat exchangers, valves, fluid handling equipment, etc.). In particular, high concentration of Ra-226, Pb-210 and Po-210 present in the deep saline fluids can be found, respectively, in the precipitants containing  $BaSO_4/SrSO_4$  and PbS (galena)/PbOHCl (laurionite), leading to enhanced levels of radionuclides in pipe scales, heat exchange panels and filtering materials [21]. In particular, radium, due to the similarity with barium and strontium (all earth alkaline with large ionic radius) can precipitate as barite by  $Ra^{2+}$  isomorphic replacement of  $Ba^{2+}$  and  $Sr^{2+}$ . Radium-rich barite has frequently observed in natural systems such as groundwater [22], thermal water [23] or scales in oil wells [24].

Several studies have been addressed scales creation processes [25] and methods for reducing their formation such as sulphate scaling inhibitor (generally high charged polymeric compounds) often used successfully in oil and gas industry [26]. In fact, scales reduction would have an impact not only on the efficiency of fluid extraction, but also on the radiation protection measures needed for handling and disposing of such scales.

## 2.2.3 Sludge

In direct cycle plants, sludge is generally collected in the tank of the cooling tower from the counter flow cooling of the vapour phase and NCGs emission. Depending on the type of plant, the water content in sludge can be very different. When the solid phase is much lower than the liquid one, sludge is generally measured as they are without any drying treatment.

## 2.2.4 Gaseous effluents (radon)

Rn-222 is a non-condensable gas (NCG) present in the geothermal fluid. If the plant is not based on a binary cycle process (Fig. 1), radon is normally extracted from the condenser of the plants and discharged by the gas compressor in the atmosphere through the stacks or the cooling towers [27].

# 2.2.5 Water discharge

For some plants, during the maintenance work, part of the geothermal fluid is collected in a temporary storage basin. This water is generally discharged in sewer system after physical-chemical treatment [28].

# 2.2.6 Residues from secondary processes (sandblasting)

During maintenance, parts of the geothermal plants are disassembled and, when necessary, sandblasted. Exhausted sand can be regenerated and employed more than once. In the end, not reusable exhausted sand and dust from the ventilation system's abatement filter of the sandblasting cabin are to be considered contaminated by radionuclides originating from the scales deposits. Moreover, exhausted sand and dust can contain corundum powder that is naturally rich in radionuclides of the radioactive decay chains of U-238 and Th-232. For this reason it is advisable to measure the powders even before use to assess their NOR content.

## 3 Materials and methods

3.1 Literature review of documents containing radiological and dosimetric data on NORM in geothermal plants

A literature review of scientific paper in peer-reviewed journal containing radiological data related to NORM in geothermal power plants has been performed using main database such as *PubMed* and *Web of Science*. The search strategy was to find all the scientific articles published in English up to September 2023 having: i) in the title or in the abstract, the words "geothermal" and "NORM" (or "radioactive" or "radionuclide"). For all the words, the asterisk character (\*) was used as wildcard to find both the singular and plural forms of the words. For example, the query used for Web of Science was the following:

 $((TI = (geotherm^*)) AND (TI = (NORM) OR TI = ((radio^*)))) OR ((AB = (geotherm^*)) AND (AB = (NORM) OR AB = ((radio^*)))).$ 

A total of 95 records and 1042 records have been found using PubMed and Web of Science, respectively. Of these records, only 14 articles passed the screening phase after the reading of the titles and the abstracts, and for 13 of them it was possible to find full-text articles (Fig. 2). The chosen eligibility criteria were to include paper containing data on NORM activity concentration in matrices of the geothermal plants as well as information regarding doses to workers and population. With these criteria only six peer-reviewed articles were found. In fact, most of the screened papers were related to geothermal spas and not on geothermal plants. Therefore, the search was extended the grey literature (conference proceedings and presentations, reports and other relevant documents). For this type of search, all the IAEA conference proceedings on NORM were screened as well as all the relevant documents available on the website of the European NORM Associations (ENA, https://ena-norm.eu/).

In the grey literature, eight documents were found: four reports (three of them not in English language); two conference proceedings and two presentations in the last *NORM X* conference organized by IAEA. Overall, a total of 14 eligible articles and documents were found.

# 4 Results of the literature review

Despite the presence of geothermal plants all over the world (Fig. 3) the literature review finds out an exiguous number of scientific papers dealing with radiological issues in this field. Most of the papers are related to European and US (Imperial Valley) plants. In general, collected data refer to nine countries: for some of them (i.e. Germany, Italy) measurements were performed in different plants. However, only for some countries information is available for different kind of matrices: in some cases (i.e. Philippines, US) only information regarding one kind of solid residues is reported and in other cases (i.e. Turkey) the description of the measured matrix is not detailed.

