The needs and feasibility of land reclamation of areas affected by enhanced natural radioactivity

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Abstract

The major efforts devoted to disposal of radioactive materials are focused on those originating from nuclear industry. Far less attention has been paid to environmental burden of waste with natural radioactivity enhanced by non-nuclear industry. Such waste differs significantly from classical nuclear ones and the radiation risk is often associated with risk caused by other pollutants. Contrary to the nuclear waste that's strictly controlled, it has been a common practice to put TENORM *(Technologically Enhanced Naturally Occurring Radioactive Material)* into heaps, where they can reach thousands of cube meters or tonnes, without any protection. Exposure to meteorological conditions sets some chemical or physical processes in motion, leading to the selective transfer and accumulation of radionuclides and disequilibrium in decay series.

As a result of the inconclusive European law, that left the decision where risk caused by TENORM is significant under each member state's competences, non-nuclear industry is hardly ever aware of environmental problems caused by natural radioactivity or expect negative consequences in case of implementing radiation protection measures. This results in the substantial underestimation of the detrimental effects to the environment originating from TENORM. EU member states apart each other try to regulate this problem case by case when but the risk caused by TENORM is rarely taken into account when the treatment of such waste is planned.

The feasibility of different methods of land reclamation was discussed including hard land reclamation, bioremediation and phytotechnology. An approach based on dilution of TENORM with inert materials was considered. Contrary to the nuclear waste, where dilution is totally forbidden, natural radioactivity, which is present in small amounts elsewhere, is a case where such approach to decrease risk sounds rationally. In extraction industry where huge amount of gangue is present, such approach is well-founded also from economic point of view.

Introduction

Natural radioactivity is ubiquitous in human environment. According to the state-of-the-art radioprotection, radiation emitted by primordial radionuclides in their natural state that has not been altered due to human activity is not considered to be a source of harmful effects neither for human beings nor environment. There are many areas in the world having elevated so called "natural background" caused either by the geological and geochemical structure of the rocks, or by the radioactive content of water flowing from underground springs. Whether or not it can cause a negative or positive effect on human beings is a matter of opinion. But if concentrations of natural radionuclides have been changed as deliberate or accidental action carried out by human being it is quite another matter. The classical case where radiation risk caused by natural radioactivity is not negligible is uranium mining and milling. It is abundantly clear that such processes must be carried out at the region where reach uranium ore occurs. But such activity is considered to be an immanent part of nuclear industry to be enclosed in radiation protection domain at the very beginning. After the enhanced natural radioactivity had been thoroughly studied in other industries it became clear that such phenomena are very frequently present in the anthropogenic environment. Many processes beyond nuclear industry lead to a situation where the activity concentration of naturally occurring radionuclides is enhanced. Such alteration of natural state can result in increased radiation risk to the people as well as to environment and non-human biota. Hence, the monitoring and prevention of occupational radiation risks caused by enhanced natural radioactivity has become obligatory in many cases of industry of concern.

Enhanced natural radioactivity is usually associated with industrial processes where a significant mass reduction of raw materials occurs. As a matter of fact, these processes are not aimed at production of natural radionuclides or the deliberate use of radiation. Therefore, radioactive nuclides are often accumulated in useless industrial residues. From the point of view of the general principles of radiation protection, the activity concentration of natural radionuclides in such materials is sometimes high enough to rank them as radioactive waste. Their amount collected in one site frequently riches hundreds of thousands of cubic meters or tonnes. For instance, in coal mining industry radium activity deposited in single tailing ponds may reach as high as 300 GBq (Michalik et al. 2005). Probably, the biggest "producer" of waste with enhanced concentration of natural radionuclides are phosphate processing plants where radionuclides remain associated with the phosphogypsum particles, being subsequently stored in a disposal sites located in the vicinity of the factories with surface dose rate reaching 350 MBq/h (Bolivar et al. 2009). In spite of that TENORM-type waste (TTW) is often deposited directly into the environment, what is strictly forbidden in case of "real" radioactive waste. Contrary to the occupational risk, which is more or less controlled, by far, less attention has been paid to the environmental burden emerging due to TENORM-type waste (TTW). This results in sites where such waste have been dumped not to undergone adequate land reclamation and often even monitoring of radioactive pollution is not carried out. There are only few examples where TENORM type waste are treated in correct way (Welbergen and Wiegers 2008). There are significant number of cases where only non-radiological parameters are effectively taken into account during presumptive land reclamation, mainly due to the lack of proper regulation.

