Radiological Impact of TENORM on the Environment in Romania

E. Botezatu, O. Iacob
Radiation Hygiene Laboratory, Institute of Public Health, 14, Victor Babes Str, Ro-700465 Iasi, Romania
E-mail: elena_botezatu@yahoo.com

Abstract. The objective of this study was to assess the radiological impact on the environment of some non-nuclear industries dealing with naturally occurring radioactive materials. The data of many years of environmental monitoring of radioactivity in the surrounding of ten coal-fired power plants (CFPPs), a phosphate fertilizer plant (PFP) and four oil fields are presented and discussed in this paper. The normal and enhanced levels of environmental natural radioactivity associated with those three non-nuclear industries are reported comparatively. The concentrations of uranium, thorium and potassium in the upper 5cm layer of soil were generally higher in the surroundings of the investigated plants and wells but were comparable to the representative values for the Romania. The natural radioactivity levels for thorium-232 and potassium-40 detected in surface water are generally comparable to those found in the other zones of Romania. The values for uranium-238 and radium-226 are significantly higher than the corresponding concentrations found generally in Romanian surface waters. The vegetation samples showed somewhat higher local activity values for all four radionuclides, with the highest uranium-238 and radium-226 content related to PFP working, being much higher than that determined in other areas. The study provides a scientific basis for decisions on the control of natural radionuclides from non-nuclear industries and on the disposal of their radioactive wastes (TENORM), even if natural radionuclides occur, into the environment. The radiological consequences of releases of radionuclides following three selected non-nuclear industrial activities are discussed related to further environmental risks.

1. Introduction

The mining, milling and industrial use of naturally occurring radioactive materials (NORM) covers a range of mineral resources and industrial activities. Due to the technological processes evolved in some of these industrial processes, the concentrations of natural radioactive elements in the products and in the wastes can be much higher than in the ore/raw materials. Technologically Enhanced Naturally Occurring Radioactive Material (TENORM) stands for technologically enhanced naturally occurring radioactive materials. "Technologically enhanced" was added to distinguish clearly between radionuclides as they occur naturally and radionuclides that human activity has concentrated or exposed. Elevated levels of NORM and consequently increased exposure to NORM have been identified in several major industries including energy production using fossil fuels, phosphate industry, and oil and gas production [1–4]. The wastes originated in these activities are released in the environment and hence, an environmental management of the highest quality is needed to reduce the resultant safety problems for both environment and population. So far, under Romanian legislation there were no radiological controls on the operation of these industries or restrictions on how waste is discharged (to the atmosphere, to landfill or being sold) which relate to its radionuclide content [5]. The growing concern amongst the population about the quality of their environment increases the significance of impact assessment of radioactive wastes disposal into the environment even if natural radionuclides occur. The objective of our study was to assess the radiological impact on the environment of these non-nuclear industries, which use and produce materials, containing technologically enhanced levels of naturally occurring radionuclides.

2. Materials and methods

Between 1989 and 2001 we studied ten coal-fired power plants (CFPPs), one phosphate fertilizer plant (PFP) and four oil fields from two major petroleum-producing region. The main radionuclides investigated in the uranium series in these industrial TENORM situations were uranium-238, polonium-210, radium-226 and radon-222 with its progeny. In the thorium series we looked at thorium-232 and radon-220 with its daughters. In addition, potassium-40 was determined. High-resolution gamma
spectrometry techniques and radiochemical and physical methods carried out in conformity with the current national standards and settlements were applied. Extensive measurements of the levels of natural radioactivity in raw materials (coal, sedimentary phosphate rocks), end products, a by-product (fertilizers, phosphogypsum), wastes (slag, ash, sale, sludge) and in environmental samples in the selected areas were performed. Soil, vegetation, snow, surface water, atmospheric deposition and air samples have been collected at 20 locations from the environment of each plant within a 5 km radius from the plant.