**Fig. 2** Flow-chart of the literature review on radiological data in geothermal plants



In the following sections all the results regarding data collected from the literature are reported.

4.1 Radiological characterization of solid matrices

In Table 2, the activity concentration of natural radionuclides in geothermal residues collected by the literature review are reported. The activity concentration of natural radionuclides is spread in a very wide range amongst the different plants: this depends not only on the different characteristic of the plants but also on the geological and mineralogical properties of the reservoir basin.

Data show that radionuclides exceeding EU-BSS EL/CLs (Exemption and Clearance Levels) in scales and filtering materials are Ra-226, Pb-210 and Po-210 for U-238 decay series, and Ra-228 for Th-232 series. Instead, the radioactivity content of the parent nuclides of the decay series (U-238 and Th-232) as well as K-40 are generally present in negligible amounts, often below the analytic detection limits.

The Hot-Water System (HWS) plants have generally high contents of NORM in scales and filter deposits: the mean values of the radionuclides of Ra-226 and Ra-228 decay segments generally exceed the Exemption Levels of the EU-BSS (values in bold in Table 2). Instead, for the Vapour-Dominated System (VDS) plants (in Italy) this kind of NORM residues show a quite low radiological content. In fact, as shown in Fig. 4, the presence of scales deposits is poor in VDS plants because the scaling and the filtering processes involve only the NCG part of the geothermal fluid that contains a lower concentration of ions respect to the whole geothermal fluid.

It is worth to be noted that, in Germany, the HWS plants located in the region of *Molasse Basin* have generally activity concentrations about two orders of magnitude lower than the plants present in the other two areas (Upper Rhine Graben and North German Basin). These differences are explained by the different NORM radionuclide concentrations of the respective geothermal fluids (see Table 3) [29].

Moreover, in most of the HWS plants, the content of Pb-210 is higher than Ra-226: in some cases (e.g. Netherlands plants) the activity concentration of Pb-210 is more than two order to magnitude higher than Ra-226 [30]. In these cases, (Belgium, Netherlands and Upper Rhine Graben—Germany) the high values find for Pb-210 can be explained by the presence of sulphate inhibitors. This kind of inhibitors are quite effective in the reduction of (Ba, Sr, (Ra)) SO<sub>4</sub> suppressing the precipitation of sulphate minerals: in these conditions the precipitation of sulphides, metals, predominantly PbS, Pb is observed [26].

As general comment, looking at the same plant, high values of natural radionuclides are found both in scales and in filtering materials. An anomalous situation appears for the Italian plant in the Po valley. According to the authors [31], for operational



Fig. 3 Distribution of Geothermal Power Plant all over the world (taken from ThinkGeoEnergy [32]

difficulties, the scales sampling was done only near the heat exchanger and not where the geothermal fluid enters. This could explain the low values found in scales. Notably, in situ gamma dose rate measurements performed near the geothermal fluid enters showed values around 1500 nSv/h.

Regarding filtering materials data from the US plant, it is important to point out that these values represent the mean of filtering cakes positioned in a sequence in different sites of the plant. Generally, when more than one filter is used on a pipeline, scale deposit taken from the last filter resulted to have (one order of magnitude) lower natural activity concentration than the other ones [33].

Looking at Table 2, it can be observed that radiological contents of NOR in exhausted sand and dust is of the same order of magnitude of the sand itself (Ra-226 and Ra-228 = 0.3 Bq/g). In particular, the NOR content in samples of corundum power increases with the decreasing of particles size (from 20 to 80 mesh). This phenomenon could be attributable to the distribution of radionuclides in the grain of mineral (sand) and the crystalline structure.

#### 4.2 Radiological characterization of non-solid matrices

Regarding geothermal fluid, it is worth noting that although uranium and thorium are present in the lithological formations from which geothermal fluid are extracted, it does not contain significant concentrations of them due to the reducing conditions in the geothermal reservoir, which limit their mobility [34, 35]. Instead, in these conditions, radium is generally present in ionic form ( $Ra^{2+}$ ) in the geothermal fluid and then it is available for transport processes [36]. This behaviour is confirmed by data of the maximum activity concentrations found in geothermal fluids from the literature (reported in Table 3).

Notably, the maximum values for  $^{226}$ Ra (about 100 Bq L<sup>-1</sup>) found for brines and production waters from the oil and gas plants have the same order of magnitude of the geothermal fluids [11].

