THE JOURNAL OF PREVENTIVE MEDICINE
2003; 11 (1): 19-27

THE HAZARD OF NATURALLY OCCURRING RADIOACTIVE MATERIALS FOR WORKERS IN PHOSPHATE FERTILIZERS INDUSTRY AND ENERGY PRODUCTION

Elena Botezatu, Olga Iacob

Institute of Public Health

Abstract. The aim of this study was to assess the exposure of workers in two industries due to work activities involving naturally occurring radioactive materials (NORMs). Our research was intended to provide a better knowledge of working practices and to provide sufficient meaningful information regarding the estimated exposures to workers in these industries due to NORM, focusing in particular on the regulatory aspects relating to naturally occurring radioactive materials. The workers involved in the production of energy in Coal Fired Power Plants (CFPPs) and in fertilizers production in phosphate fertilizer plant (PFP) are subject to radiation exposure through internal and external pathways. In CFPPs, the inhalation of dusts containing naturally occurring radionuclides was found to be the most significant exposure pathway. The highest risk was associated with the intake via inhalation of thorium-232, which contributes up to 41 % to the annual effective dose. Exposures to individuals resulting from inhalation of radon and thoron progeny represent only 5%, respectively 1.5% from the whole dose. In PFP, the terrestrial gamma external radiation delivers the highest contribution to overall exposure of workers from natural sources (42%), followed by radon along with its decay products (36%). Inhalation of dusts containing naturally occurring radionuclides was found to be an important exposure pathway (22%). For workers employed in the CFPPs, the higher risk is associated with the intake (via inhalation) of thorium-232, while in PFP the higher risk is associated with the intake (via inhalation) of radium-226 and uranium-238.

The results show that occupational radiation doses for some workers reach relevant levels compared to protection limits in the nuclear industry. These individual dose levels therefore should be carefully measured, controlled and registered. We consider that in PFP there are work activities that must be subject to control. Optimization techniques to reduce individual and collective doses in the phosphate production should be established.

Key words: non-nuclear industries, naturally occurring radioactive materials, workplaces, radiation exposure

Rezumat. Industriile non-nucleare identificate ca surse de radioelemente naturale includ producția de energie prin utilizarea combustibilor fosili şi industria îngrăşămintelor fosfatice. In centralele termice pe bază de cărbune sau in combinatele de îngrăşăminte chimice fosfatice, prin procesele tehnologice are loc concentrarea substanțelor radioactive în produsele finale sau secundare precum şi în eliminările gazoase, lichide sau solide. Datorită acestei concentrări, deşi cantitățile de radionuclizi din unele materiale prelucrate sunt considerate destul de mici, poate apare pericolul iradierii în unele medii de muncă. Problema se complică prin faptul că această expunere poate apare în locurile de muncă unde nu este percepută necesitatea protecției radiologice. Prezentul studiu are drept scop obținerea unui set comprehensiv de informații care să constituie baza științifică pentru deciziile asupra controlului radionuclizilor naturali din cele doua industrii non-nucleare. In centrale electrotermice (CET) moderne ce utilizează cărbune, situate în Nord-Estul României şi într-o uzină de îngrăşăminte fosfatice (UIF) s-au estimat expunerile la radiații ionizante pentru angajați, exprimate în termeni de doză efectivă, ca şi riscul asociat. În CET, inhalarea pulberilor care conțin radionuclizi naturali este principala cale de expunere, riscul cel mai mare pentru lucrători fiind asociat inhalării de toriu-232, care contribuie cu până la 41 % la doza efectivă anuală. Expunerea rezultată din inhalarea descendenților radonului şi toronului reprezintă numai 5% respectiv 1,5% din întreaga doză. In UIF iradierea gama externă are cea mai mare contribuție la iradierea muncitorilor din toate sursele naturale de iradiere (42%), urmată de radon împreună cu descendenții săi (36%). Inhalarea pulberilor care conțin radionuclizi naturali este o cale importantă de expunere (22%), riscul cel mai mare pentru lucrători fiind asociat inhalării de radiu-226 şi uraniu-238. In uzina de îngrăşăminte fosfatice există activități care trebuie supuse unui control adecvat din punct de vedere al radioprotectiei.

Cuvinte cheie: industrii non-nucleare, materiale ce conțin radioelemente naturale, locuri de munca, expunerea la iradierea naturală

INTRODUCTION

The technological processes evolved in the phosphate industry and during the production of energy on the fossil fuels basis, the concentrations of natural radioactive elements in the products and in the wastes can be much higher than in the ore/raw materials. These raw materials, their by-products and the end products from the chemical or physical processing may lead to workers exposure (1). Such exposures often occur in workplaces where there is no perception among staff of the various relevant radiation protection problems.