The soil (top 5 cm and 5-15 cm), vegetation and snow samples were collected from 400 sq. cm surface as in oil-field area, too. All the solid samples were dried at 105°C and after grinding were passed through 12-mesh sieve. The sieved samples were placed in the plastic Marinelli beaker and stored for 30 days to allow build up and reach radioactive equilibrium of radon and its daughters. After this period those samples were measured. Cumulated wet and dry atmospheric deposition were sampled monthly and analyzed seasonable weathers. These samples and vegetation samples were dried, calcined and wet mineralized. Arising acid extracts were utilized for the assay of natural radioelements like in water. Uranium-238 and thorium-232 were calculated after determining the content of natural uranium and thorium by the method based on their separation and purification on a strong basic anion exchange resin and spectrophotometric measurements in the form of their Arsenazo III complex. Radium-226 was determined through its decay descendent radon-222, and by alpha rays measurement in a scintillating chamber. The assay of polonium-210 was done by electrochemical deposition and alpha counting of polonium-210 deposited on the nickel disc in a low background ZnS (Ag) scintillation counter. Potassium-40 was found by calculation after the photometry dosing in flame emission mixture of potassium natural isotopes. Air was sampled every day at the same time for a week at 160 cm above the soil. The 222Rn and 220Rn short-lived decay product concentrations in air were measured, and the radon equilibrium equivalent concentrations (EEC) were calculated. The individual specific activity values for the attached fractions of 218Po, 214Pb, 214Bi and 212Pb were determined. The method for measuring radon decay products was an active one.

3. Results and discussion

3.1. Energy production using fossil fuels

Coal based thermal power plants constitute about 35% of quantum of energy supply in Romania. The combustion of coal results in partitioning of radionuclides included in the non-combustible mineral matter, between the bottom ash and fly ash. The coal-fired power plants are a potential radioactive polluting source for the environment due to the atmospheric discharges of escaping fly ash as well as by the large amounts of coal ash that requires proper management and disposal, either at the point of use or elsewhere in ash stored facilities. We were interested in knowing possible modifications of natural radiation background around the coal-fired power plants (CFPPs). With that end in view we selected ten representatives CFPPs, from which seven relatively modern and three older ones. Since 1993 the oldest plants and two relatively modern ones were modernized and equipped with electrofilters allowing ash retention of 99%. The coal-fired power plants are burning brown coal, lignite and/or mixture of different kinds of coal with an ash content of 26-55 %. Coal consumption is ranging from 5×10^9 up to 20×10^9Kg/Gwa. Large amounts of fly ash and bottom ash result from coal combustion, total ash production varying from 2×10^9Kg/Gwa up to 13×10^9Kg/Gwa. Roughly a third of coal ash is re-used mainly as substitutes or as an additive in cement and concrete, as structural fills for buildings and road construction, in land reclamation, in roofing materials and various products used as building materials [6, 7]. The wastes are hydraulic removed to the piles, which are placed close to surface water. 70 to 80% from the bulk quantity of the coal ash is disposed of in landfills or in ponds. There are many off-site coal-ash landfills and surface piles, at least one per plant.

The measurements performed on coal and wastes in the coal-fired power stations clearly demonstrated the enhancement with one order of magnitude of activity concentrations over the different processing stages from coal to ash (see Table I).
Table I. Activity mass concentrations of natural radionuclides in coal and wastes.

<table>
<thead>
<tr>
<th>Type of sample</th>
<th>Mean and range activity mass concentrations (Bq/Kg)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$^{238}$U</td>
</tr>
<tr>
<td>Coal</td>
<td>26</td>
</tr>
<tr>
<td></td>
<td>15 – 150</td>
</tr>
<tr>
<td>Slag and bottom ash</td>
<td>39</td>
</tr>
<tr>
<td></td>
<td>17 – 100</td>
</tr>
<tr>
<td>Fly-ash collected</td>
<td>144</td>
</tr>
<tr>
<td></td>
<td>45 – 276</td>
</tr>
<tr>
<td>Escaping fly-ash</td>
<td>226</td>
</tr>
<tr>
<td></td>
<td>75 – 390</td>
</tr>
</tbody>
</table>

The natural radionuclide concentrations in fly ash were significantly higher than the corresponding values for coal. These smaller particles are less efficiently collected by the filtering system and thus preferentially escape from the plant, falling out in the environment. The natural radioelements from the fly ash spread in the atmosphere as sedimentable powder and fall out on the soil surface either through falling, or in association with impurities or rain. The atmospheric discharges of natural radionuclides from CFPPs and consequently the surrounding natural radioactivity depend heavily on the efficiency of the emission control devices, the coal quality and ash content, and the tall of the stack. The taller stack results in a much greater atmospheric dilution.