Regarding radon, measurements in the geothermal fluid were performed in VDS and HWS located in different plants (see Table 4), as reported by UNSCEAR [37]. In Table 4, average values are reported, but it is worth to note that the variability of the radon concentration amongst the different wells of the same plant can be very high. For example, for the New Zealand plant, the values vary from 16 to about 6600 Bq  $L^{-1}$ .

For VDS, radon concentration measurements were performed in the condensed steam after the sampling [38]. It is worth noting that in steam fields, the radon concentration is inversely proportional to the corresponding volume of steam at the wellhead, to

Table 2 Radiological content of NOR in solid matrices in Bq/g from some geothermal power plants in the world. Bold values indicate the Exemption Levels of the EU-BSS [10]

| Matrix                  | Type of plant | Country     | Country Location      |        | Ra-226 Pt |  | Pb-210 |   | Po-210 |   | Ra-228 |        | Th-228 | Ref                 |
|-------------------------|---------------|-------------|-----------------------|--------|-----------|--|--------|---|--------|---|--------|--------|--------|---------------------|
|                         |               |             |                       | AM     | Max       | AM   | Max    | AM  | Max    | AM  | Max    | AM     | Max    |                     |
| Solid residue           | LE-LDS        | Turkey      | Denizli               | 0.5    | 2.7       |  |        |   |        | 0.4   | 2.4    |        |        | [39]                |
| Scales                  | ME-LDS        | Iceland     | Reykjanes             | < 0.01 |           | 43   | 52     | 169   | 214    |   |        |        |        | [ <mark>40</mark> ] |
|                         | HE-LDS        | Philippines | Bac-Man               | < 0.01 |           |  |        |   |        | <dl< td=""><td></td><td></td><td></td><td>[41]</td></dl<> |        |        |        | [41]                |
|                         | VDS           | Italy       | Larderello            | < 0.01 | 0.04      | 0.06   | 0.4    |   |        | < 0.01  | 0.02   |        |        | [42]                |
|                         | EGS           | France      | Soultz                | 1015   |           |  |        |   |        | 338   |        | 220    |        | [21]                |
|                         | HWS           | Belgium     | Balmatt               | 4.8    |           | 170  |        | 9160  |        | 1.2   |        |        |        | [25]                |
|                         | HWS           | Germany     | Upper Rhine<br>Graben | 34     | 770       | 500  | 5500   |   |        | 18  | 430    | 6.1    | 130    | [ <mark>29</mark> ] |
|                         | HWS           | Germany     | North German<br>Basin | 38     | 430       | 170  | 770    |   |        | 41  | 300    | 16     | 240    | [ <mark>29</mark> ] |
|                         | HWS           | Germany     | Molasse Basin         | 0.04   | 0.4       | 0.03   | 0.07   |   |        | 0.01  | 0.03   | < 0.01 | 0.02   | [ <mark>29</mark> ] |
|                         | HWS           | Italy       | Po valley             | 0.6    |           | <dl< td=""><td></td><td></td><td></td><td><dl< td=""><td></td><td></td><td></td><td>[31]</td></dl<></td></dl<> |        |   |        | <dl< td=""><td></td><td></td><td></td><td>[31]</td></dl<> |        |        |        | [31]                |
|                         | HWS           | Netherlands |                       | 4      |           |  | 1600   |   |        | 1.9   |        | 1.6    |        | [ <mark>30</mark> ] |
| Filter deposits         | HE-LDS        | USA         | Imperial Valley       | 4.9    | 9.4       | 3.6  |        | 3.6   |        | 3.4   | 7      | 0.9    |        | [33]                |
|                         | VDS           | Italy       | Larderello            | 0.02   | 0.03      | 0.04   | 0.1    | -   |        | 0.02  | 0.04   |        |        | [42]                |
|                         | HWS           | Belgium     | Balmatt               | 22     | 27        |  | 250    | <dl< td=""><td></td><td>2.5</td><td>2.7</td><td></td><td></td><td>[25]</td></dl<> |        | 2.5   | 2.7    |        |        | [25]                |
|                         | HWS           | Germany     | North German<br>Basin |        | 102       |  | 24     |   |        |   | 75     |        | 30     | [34]                |
|                         | HWS           | Italy       | Po valley             | 7.2    |           | 19   |        |   |        | 0.09  |        |        |        | [31]                |
|                         | HWS           | Netherlands |                       | 0.8    |           | 50   | 1000   |   |        | 0.06  |        |        |        | [30]                |
|                         | EGS           | France      | Soultz                | 1033   |           |  |        |   |        | 280   |        | 269    |        | [21]                |
| Sludge-cooling<br>tower | VDS           | Italy       | Larderello            | 0.01   | 0.03      | 0.45   | 1.9    | 0.5   |        | < 0.01  | 0.02   |        |        | [42]                |
| Sandblasting<br>dust    | VDS           | Italy       | Larderello            | 0.2    | 0.3       | -  |        | 0.23  |        | 0.2   | 0.25   |        |        | [42]                |