The aim of this study was to assess the exposure of workers in these two industries due to work activities involving naturally occurring radioactive materials (NORMs) being the first one on this issue. Our research was intended to provide both a better knowledge of working practices and meaningful information on workers' exposure to NORM in two non-nuclear industries, focusing on the regulatory aspects relating to naturally occurring radioactive materials.

MATERIALS AND METHODS

Between 1996 and 1999 four modern coal-fired power plants (CFPPs) and one phosphate fertilizer plant (PFP) have been investigated. Knowing that the relevant pathways of exposure to workers are external gamma irradiation, dust and radon inhalation, the following steps were taken:

- Extensive measurements of natural radioactivity content in raw materials (coal, sedimentary phosphate rocks), end products, by-products (fertilizers, phospho-gypsum), wastes (slag, ash) and dust at workplaces in these selected plants by high-resolution gamma spectrometry techniques and radiochemical and physical methods carried out in conformity with the current national standards (2).

- Active measurements of gamma ray dose rate were carried out in all working places using Inspector Nuclear Radiation Monitor (MEDCOM) – SUA (autoscale of 0.01 μSv/h – 1 mSv/h).

- The ^{222}Rn and ^{220}Rn short-lived decay product concentrations in air in all of the workplaces were measured.

During a month, the air was sampled daily with respect to the day moment and to 60 cm height above the floor. The individual specific activity values for the attached fractions of ^{218}Po, ^{214}Pb, ^{214}Bi and ^{212}Pb were determined in accordance with the national standard (3). The relative errors affecting the specific activities determined for radon daughters were 15% for ^{214}Pb and ^{214}Bi, 30% for ^{218}Po and 10-20% for ^{212}Pb. The method for measuring radon decay products involved pumping a known volume of sample air through an open-faced, high-efficiency filter paper at a known flow rate for a certain sampling time. The filter paper was removed and then the deposited activity, which was assumed to be due solely to ^{222}Rn and ^{220}Rn daughters, was counted using a ZnS scintillation counter over a specified period of time.

Equilibrium equivalent concentration (EEC) values were calculated on the basis of the following equations provided in the UNSCEAR Repport of 2000 (1):

$$EEC\ (^{222}Rn\ series) = 0.105C_1 + 0.515C_2 + 0.380C_3$$

$$EEC\ (^{220}Rn\ series) = 0.913\ C_1 + 0.087\ C_2$$

where the symbols C_1, C_2 and C_3 represent respectively the activity concentrations of ^{218}Po, ^{214}Pb and ^{214}Bi, for the ^{222}Rn series, and the symbols C_1 and C_2 represent likewise the activity concentrations of ^{212}Pb and ^{212}Bi from the thoron series.

- The total amount of inhalable dust was determined using the Casella Method MDH S14 using an open-face head fitted with a type A glass fibre filter (∅ 37 mm), manufactured by Whatman (RB). The indoor atmosphere in the work places was also characterized by grading the particle distribution (4).

- Radiation exposure to workers in both industries (298 employed in CFPPs, and 151 in the PFP) was expressed in terms of individual and collective effective dose, using the latest ICRP models and the dose-conversion coefficients published by the ICRP and UNSCEAR (3,5,6). The organ doses for the workers were calculated using the average breathing weighted rate of 21.3 $m^3 . d^{-1}$ (6) and the ^{226}Ra, ^{232}Th, and ^{238}U dose coefficients for inhalation, relating to an adult (7).

RESULTS AND DISCUSSION

Workers radiation exposures have been evaluated from measurements and samples drawing in all workplaces. So, for CFPPs workers were: the coal storage areas and bunkers, coal breakers and mills, conveyor belts, stoker platforms, electro-filters, steam turbines, pumps, workrooms and workshops, laboratories.

For phosphoric acid and fertilizer sections of the PFP the following places were investigated: the unloading ramp, storage, drying and grinding areas, the conveyer belt for phosphateous rock, the areas for burdening preparation, filtration, concentration, defluorination, storage of H_3PO_4, conveyer belts, areas for the drying and storage of phosphogypsum, neutralization, drying, graining, crushing, sorting, the conveyer belt and storage areas for chemical fertilizers.