The atmospheric discharges of natural radionuclides ($^{40}$K, Uranium and Thorium decay series) for these ten plants varied in a range from 22 – 2100 GBq/Gwa being much higher than the emissions of the reference older and more modern CFPPs from the world [1, 4, 8]. Firstly accounting for this was the less ash retention efficiency (lowering till 80%) of the particulate control devices of power stations and secondly, the high ash content of the coal. The mass of fly ash ($10^6$kg) annually released into the atmosphere until 1993 may be the main cause of disturbance of natural radiation background. The activities discharged in the spring – summer period are smaller than what they used to be in winter as a consequence of a difference in the intensity of energy production of CFPP and repair work usually carried out in the summer. Our recent measurements have failed to reveal a significant increase in the natural radiation. The resulting normalized atmospheric discharges have decreased at 6 - 150 GBq/Gwa in the last years because CFPPs were modernized and equipped with high efficiency filtering devices, even if without rising of the stack height. The unsuitable installations have been eliminated. The average values of the natural radioactivity of the particulate deposited at different km distant from the emission source, for two plants using the same coal, having the same power but different ash retention efficiency and tall of the stack are shown in Figures 1 and 2.

As a result of the activity deposited on soil, we found two times higher concentrations of uranium, thorium, radium-226 and potassium-40 in the upper 5 cm layer than in the 5-15 cm layer of soils in the surrounding of the CFPPs, especially for the older ones. The only minor excess in specific activity results in an increase in exposure to gamma rays. The absorbed dose rate in air (at 1 m height) from gamma field of $^{238}$U, $^{232}$Th series and $^{40}$K was estimated to be of 57 – 180 nGy/h the values being of two times higher than typical background in other zones not related with CFPPs. It was obvious that the doses were highest at locations 1-2 km from the stack as well as on the grounds of the CFPP in the direction of winds. One can notice that in the last years, the radiation exposure levels diminished strongly (27 – 90 nGy/h) and these last values are comparable with that reported in literature [9, 10]. The vegetation samples showed higher local activities for all the radionuclides with the highest thorium-232 content (up to 80 Bq/kg). Measurements in surface air had shown the presence of enhanced concentrations of up to 44 Bq/m$^3$ of $^{222}$Rn around the older plants, compared with the EEC maximum value of about 6 in Romania [11].
FIG. 1. *The average natural radioactivity of the atmospheric deposition around a CFPP with fly ash removal efficiency of 90% and stack of 80m high.*

FIG. 2. *The average natural radioactivity of the atmospheric deposition around a CFPP with fly ash removal efficiency of 98 % and stack of 180m high.*

Snow showed high local activity values for uranium-238, radium-226 and thorium-232, as direct results of CFPPs emissions. From those three radionuclides, the values of radium-226 concentrations were highest ones ranging from 1 up to 95 mBq/kg. The average concentration of the natural radionuclides in snow around two CFPPs are presented in Table II. The first plant (A) has fly ash removal efficiency of 90 % and stack of 80m high. The second one (B) is equipped with electrofilters allowing ash retention of 99% and has the tall of the stack of 180m.

The huge amounts of solid wastes (which are TENORM) originating in CFPPs, cover large areas of ground, (a typical ash disposal landfill being anywhere from 30 to 60 hectares), giving rise to imminent ecological problems. The background radiation levels were two to fourfold higher in ash pond or dump areas, as compared to other locations in relation with operation of CFPPs. On the ash dumps the gamma dose rates ranged between 90 and 320 nGy/h. Some of the oldest dumps waste have been filled and have been covered with soil and used as plough land [7]. The higher levels of the radionuclide content in coal
ash stored respectively in the upper soil are reflected in the finding values for the food samples from this area. (Table III).