LE-LDS = Low enthalpy, liquid-dominated systems; HWS = Hot-water systems; ME-LDS = Medium enthalpy, liquid-dominated systems; EGS = Enhanced Systems; HWS = Hot-water systems; ME-LDS = Medium enthalpy, liquid-dominated systems; EGS = Enhanced Systems; ME-LDS = Medium enthalpy, liquid-dominated systems; EGS = Enhanced Systems; ME-LDS = Medium enthalpy, liquid-dominated systems; EGS = Enhanced Systems; ME-LDS = Medium enthalpy, liquid-dominated systems; EGS = Enhanced Systems; ME-LDS = Medium enthalpy, liquid-dominated systems; EGS = Enhanced Systems; ME-LDS = Medium enthalpy, liquid-dominated systems; EGS = Enhanced Systems; ME-LDS = Medium enthalpy, liquid-dominated systems; EGS = Enhanced Systems; ME-LDS = Medium enthalpy, liquid-dominated systems; EGS = Enhanced Systems; ME-LDS = Medium enthalpy, liquid-dominated systems; EGS = Enhanced Systems; ME-LDS = Medium enthalpy, liquid-dominated systems; EGS = Enhanced Systems; ME-LDS = Medium enthalpy, liquid-dominated systems; EGS = Enhanced Systems; ME-LDS = Medium enthalpy, liquid-dominated systems; EGS = Enhanced Systems; ME-LDS = Medium enthalpy, liquid-dominated systems; EGS = Enhanced Systems; ME-LDS = Medium enthalpy, liquid-dominated systems; EGS = Enhanced Systems; ME-LDS = Medium enthalpy, liquid-dominated systems; EGS = Enhanced Systems; ME-LDS = Medium enthalpy, liquid-dominated systems; EGS = Enhanced Systems; ME-LDS = Medium enthalpy, liquid-dominated systems; EGS = Enhanced Systems; ME-LDS = Medium enthalpy, liquid-dominated systems; EGS = Enhanced Systems; ME-LDS = Medium enthalpy, liquid-dominated systems; EGS = Enhanced Systems; ME-LDS = Medium enthalpy, liquid-dominated systems; ME-LDS =geothermal system; HE-LDS = Liquid dominated, high-enthalpy systems; VDS = Vapour-dominated systems; DL = Detection level

| <b>Table 3</b> Radiological content ofNOR in non-solid matrices(geothermal fluid) fromgeothermal power plants:maximum values in Bq $L^{-1}$ | Country | Location           | Ra-226 | Pb-210   | Ra-228                         | Reference |
|---|---------|--------------------|--------|--|--------------------------------|-----------|
|   | Germany | Upper Rhine Graben | 48     | 26   | 31                             | [28, 43]  |
|   | Germany | North German Basin | 35     | 160  | 35                             | [29]      |
|   | Germany | Molasse Basin      | 1.5    | < 0.1  | 0.5                            | [29]      |
|   | Italy   | Po valley          | 42     | <dl< td=""><td><dl< td=""><td>[31]</td></dl<></td></dl<> | <dl< td=""><td>[31]</td></dl<> | [31]      |
|   | Turkey  | Denizli            | 9      |  |                                | [39]      |
| DI – Detection level  | USA     | Salton Sea         | 55     | 97   | 51                             | [44]      |
|   |         |                    |        |  |                                |           |

Table 4 Average radon activity concentrations in geothermal fluids in different types of plants

| Fluid Type    | Country     | Location        | Rn-222 (Bq L <sup>-1</sup> ) | Reference |
|---------------|-------------|-----------------|------------------------------|-----------|
| VDS-Dry steam | Italy       | Larderello      | 1280                         | [38]      |
| VDS-Dry steam | USA         | The Geysers     | 620                          | [38]      |
| VDS-Dry steam | USA         | Salton Sea      | 110                          | [38]      |
| Hot water     | USA         | Imperial Valley | 1.1                          | [38]      |
| LE-LDS        | New Zealand | Wairakei        | 630                          | [45]      |

**Fig. 4** Scales deposit in parts of a VDS plant in Tuscany (Italy)





temperature and pressure. For these reasons, the high content of Pb-210 and Po-210 in sludge and scales can be due to the elevated presence of radon gas in the geothermal fluid.