The gonadal doses from external gamma exposure in workplaces varied from 0.12 μSv/h to 0.75 μSv/h for workers employed in **CFPP**s and from 0.17 μSv/h to 1.07 μSv/h for workers employed in the **PFP**, respectively. Radiation surveys conducted in area where large volumes of phosphate ore are stockpiled have yielded gamma exposure rates ranging from 0.48 μSv/h to 1.07 μSv/h. These values are much higher than the average per capita outdoor dose in Romania due to terrestrial gamma radiation (0.059 μSv/h, range values: 0.021-0.122 μSv/h). The assessed values of annual external gamma radiation doses in both industries lay within a range from 0.23 mSv up to over 2 mSv.

The measurements of the individual concentrations of ^{222}Rn and ^{220}Rn daughters in **CFPP** lead to radon and thoron equilibrium equivalent concentrations varying in a large range. The highest values were found in workrooms, workshops and laboratories, due to the radioactive content of the building materials used. The EEC varied within a range from 0.07-35 Bq/m^3 for radon and within one of 0.03-1.11 Bq/m^3 for thoron, respectively. It was difficult to measure reliably the amount of radon present in the air of working places with high levels of suspended dust (45-200 mg.m^{-3}), the latter representing the major source of contamination in these workplaces (9). We do not know how much from ^{222}Rn is retained within dust particle and which is the proportion of radon gas escaping from dust particles.

As was to be expected, in the **PFP** the highest values for ^{222}Rn gas were found in the areas for phosphate rock processing (35-835 Bq/m^3), phosphogypsum production (73-655 Bq/m^3) and at some locations in the phosphoric acid section (22-430 Bq/m^3), but without exceeding the action level of 1000 Bq/m^3 (10). EEC values ranged from 15-156 Bq/m^3 for radon, higher than those of CFPPs. The thoron EEC values ranged between 0.003 and 0.6 Bq/m^3, lower than those of CFPPs.

In Romania, the average indoor EEC is 25 Bq/m^3 for ^{222}Rn and 1.1 Bq/m^3 for ^{220}Rn (8). The radon and thoron equilibrium equivalent concentrations and the annual effective doses received by an individual worker in both the CFPPs and the PFP are presented comparatively in table 1.

Table 1. Annual internal and external exposures to workers in workplaces

Location	Mean values (range)					
	Internal irradiation				External irradiation	
	EEC (Bq/m^3)		E (μSv/y)		Dose rate (μGy/h)	E (μSv/y)
	^{222}Rn	^{220}Rn	^{222}Rn	^{220}Rn		
CFPP	6.5 ± 6.9 (0.07-35)	0.36 ± 0.29 (0.03-1.1)	112 (1.2-605)	27.5 (2.3–84.5)	0.43 (0.12-0.75)	825 (230–1,440)
PFP	48 ± 34 (15-156)	0.26 (0.003-0.6)	829 (259-2,690)	20 (0.2-46)	0.46 (0.17-1.07)	880 (325–2,054)

The annual exposures received by the individual workers were determined taking into account the EECs for radon and thoron, and assuming a total working time of 1920 hours per year. Effective doses (E) were calculated using dose conversion factors of respectively 9 nSv/Bq h m^{-3} for radon progeny and 40 nSv/Bq h m^{-3} for thoron progeny (1).

The average values for effective doses due to the inhalation of radon and thoron and their daughters were lower in the **CFPPs** than those found in the dwellings (550 μSv/y for radon, respectively 165 μSv/y for thoron) (8,11).

In CFPPs the particle size varied from less 0.4 μ (3%) up to greater than 6μ (20%), with prevailing dust particles of 2-3 μ (37%). The up to 4 μ particles, which are considered to be breathable particles (12), represented about 75% of the total. The lighter fly ash material with the highest radioactivity was present in many workplaces as dust. The amount of dust varied within a range of 3.14 mg m^{-3} up to 206 mg.m^{-3}, exceeding in all workplaces the admissible concentration of 2 mg.m^{-3} (13).

In **PFP** the radioactive dust arise during the operations of the discharge, transport and crushing of phosphate rock, but also during the process of the re-suspension of solid radioactive wastes (phosphogypsum and sludge). Particle sizes varied from a value of less than 0.4 μ (2.6%) up to values of higher than 6μ (8%), with dust particles 1-2 μ (39%) prevailing. The breathable particles were present in proportion of about 81%. Dust concentrations inside the plant ranged between 10 and 100 mg/m^3, with corresponding values of up to 0.2 Bq ^{226}Ra/m^3. Although the specific activities of uranium (0.01-0.25 Bq/m^3) varied from one work place to another, they always exceeded the typical value for environmental air of (0.3 . 10^{-5} Bq/m^3) (14).