### Table II. The average concentration (mBq/kg) of the natural radionuclides in snow around two CFPP

<table>
<thead>
<tr>
<th>CFPP</th>
<th>Radionuclide</th>
<th>Distant from the emission source</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>100 m</td>
</tr>
<tr>
<td>A</td>
<td>Radium-226</td>
<td>27±43</td>
</tr>
<tr>
<td>B</td>
<td></td>
<td>4.5±3.1</td>
</tr>
<tr>
<td>A</td>
<td>Uranium-238</td>
<td>5.3±2.8</td>
</tr>
<tr>
<td>B</td>
<td></td>
<td>2.5±1.9</td>
</tr>
<tr>
<td>A</td>
<td>Thorium-232</td>
<td>2.6±4.2</td>
</tr>
<tr>
<td>B</td>
<td></td>
<td>1.8±2.3</td>
</tr>
</tbody>
</table>

### Table III. Average and range of values for specific activity of natural radionuclides in samples related to ash dump

<table>
<thead>
<tr>
<th>Type of sample</th>
<th>(^{238}\text{U})</th>
<th>(^{226}\text{Ra})</th>
<th>(^{210}\text{Pb})</th>
<th>(^{232}\text{Th})</th>
<th>(^{40}\text{K})</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cereals (grains)</td>
<td>0.085 ± 0.021</td>
<td>0.58 ± 0.08</td>
<td>0.31 ± 0.09</td>
<td>0.115 ± 0.011</td>
<td>284 ± 27</td>
</tr>
<tr>
<td>Leafy vegetables</td>
<td>0.036 – 0.160</td>
<td>0.18 – 0.94</td>
<td>0.11 – 0.53</td>
<td>0.028 – 0.392</td>
<td>65 – 490</td>
</tr>
</tbody>
</table>

The values of measured concentrations in harvest on oldest piles are comparable but higher than the corresponding values for the same crops in Romania, especially for uranium and radium [6, 12].

### 3.2. Phosphate industry

In the case of the phosphate industry, the products, by-products and wastes resulting from the applied wet processing procedures contain the different fractions of uranium, radium, thorium and potassium, which were initially present in the phosphate rock [4,6, 13]. Four plants are processing the phosphate rocks in Romania. We have studied one phosphate fertilizer plant (PFP) in Bacau district from eastern Romania, which uses the sedimentary phosphate rock with \(^{238}\text{U}\) specific activity of 700 up to 2200 Bq/kg. Table IV summarizes the characterization of raw material, end products and by product waste from the PFP in terms of activity concentration.

### Table IV. Activity mass concentrations of natural radionuclides.

<table>
<thead>
<tr>
<th>Type of sample</th>
<th>Mean and range activity mass concentrations (Bq/Kg)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>(^{238}\text{U})</td>
</tr>
<tr>
<td>Phosphate ore</td>
<td>967 – 798</td>
</tr>
<tr>
<td>Fertilizer (NPK)</td>
<td>382</td>
</tr>
<tr>
<td>Fertilizer (NP)</td>
<td>528</td>
</tr>
<tr>
<td>Dicalcic phosphate</td>
<td>465</td>
</tr>
<tr>
<td>Phosphoric acid</td>
<td>789</td>
</tr>
</tbody>
</table>
During the wet process, there is a selective separation and concentration of radionuclides. About 80% of the radium-226 follows the phosphogypsum that is the principal waste by-product generated during the phosphoric acid production process (wet process). The greatest part of the uranium (about 85%) and of the thorium (about 70%) is transferred to the phosphoric acid. The redistribution of uranium and their daughters in the varied wares, by-product, wastes and liquid or gaseous effluent gives rise to a multitude of radiation sources.