## 4.3 Estimated dose for workers and population

For workers, the operations that generally produce an increase in the effective dose, generally due to the external irradiation, occur during the maintenance of the plants. Near pipelines and heat exchangers dose rate of up to 12  $\mu$ Sv/h was measured at 1 m from plant components in some German plants [29]. Instead, the dose rate measured in the Italian plants were in the range 0.1–3  $\mu$ Sv/h [31] for HWS and below 1  $\mu$ Sv/h for VDS [42].

In addition to external radiation, the contribution of dust ingestion or inhalation has to be considered if inspection and maintenance were carried out on open parts of the plants, for example, in the operations for removing scales. However, exposure to radon for workers is not expected to be significant since scales resulted to have radon emanation fraction typically around 5%, according to measurements performed in scales from oil and gas industry (very similar to those from geothermal power plant). This is probably due to the hard, solid structure of the scale that inhibits the release of radon [46]. Moreover, during the removal process the scales area are commonly wetted [47] and workers are equipped with occupational safety measures, generally used for other hazardous substances, such as wearing a mask or gloves that highly reduce the risk of ingestion and inhalation [29].

For the population, radon discharge is surely a scenario to be considered for dose assessments. Notably, the dispersion of Rn-222 and its progeny from premise and abroad had been modelled in Germany by different authors leading to the conclusion that Rn-222 will not contribute to an enhanced dose level around the plant [28, 36]. This is also confirmed by measurements performed at Wairakei power plant (New Zealand), from [45], which found that Rn-222 was diluted in atmosphere to low concentrations, presenting no health hazards. Regarding radon progeny, measurements of Pb-210 deposition rate were performed on Epiphytic lichens (used as bio accumulator of radionuclides) both at Wairakei power station (New Zealand) and in the Travale geothermal area (Italy), showing that radioactivity in geothermal fields is similar to areas not subject to geothermal exploitation [8].

#### 5 A tailored graded methodology for geothermal plants

Starting from the general methodology for applying the graded approach in NORM involving industries, as described in a previous paper [12], from the analysis of data collected and from some measurements performed in field, a tailored graded methodology for geothermal plants was developed.

The general methodology consists of two phases, each one divided into four consecutives steps (Fig. 5). The aim is to identify the most critical exposure scenarios and verify the compliance of exposure levels for workers and members of the public to the relevant ELs.

## 5.1 Identification of solid matrices

Starting from the data reported in Table 2 and from the literature review, a list of solid matrices and of the most critical radionuclides that need to be measured are identified and reported in Table 5.

According to RP 122-II approach [48], in Table 5 some radionuclides are followed by a "+" (i.e. Ra-226 +), this means that for that specific chain segment shorter-lived daughter nuclides are in secular equilibrium with parent.

With the exception of Po-210 that requires *alpha* spectrometer to be detected, all the other radionuclides listed in Table 5 can be detected by laboratory *gamma* spectrometry.

It is important to point out that parts of the plant are generally measured by in situ gamma spectrometry since is not possible to remove them from the plant unless their replacement during maintenance operation. Moreover, in this case Pb-210 were not considered as critical radionuclide since its (low) gamma energy emission was shielded.



Fig. 5 Methodology proposed to apply the graded approach for NORM involving industries (adapted from [12])

| Table 5 Solid matrices (residues)         of interest to be addressed in the    | Materials to be sampled                    | Description of the samples                               | Most critical radionuclides             |  |  |
|---|--|--|---|--|--|
| step 2 of the graded approach   | Sludge—dry and wet <sup>*</sup> residues   | Sludge of the collection tank of the cooling tower       | Ra-226 + , Ra 228 +                     |  |  |
|   | Parts of the plants                        | Valves, tubes, vessels,                                  | Ra-226 + , Ra 228 +                     |  |  |
|   | Scales deposits                            | Scales from tubes, vessels and other parts of the plants | Ra-226 + , Ra 228 + , Pb-210,<br>Po-210 |  |  |
|   | Exhausted adsorbent and filtering material | Filter cakes, sorbents and catalyst                      | Ra-226 + , Ra 228 + , Pb-210,<br>Po-210 |  |  |
| *Sludge with high water content<br>or supernatant liquid phase of the<br>sludge | Other exhausted materials                  | Corundum powder, other sands used for sandblasting       | Ra-226 + , Ra 228 + , Pb-210            |  |  |

High values of Pb-210 and Po-210 both in scales and filtering materials (see Table 2) were generally found in *Hot-water systems* plants (such as Reykjanes, Balmatt). This evidence suggests the importance to measure also these radionuclides despite the fact that their measurements may present some operational difficulties due to the low energy emission for Pb-210 and to the need of radiochemical preparation of the sample for Po-210 measurement by alpha spectrometry. However, if residues are stored for several months, Po-210 will be in equilibrium with Pb-210 and the measurement of Po-210 could be avoided.