The comparative data of occupational exposure and radiation risk in both CFPP and PFP are shown in table 2.

Table 2. Occupational exposure and radiation risks in CFPP and PFP

	Measure unit	CFPPs	PFP
Workers	Number	298	151
Work time	hours/year	1920	1920
Dust loading	mg/m^3	3 - 206	10 – 100
Particle sizes	microns	< 0.4 μ (3%)	< 0.4μ (2.6%)
		> 2μ (37%) < 3μ	>1μ (39%) < 2μ
		> 6μ (20%)	> 6μ (8%)
Breathable particles	microns	Up to 4μ (75 %)	Up to 4μ (81 %)
Radioactive content Uranium– 238 Radium – 226 Thorium – 232	mBq/g powders	 28 - 176 37 - 206 30 - 360	 87 - 23750 170 - 18600
Effective dose from intake via inhalation Uranium – 238 Radium – 226 Thorium – 232	µSv/y	 0.5 – 16.6 1.6 – 110 24 – 3900	 25 – 618 29 – 580
Effective dose from inhalation of radon and its short-lived progeny Radon - 222 Radon – 220	µSv/y	 1.2 – 605 2.3 – 84.5	 259 – 2,690 0.2 –46
Effective dose from external irradiation	µSv/y	230 – 1,440	365 – 1,997
Potential radiation induced fatal cancers	Cases/no of workers	3×10^{-2}	2×10^{-2}
Potential radiation induced fatal cancers Internal exposure External exposure	Cases per 10^5 people	10.1 6.0 4.1	13.6 9.4 4.2

The annual intake via inhalation of breathable particles containing ^{238}U, ^{226}Ra and ^{232}Th were taking into account to assess the effective doses per unit of intake. The annual committed effective doses were at levels of 0.5-16.6 µSv from ^{238}U; of 1.6-110 µSv from ^{226}Ra and 24-**3900** µSv from ^{232}Th for the workers from **CFPPs**. The resultant annual committed effective doses were at levels of 25-618 µSv from intake of ^{238}U and of 29-580 µSv from intake of ^{226}Ra, for the workers from **PFP**.

The workers employed in the phosphate industry receive the highest total effective doses from the inhalation of radon and thoron and their progeny, as well as from external exposures at work.

Radiation-induced cancer risk was estimated from the annual effective collective doses using two different risk coefficients: $7.3 \cdot 10^{-2} Sv^{-1}$ for internal exposures from inhalation of radon progeny in both workplaces and dwellings (5), and $4 \cdot 10^{-2} Sv^{-1}$ for internal exposures from ^{238}U, ^{226}Ra, ^{232}Th, and for external gamma exposure (15). Exposures in workplaces could be responsible for $3 \cdot 10^{-2}$ potentially radiation induced fatal cancers occurring in workers employed in the CFPPs (298), and $2 \cdot 10^{-2}$ cases in workers employed in the PFP (151). The estimated risk is expressed as the number of potential fatal cancers per 10^5 persons so as to make it possible to compare values. As can be seen the values for risk from external exposures are at the same level in both industries. Fatal cancer risk is, however, higher for internal exposures of workers in phosphate fertilizer plant.

Taking into account the repartition of workers in every workplace with its specific radiological features, the individual and collective annual doses were estimated (tables 3 and 4).

Table 3. Workforce and annual doses

Year	Number of monitored workers		Annual average effective dose		Annual total collective effective dose	
			mSv		manSv	
	CFPPs	PFP	CFPPs	PFP	CFPPs	PFP
1996	376	156	2.51 ± 0.11	4.0 ± 0.19	0.944	0.635
1997	315	151	2.18 ± 0.07	3.10 ± 0.13	0.687	0.469
1998	307	177	1.70 ± 0.08	3.42 ± 0.15	0.522	0.605
1999	298	151	1.44 ± 0.06	2.87 ± 0.14	0.429	0.433

Table 4. Annual collective dose (man Sv) from workers in effective dose intervals

Year	0.2 - 0.99 mSv		1.0 – 4.99 mSv		5.0 – 9.99 mSv	
	CFPPs	PFP	CFPPs	PFP	CFPPs	PFP
1996	0.028	0.033	0.733	0.302	0.200	0.294
1997	0.017	0.029	0.543	0.295	0.126	0.145
1998	0.012	0.027	0.431	0.322	0.090	0.251
1999	0.028	0.029	0.334	0.270	0.066	0.131

It becomes evident that in both type of plants the effective dose limit of 1 mSv/y for public exposure is exceeded (10).