Gases, aerosols and radioactive dusts represent the airborne discharge. The radon gas and the radioactive aerosols engendered from it undergo a very strong atmospheric dilution, and it is difficult to measure reliably the amount of radon present in air. The ambient air samples outside plant had $^{222}\text{Rn}$ concentration very close to those founded in the whole district ($5 – 53 \text{ Bq/m}^3$).

Liquid discharges are represented by the industrial used waters that contain hundreds GBq (uranium with its descendents) when they enter in the treatment water station. Through chemical treatment these waters with a radium-226 concentration of (3-18 Bq/L) and uranium-238 (0.2 - 0.9 Bq/L) suffer an important decontamination of about two orders of magnitude and then are withdrawn in Bistrita river, by an open canal. Nevertheless, they send both in suspension and in solution towards emissary about $10^8$ Bq of each radioelement ($^{238}\text{U}, ^{226}\text{Ra}, ^{210}\text{Pb}-^{210}\text{Po}$) belonging to the uranium series. The radioactive content of the suspensions reached a level of $\sim 11$ Bq/L involving 98% of existent radium-226. The influence of liquid effluents on the radioactivity of emissary was investigated by water and sediment samples drawing upstream and down stream of the spillway point. The natural radioactivity of Bistrita river down stream of chemical plant raised by an order of magnitude the level found upstream but lower than the Maximum Activity Concentrations admitted in drinking water in Romania. The results showed obvious a lowering by degrees of radioactivity concentrations as far as the junction of Bistrita and Siret rivers (2 km downstream).

No increase of $^{226}\text{Ra}$ concentration in the underground water could be discerned excepting the phosphogypsum dump zone. Underground sources inside or outside of plant show low variations of natural radioactive content. Only the water from well nearest the phosphogypsum stack dump have highest activity values indicating an underground infiltration by a slowly dissolving process of radioelements [14].

Solid wastes represented especially from phosphogypsum were deposited in one pile, which contain rough $10^7$ tons (about 1 MBq/t of radium-226). The storage of phosphogypsum in dump can rise many ecological problems. This stack of phosphogypsum is just near by the town. Radon flux rates from phosphogypsum stack have been founded to vary from 0.12 to 0.44 Bq/m$^2\cdot$sec, with an average value of 0.30 Bq/m$^2\cdot$sec, most likely due to the radium concentration in the parent rock. Because the gamma radiation exposure rates have been around 0.35 $\mu$Gy/h, exceeding specific values for the other areas with normal radioactive natural background [11], only just near the phosphogypsum pile, we consider this pile do not pose a significant radiation protection problem. At present a new dump in conformity with environmental radiation protection rules is built. But, for the improvement of actual situation, it is trying the remediation of the old pile to reduce direct gamma radiation and radon exhalation. The phosphogypsum is not used as building material. There are most important reasons why this by-product waste is unsuitable building material, like its high hygroscopicity. But the main reason is its high radium-226 content, which is one hundred times that of natural gypsum and 10-15 times higher than usual building materials [15, 16].

The soil contamination originates in the spreading of dust from rock and solid wastes dump as well as in the overflow of wastewater from treatment ponds during excess rainfall. It did evident that only the soil inside of plant precincts has a radioactivity exceeding the mean values found in Bacau district. Measurements in soil samples had shown the enhanced $^{226}\text{Ra}$ concentrations of up to 265 Bq/kg (with an average value of 78 ± 44 Bq/kg) around the plant compared with the normal concentration of about 38 Bq/kg in Romania [6, 10, 11].
3.3. **Oil/gas industry**