Regarding filtering materials, when more than one filter is present on the pipeline, is advisable to measure the one in proximity of the geothermal fluid inlet.

## 5.2 Identification of exposure scenarios for workers and population

If concentration in solid residues exceeds ELs, the procedure requires dose assessment for workers and population. The most relevant exposure scenarios are reported in Table 6. This table reports the description of the specific exposure scenarios for workers and members of the public, generally taken from RP 122-II [48] and RP 107 [47], with the exception of the radon release in atmosphere. Indeed, as pointed out in Sect. 4.3, radon discharge is surely a scenario to be considered for dose assessments for members of the public, especially for plants with very high radon concentrations releases [49].

In Table 6 are also mentioned matrices significant for the dose evaluation not radiologically characterized in Phase 1, in particular non-solid matrices.

Scenarios regarding the reuse and (mostly) the landfilling of residues have also to be take into account for the dose assessment for members of the public. Generally, residues from the geothermal plants (e.g. scales or filtering materials) have the same disposal route of those coming from the *oil and gas* energy production industries. As a consequence, whereas Ra-226 and its progeny are present, also the doses resulting from disposal could be quite similar to those estimated for the oil and gas industries [28].

5.3 Radiological characterization of other matrixes of interest and measurements useful for the dose evaluation

From the analysis of the geothermal energy production processes, non-solid matrices of possible concern have been identified and a list is reported in Table 7. Reinjection waters are not considered as liquid discharge since they are reinjected into power plant circuit.

The inclusion of other matrices is finalized to the dose assessment for workers and member of public. In order to assess the exposure of workers, in situ gamma dose rate are generally performed. The natural radioactivity content of the geothermal fluid

 
 Table 6 Scenarios identified for estimating doses to workers and population

| Specific exposure scenarios                                       | Exposure scenarios defined in RP docs                        | Type of matrices         | Material   | Exposure pathways |
|---|--|--------------------------|--|-------------------|
| Worker—exposure<br>from removing<br>residues                      | Removal of scales RP<br>107 [47]                             | Residues                 | Scales<br>Sludge   | Ex, In, Ig, Rn    |
| Worker—exposure<br>from process vessels                           | RP 107 [47]  | Raw material<br>Residues | Geothermal fluid<br>Scales                                 | Ex                |
| and pipes<br>Worker—exposure to<br>stockpiles of<br>materials     | Storage of moderate or<br>large quantities—RP<br>122 II [48] | Residues                 | Scales<br>Sludge<br>Filtering materials                    | Ex, In, Ig, Rn    |
| Worker—transport of<br>materials (including<br>loading/unloading) | Transport RP 122-II<br>[48]                                  | Residues                 | Scales<br>Sludge<br>Corundum powder<br>Filtering materials | Ex, In, Ig        |
| Population—radon<br>discharge                                     | not defined  | Effluents                | -  | Rn                |
| Population—reuse or<br>landfilling of residues                    | Landfill RP 122-II [48]                                      | Residues                 | Scales<br>Sludge<br>Filtering materials                    | Ex, In, Ig        |

Eur. Phys. J. Plus

(2024) 139:195

Ex = external exposure; In = inhalation; Ig = ingestion; Rn = radon

**Table 7** Other matrices(liquid/gaseous) of interest to beaddressed in the *step 6* of thegraded approach

| Type of matrix       | Matrix                  | Description                     | Radiological content or other physical quantities |
|----------------------|-------------------------|---------------------------------|---|
| Liquid               | Geothermal fluid        | Fluid at the entry of the plant | Ra-226 + , Ra 228 + , Pb-210<br>Gamma dose rate   |
| Gaseous              | Vapour containing radon | Release from the cooling tower  | Rn-222  |
| Other solid matrices | Parts of the plant      | Tube, vessel,                   | Gamma dose rate                                   |

could be measured by in situ gamma spectrometry or, when sampled at well, measured in laboratory in order to have an indication about the Rn-222 release and the consequent dose to members of public.