It can be observed that the doses diminished continuously in the period 1996-1999. After 1999, the assessments of radiological situation relied on our sporadic measurements revealed the same decrease of annual effective dose to an average value of 0.57 mSv in CFPPs (2001y), respectively 0.87 mSv in PFP (2002y). This situation can be account on the follows:

- improvement of working conditions as result of an intensive natural and artificial ventilation system;
- modernization of working devices and elimination of absolute and unsuitable installations;
- change of phosphorite with one of lower radioactivity level;
- periodical unemployment (about three month yearly) and decreased industrial activity.

CONCLUSIONS

- The workers involved in the production of energy in CFPP and in fertilizers production in PFP are exposed to radiation through internal and external pathways in addition to noxious mixture of chemicals.
- Inhalation of dusts containing naturally occurring radionuclides was found to be a significant exposure pathway in both industries.
- For workers employed in the CFPPs, the high risk is associated with thorium-232 inhalation, which contributes up to 41 % to the annual effective dose. The higher the content of ^{232}Th in coal and ash, the higher intake of this radioelement is. Exposures to individuals resulting from the inhalation of radon and thoron progeny represent only 5% respectively 1.5% from whole dose.
- In PFP, the terrestrial gamma external radiation has the highest contribution to overall exposure of workers from natural sources (42%), followed by radon along with its decay products (36%).
- Occupational radiation doses for some workers reach relevant levels compared to protection limits in the nuclear industry. These individual dose levels therefore should be carefully measured, controlled and registered.
- In PFP there are work activities that must be subject to control. Optimization techniques to reduce individual and collective doses in the phosphate production should be established.

REFERENCES

1. United Nation. *Sources and effects of ionizing radiation*. UNSCEAR 2000 Report Annex B, 83-157.
2. Botezatu E, Aflorei A: *Non-nuclear industries involving natural radioactivity: radiological impact on environment and workplaces*", Journal of Preventive Medicine, 1997, 5 (4): 13-21.

3. IRS, Air Quality. *Volumetric activity determination of polonium-218, lead-214, bismut-214 and lead-212*, SR 13397, Romanian Standard, Bucureşti, 1997 (in Romanian).
4. Dallas MS: *Air Sampling Handbook*. 1, 3rd Ed., Casella London Ltd. 1994.
5. ICRP. *Protection Against Radon-222 at Home and at Work*. Publication 65, Ann. ICRP, Oxford, 1994, 22 (2).
6. ICRP. *Human Respiratory tract model for Radiological protection,* Publication 66, Ann. ICRP, Oxford, 1994, 24 (1-3).
7. Phipps AW, Kendall GM, et al, *Memorandum. Committed Equivalent Organ Doses and Committed Effective Doses from intakes of radionuclides*, NRPB-M288, Chilton, Didcot 1991.
8. Iacob O, Botezatu E, Grecea C, *Update on natural radiation exposures in Romania*, Proc. 5th Int. Conf. on HLNRRA: Radiation Dose and Health Effects, Munich, 2002, II: 7–10.
9. Botezatu E, Grecea C, Iacob O, *Exposure to radon and thoron of workers in coal fired power plants,* Journal of Preventive Medicine, 1998, 6 (2): 23-28.
10. IAEA. *International BSS for Protection against Ionizing Radiation and for the Safety of Radiation Sources*, 1994.
11. Botezatu E, Iacob O, Branisteanu R, Grecea C, *Assessment of Exposures of Workers to NORMs in Two Non-Nuclear Industries*, Proc. 5th Int. Conf. on HLNRRA: Radiation Dose and Health Effects, Munich, 2002, II: 7-10.
12. ACGIH. *Threshold limits values for chemical substances and physical agents*, ACGIH Publishing Cincinnati, Ohio, 1999.
13. RHM 1957. *Romanian Norms of Occupational Medicine*, 1995.
14. Botezatu E, Iacob O, Clain L, Grecea C, *Radiation Exposure in Some Non-Nuclear Industries Involving natural radioactivity*, Proc. 6th Int. Symp. on Achievements & Challenges: Advancing Radiation Protection into the 21st Century, Southport, UK, 1999, 77.
15. ICRP. *Recommendations of the ICRP*, Publication 60, Ann. ICRP, Oxford, 1994, 21 (1-3).