The exploitation of natural gas/oil reservoirs is associated with the production of significant field water quantities. The formation water volumes resulted in certain segments adjacent to the complex gas/oil production process may amount for large structures to million liters per year. Ground water that coexists with deposits of oil can have unusually high concentrations of dissolved constituents that build up during prolonged periods of water/rock contact. Within the field water composites, besides a series of acid and basic ions, there are variable sodium chloride quantities dissolved in its mass (80 – 130 g/L). Uranium and thorium compounds are mostly insoluble and as oil and gas are brought to the surface, remain in the underground reservoir. Many oil-field waters are particularly rich in chloride, and this enhances the solubility of other elements including the radioactive element radium. Radium concentrations tend to be higher in more saline water. Some of this saline, radium-bearing water is also extracted with the oil and gas. Some radium and radium daughter compounds are slightly soluble in water and may become mobilized when this production water is brought to the surface. This field wastewater is a strong polluting agent and must be separated and then disposed, usually by return to depth in depleted formations by means of injection wells. NORM levels in produced water, scale and sludge usually are characterized in terms of radium activity level (Radium-226 and Radium-228). Because the activity level of $^{226}$Ra usually is three times that of $^{228}$Ra, Radium-226 is the primary isotope of concern with respect to long term radiological concerns in environmental impact [17,18].

Since in the Eastern Romania there are four oil field which represent about 30% from Romanian oil-field exploitation (over 10,000 wells) we aimed to the assessment of radium-226 content of oil-field waters and possible pollution of these zones with this radionuclide. Radium-226 concentrations from 0.005 Bq/L up to more 10 Bq/L were to be found in the formation water samples arising from oil wells, water injection wells and treating stations. (Table V).

<table>
<thead>
<tr>
<th>DISTRICT</th>
<th>OIL FIELD</th>
<th>Water type</th>
<th>Radium-226 (Bq/L)</th>
<th>Average ± SD</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td><strong>Range</strong></td>
<td><strong>Average ± SD</strong></td>
</tr>
<tr>
<td>Bacau</td>
<td>Moinesti</td>
<td>Formation water</td>
<td>0.07 – 10.54</td>
<td>3.72 ± 1.99</td>
</tr>
<tr>
<td></td>
<td>Modarzau</td>
<td>Formation water</td>
<td>0.17 – 4.45</td>
<td>2.53 ± 1.62</td>
</tr>
<tr>
<td></td>
<td>Zemes</td>
<td>Formation water</td>
<td>0.15 – 4.39</td>
<td>1.72 ± 1.53</td>
</tr>
<tr>
<td></td>
<td>Moinesti, Zemes, Zemes, Modarzau</td>
<td>Ground and surface water</td>
<td>0.003 – 0.199</td>
<td>0.053 ± 0.039</td>
</tr>
<tr>
<td></td>
<td>Bordei Verde</td>
<td>Formation water</td>
<td>0.052 – 0.077</td>
<td>0.067 ± 0.008</td>
</tr>
<tr>
<td></td>
<td>Oprisenesti</td>
<td>Formation water</td>
<td>0.005 – 0.019</td>
<td>0.009 ± 0.005</td>
</tr>
<tr>
<td></td>
<td>Ianca</td>
<td>Formation water</td>
<td>0.034 – 5.05</td>
<td>1.70 ± 2.05</td>
</tr>
<tr>
<td></td>
<td>Bordei Verde, Oprisenesti, Ianca</td>
<td>Ground and surface water</td>
<td>0.007 – 0.127</td>
<td>0.044 ± 0.032</td>
</tr>
<tr>
<td>Braila</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

On should remark that 80% of samples exceeded the specific activity of 0.88 Bq/L admitted in Romania for surface water. We must give this problem our careful consideration taking into account that these formation waters are not reinjected in all situations, being accidentally discharged into watercourses and into cultivated or meadow lands. The radium-226 concentrations of the ground and surface water in the same territory were much smaller than that in oil-fields water, for the time being. The soil samples showed somewhat higher local activities up to about 300 Bq/kg for radium-226, respectively 120 Bq/kg for uranium-238.