# 6 Application of the graded methodological approach to the geothermal energy production in Italy

In the following sections, after a brief description of the geothermal energy production in Italy, the application of the tailored graded approach (described in the previous paragraph) to Italian geothermal sector is reported.

## 6.1 Geothermal energy production in Italy

According to updated Italian NORM inventory [12], in Italy, there are 34 geothermal power plants with 37 production units. Almost all the 34 geothermal power plants are based on direct cycle technology and the geothermal fluid can be a superheated steam, or saturated steam or a two-phase fluid with a NCG (non-condensable gas) content of about 2–10% by weight.

In particular, Italy is characterised by four areas of underground heat. Due to the characteristics of the geothermal fluid, only in one area (Tuscany) geothermal high-enthalpy power plants with vapour-dominated systems (VDS) are present. In particular, *Larderello* today hosts the largest geothermal power plant in Europe, as well as the oldest in the world [27].

Moreover, in Italy there is also a hot-water system (HWS) plant located in the *Po valley* in which fluids are exploited through binary cycle. In this plant, used only for district heating and not for the production of electric energy, the geothermal fluid yields heat to a low-boiling fluid in the heat exchanger.

In the following section, graded approach was applied using data from both vapour-dominated plants in *Larderello* and hot-water system plant (HWS) located in the *Po valley*.

| Table 8 Maximum values for the           critical radionuclides resulted          | Radionuclide  | HWS                     |                                    | VDS                      | Italian EL                   |         |
|---|---------------|-------------------------|------------------------------------|--------------------------|------------------------------|---------|
| from the radiological<br>characterization of residues in                          |               | Max value $(Bq g^{-1})$ | Type of sample                     | Max value (Bq $g^{-1}$ ) | Type of sample               | (Bqg ') |
| plants  | Ra-226        | 7                       | Exhausted<br>filtering<br>material | 0.4                      | Corundum<br>powder           | 1       |
|   | Pb-210        | 19                      | Exhausted<br>filtering<br>material | 1.9                      | Sludge from<br>cooling tower | 5       |
|   | Po-210        | ND                      |                                    | 0.5                      | Sludge from<br>cooling tower | 5       |
|   | Ra-228        | < 0.09                  | Exhausted<br>filtering<br>material | 0.4                      | Corundum<br>powder           | 1       |
| In bold values above the EL   |               |                         |                                    |                          |                              |         |
| Table 9 Gamma dose rate   | Distance from | the filter (m)          | D                                  | ose rate (µSv/h)         |                              |         |
| measurements performed in<br>proximity of six filters of the<br>Italian HWS plant |               |                         | Ā                                  | М                        | Median                       | Max     |
|   | 0             |                         | 0.                                 | 5                        | 0.3                          | 1.4     |
|   | 0.5           |                         | 0.1                                | 2                        | 0.2                          | 0.4     |
| AM: arithmetic mean   | 1.0           |                         | 0.1                                | 2                        | 0.2                          | 0.3     |

6.2 Application of the graded approach to some Italian geothermal plants

The EU-BSS in the Annex VI identified the entire industrial process of *geothermal energy production* as a practice. In Italy, the national legislation specifies as critical exposure situation the "Maintenance of high or medium-enthalpy geothermal energy systems" [50] so no further analysis of the industrial process is needed (*step 1*).

Regarding the *step 2*, both EU-BSS and the Italian legislation require the characterization of only solid matrices to verify the compliance with EL/CLs in terms of activity concentrations. Since the raw material used in geothermal energy production is the geothermal fluid, the only types of solid matrix to be characterized are the residues reported in Table 5 together with the most critical radionuclides.

Regarding the *step 3*, from the radiological characterizations performed in the solid matrices of the Italian plants resulted that for the VDS plants the maximum value of activity concentration (Table 8) does not exceed the general exemption levels (ELs) of the Italian legislation (*step 4*). Therefore, according to the graded approach, on the basis of the collected data, no further investigation regarding dose assessment for workers and population are required.

Instead, for the HWS plant, the maximum values of Ra-226 and Pb-210 activity concentration in an exhausted filtering material are higher than the Italian ELs. Therefore, for this plant, a dose evaluation is necessary to verify the compliance with the exemption levels in terms of annual effective doses (*Phase 2*), which are equal to 1 mSv/y and 0.3 mSv/y for workers and members of the public, respectively.