The present NORM in oil and gas production streams occasionally accumulates as scale or sludge in tubing and surface equipment. At oil-field sites the pipes and tanks that handle large volumes of the "produced water" can become coated with scale deposits that contain radium. Radium-bearing scale is the
type of "diffuse NORM waste" that commonly occurs in the oil industry. Since the radium concentrations in the original formation are highly variable, the concentrations that precipitate out in sludge and as scale on internal surface of oil and gas production and processing equipment are also variable. The scale is relatively insoluble and may vary in thickness from a few millimeters to more than 2 cm. We found the highest concentrations of radium reaching up to ten thousand of Bq/kg in scale deposited in wellhead piping and in production tubing near wellhead. There was a decrease of this concentration of radium deposited in separators (up to one order of magnitude), the lowest value being found in heater treatment devices.

Some of the solids in the original product stream are removed in the separator, the treatment equipment and tanks and accumulate there as sludge. Radium-226 concentrations in sludge were much lower than concentrations in pipe scale. (Table VI). Radium-226 dominates sludge and pipes scale accumulations, while deposits on interior surfaces of gas plant equipment are predominantly Lead-210 with its decay product Polonium-210, up to 3000 Bq/kg.

Table VI. Activity concentration of $^{226}$Ra in scale and sludge samples

<table>
<thead>
<tr>
<th>Type of sample</th>
<th>Radium-226 (Bq/kg)</th>
</tr>
</thead>
<tbody>
<tr>
<td>SCALE</td>
<td>287 – 9260</td>
</tr>
<tr>
<td>SLUDGE</td>
<td>21 – 330</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Range</th>
<th>Average</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>3200 ± 1310</td>
</tr>
<tr>
<td></td>
<td>120 ± 67</td>
</tr>
</tbody>
</table>

Therefore, all the oil field equipment must be surveyed for the presence of enhanced natural radioactivity. Some scale is removed at the well site during work over operations. Most scale is recovered from equipment when this is sent to a facility for cleaning. The relatively higher external dose rates up to 400 nGy/h, (two-four times natural radiation background) were associated with areas around water discharge and handling system and descaling of pipes.

4. Overview of NORM in CFPPs, PFP and petroleum industry

The normal and enhanced levels of environmental natural radioactivity associated with those three non-nuclear industries are reported comparatively in Table VII.

Table VII. Typically encountered activity concentration of natural radionuclides associated with non-nuclear industries.

<table>
<thead>
<tr>
<th>Component of Environment</th>
<th>Industry</th>
<th>$^{238}$U (Bq/kg)</th>
<th>$^{226}$Ra (Bq/kg)</th>
<th>$^{232}$Th (Bq/kg)</th>
<th>$^{40}$K (Bq/kg)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Soil</td>
<td>CFPPs</td>
<td>10 – 86</td>
<td>8 – 156</td>
<td>11 – 96</td>
<td>400 - 1200</td>
</tr>
<tr>
<td></td>
<td>PFP</td>
<td>5 – 153</td>
<td>30 – 265</td>
<td>7 – 44</td>
<td>105 - 780</td>
</tr>
<tr>
<td></td>
<td>Oil-field</td>
<td>2.4 – 120</td>
<td>60 – 330</td>
<td>8 – 87</td>
<td>53 - 960</td>
</tr>
<tr>
<td></td>
<td>Usual values</td>
<td>8 – 60</td>
<td>8 – 72</td>
<td>11 – 75</td>
<td>250 – 1100</td>
</tr>
<tr>
<td></td>
<td>CFPPs</td>
<td>1.9 – 11.2</td>
<td>2.4 – 10.3</td>
<td>1.6 – 15.9</td>
<td>76 - 655</td>
</tr>
<tr>
<td>Surface water</td>
<td>PFP</td>
<td>5.9 – 160</td>
<td>3 – 180</td>
<td>3.1 – 19.5</td>
<td>43 - 580</td>
</tr>
<tr>
<td></td>
<td>Oil-field</td>
<td>0.04 – 1.1</td>
<td>23 – 45</td>
<td>0.2 – 8.0</td>
<td>221 - 899</td>
</tr>
<tr>
<td></td>
<td>Usual values</td>
<td>0.35 – 18.5</td>
<td>1.8 – 22.5</td>
<td>1.5 – 12.2</td>
<td>25 - 670</td>
</tr>
<tr>
<td>Spontaneous vegetation</td>
<td>CFPPs</td>
<td>0.1 – 61.0</td>
<td>0.7 – 40</td>
<td>1.5 – 83</td>
<td>220 - 1450</td>
</tr>
<tr>
<td></td>
<td>PFP</td>
<td>34 – 376</td>
<td>8.0 – 549</td>
<td>3.0 – 53</td>
<td>523 - 1390</td>
</tr>
<tr>
<td></td>
<td>Oil-field</td>
<td>0.2 – 55</td>
<td>3.7 – 59.2</td>
<td>0.005 – 0.012</td>
<td>710 - 1100</td>
</tr>
<tr>
<td></td>
<td>Usual values</td>
<td>0.7 - 48</td>
<td>1.8 – 18.7</td>
<td>1.6 – 3.5</td>
<td>350 - 640</td>
</tr>
</tbody>
</table>
The concentrations of uranium, thorium and potassium in the upper 5cm layer of soil were generally higher in the surroundings of the investigated plants and wells but were comparable to the representative values for the Romania. The average values of annual absorbed dose rate in air from terrestrial gamma radiation (mGy/y) related to these non-nuclear industries are respectively of 0.74, 0.65 and 0.61 for CFPPs, PFP and oil field. These values are comparable, higher however, than the average annual absorbed dose rate in air from terrestrial gamma radiation in Romania of 0.52 mGy/y [11, 17].

The natural radioactivity levels for thorium-232 and potassium-40 detected in surface water are generally comparable to those found in the other zones of Romania. The values for uranium-238 and radium-226 are significantly higher than the corresponding concentrations found generally in Romanian surface waters [10, 18, 19]. One should remark that the values for neither of the analyzed groups of samples exceeded the level for specific activity admitted in Romania even for fresh water, for any of the natural radionuclides under investigation excepting some values for radium-226.

The vegetation samples showed somewhat higher local activity values for all four radionuclides, with the highest uranium-238 and radium-226 content of up to 380 Bq/kg, respectively 550 Bq/kg, related to PFP working, being much higher than that determined in other areas [18].

It can be observed that natural radioactivity levels in the environmental media, which could represent a risk for the people living in the areas influenced by these non-nuclear industries, do not indicate an unquestionable increase in the natural radiation background, excepting some particular areas very close to these non-nuclear activities. It should be noted that many questions regarding dangers in the area of radiation protection versus natural radiation protection remain controversial.

5. Concluding remarks

♦ The releases from non-nuclear industries represent a significant source of chronic environmental contamination with natural radioactive elements. The NORM concentrations in these non-nuclear industries vary from background levels, requiring no special precautions, to elevate levels (TENORM). The continued releases of these materials to environment may result in a buildup in the air, water and soil of the radionuclides, particularly radium-226. There will be an increase of the basic radiation rate in the neighborhood area of these non-nuclear activities or/and plants.

♦ The relatively large levels of radioactivity that result have to be disposed of in a responsible way. The solid wastes originated in some non-nuclear industries cover large earth areas giving rise to ecological problems. The improper disposal and re-use of these wastes should partially clear up such ecological problems but should lead to circumstances resulting in contamination events and increase the potential for unpredictable exposures.

♦ From a radiological point of view, the situation does not pose any immediate concern. Although adverse health effects from increased natural background radioactivity may seem unlikely for the near term, long-term accumulation of radioactive materials from the investigated non-nuclear industries could pose serious health hazards.

♦ Some places however need further investigations, with special emphasis on the control of $^{226}$Ra releases. In some places restriction of land use, or even remediation would be advisable.

♦ Our data enable comparisons to be made with the other practices and other countries. They suggest that the occurrence of TENORM is significant enough to warrant increased control of radioactivity aiming to regulate this. Our results will be useful for the future Romanian radiation protection
standards regarding the possession, use, transfer and disposal of Technologically Enhanced Naturally Occurring Radioactive Materials.

6. References

5. CNCAN, Norme Fundamentale de Securitate Radiologica, (Basic Safety Standards in Romania), 2000