Starting from general exposure scenarios listed in Table 6, for the Italian HWS plant "exposure from removing residues" and "exposure from process vessels and pipes" are identified for workers, instead no exposure scenario is relevant for population since the plant is based on binary cycle, so it does not determine any liquid and atmospheric release (*step 5*). In this case, there are not any other matrices to be characterized. On the other side, gamma dose rate measurements at different distances from pipeline to evaluate the dose for workers were performed in the frame of the *step 6* activities. In Table 9 the results of these measurements are given. During the operation of replacing exhausted filters, maintenance workers are exposed to scales without the shielding of the tubes (thickness = 2 cm of steel). Since it was impossible to carry out gamma dose rate measurements during this phase of maintenance, it was considered a conservative evaluation of the workers doses doubling the values reported in Table 9.

For the workers who perform visual inspection of the plants recording the operating parameters, a personal dosimeter was used providing a maximum dose rate equal to 0.01  $\mu$ Sv/h [31]. Details of the workers exposure scenarios and relevant doses are reported in Table 10 (*step 7*). It is worth to note that, for both the ordinary and maintenance interventions, the annual doses evaluated are well below the Italian EL of 1 mSv/y (*step 8*).

As previously said, since this plant is based on binary cycle, it does not provide any atmospheric release from non-condensable gases (NCG) of the geothermal fluid. Therefore, regarding the population, the only scenario of concern is the landfilling of the exhausted filters. By comparison of the maximum activity concentration of Ra-226 and Pb-210 (see in Table 8) in residues with the

| Table 10 Annual dose assessed           for workers performing ordinary | Specific exposure scenario  | Annual working time (h/y) | Annual dose (mSv/y) |
|---|---|---------------------------|---------------------|
| and maintenance interventions in<br>an Italian HWS plant [31]           | Ordinary intervention—Visual inspection of the plants and recording of operating parameters | 500                       | 0.005               |
|   | Maintenance interventions—Replacement of exhausted filter                                   |                           |                     |
|   | Setting up and dismantling the scaffolding  | 18                        | 0.007               |
|   | Unbolting and subsequent tightening of the upper cap of the filter units                    | 12                        | 0.017               |
|   | Replacing of filters and operations with open equipment                                     | 24                        | 0.070               |
|   | Handling of the container containing exhausted filters                                      | 2                         | 0.001               |

specific EL/CLs for the landfill scenario reported in RP 122-II [48] it emerges that this kind of residues cannot be disposed of in landfills for conventional wastes.

### 7 Conclusions and perspectives

In the present paper, the general methodological approach developed to manage legislative requirements for NORM involving industries was fitted to the geothermal industrial sector, which is in the indicative list of NORM involving industrial sectors in the EU-BSS.

A literature review about the radiological characterization of NORM in geothermal plants has been performed to identify the most critical matrices and exposure scenarios from the radiological point of view. From the analysis of collected data, it emerges that radiological content of NORM residues is generally spread in a very wide range depending on the physical-chemical properties of geothermal fluid as well as from the different characteristic of the plants. Moreover, it emerges that VDS plants seem to be not highly radiologically impacting considering the scarce amount of scales and other solid residues because the scaling and the filtering processes involve only the NCG part of the geothermal fluid that contains a lower concentration of ions respect to the whole geothermal fluid. Otherwise, residues (both scales and filtering materials) from HWS plants show generally high activity concentrations, especially for Ra-226 and Ra-228 decay segments, exceeding Exemption Levels of the EU-BSS.

On the basis of review results, several tables are presented as tools to support the application of the tailored graded approach to the geothermal industrial sector. In particular, the procedures sketched in these tables helps to identify NOR matrices of interest, relevant radionuclides, suitable analytical methods and scenarios for dose assessment for workers and members of the public.

In conclusion, the analysis of the review data and the results of application of the methodology to Italian plants shows that the most critical exposure scenario for workers is the maintenance of the plant, and the main pathway is the external radiation. In fact, the occupational safety measures with which workers are equipped reduce the risk of ingestion and inhalation of radionuclides.

In perspective, further investigations needed to evaluate radon contribution to doses for both workers and members of the public are planned as well as the modelling and analysis of exposure scenarios regarding the reuse and landfilling of residues. Finally, data gathered by the on-going surveys on NORM involving industries promoted by RadoNorm project [51, 52] will contribute to a deeper knowledge of radiological aspects in this industrial sector.

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Data Availability Statement This manuscript has associated data in a data repository. [Authors' comment: Data sets generated during the current study are available from the corresponding author on reasonable request. Restriction can be applied by the owner of the geothermal plants.]

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