Environmental burden of TENORM residues

There is a lot of data dealing with the behaviour of natural radionuclides being in the natural state on the border of abiotic and biotic matter. Frequently, the processes of metabolism lead to concentration of some long-lived natural radionuclides in particular tissues of fungi, plants as well as animals (McDonald et al. 1996). Derived committed dose can be higher than doses resulted from artificial radionuclides accumulated simultaneously (Aarkrog et al. 1997). There are some examples of societies based on limited trophic chain where the related committed dose to individuals caused by biologically accumulated natural radionuclides such as polonium and lead ingestion is significant. J. Van Oostdam et al. (2005) indicated ^{210}Po derived doses as high as 10 mSv per year for some aboriginal northern communities consuming large amounts of caribou. Either, annual doses reaching 3 mSv were reported for population of fisherman just living on seafood (Alonso-Hernandez et al. 2002), (Camplin et all. 1996). If such processes are going on in unchanged environment, one can easily image what is going to happen in the vicinity waste dump where residues containing enhanced concentration of natural radionuclides had been collected.

Each particular occurrence of TTW presents a unique scenario of exposure – usually different from those caused by artificial radionuclides present in radioactive waste or spent nuclear fuel. As a result of the direct contact with environment, some transformation processes such as mobilisation of radionuclide species from solid phases or interactions of mobile and reactive radionuclide species with components in soils and sediments may be set in motion (Vandenhove and Van Hees 2007). Also considerable transfer of radionuclides to biota can be observed (Soudek et al. 2007). All these result in that the original distribution of radionuclides deposited in environment can change over time. Moreover, natural radionuclides are often associated with other pollutants as heavy metals or hydrocarbons that can escalate negative impact on environment if dumped out of plants. That’s why, to plan the land reclamation of a site affected by TTW, information on radionuclide species deposited, interactions within affected site either local ecosystems or environment compartment and the varying in time distribution of radionuclide species influencing mobility and biological uptake is essential (Tamponnet et al. 2008).

TENORM-type waste characterization

TTW produced by non-nuclear industrial activities such as mineral production, mining activities or coal fired power plants contain a number of long-lived natural occurring radionuclides from the uranium and thorium decay series. The main, from radiation protection point of view, properties of TTW which make them significantly different from “classical” radioactive waste are (Michalik 2007);

- the occurrence in bulk quantities deposited directly in the environment,

- the wide variety of radionuclides speciation and different minerals content,
- the possible coexistence of other pollutants as heavy metals, sulphates, hydrocarbons

Taking into consideration the radionuclides that occur in TTW and either their activity concentration or total activity, some part of them should be classified as radioactive waste containing alpha emitters, (i.e. the limit of activity concentration for radium isotopes is 10 kBq/kg). Actually, the decision to rate TTW among the radioactive waste at all, not only as alpha emitters is rarely taken. It results in technical problems and economical consequences that would follow such decision (to remind, such waste must be sealed and deposited in a repository, in case of alpha emitters in deep underground repository). After they are classified as radioactive waste TTW would generate enormous cost- related implications and fill available repository very quickly. There is no possibility to treat them as such with regard to existing regulation, which had been prepared when thinking about nuclear waste. Finally, there are gaps, no special regulation and no correct way of deposition. Moreover, from environmental point of view, very interesting is, what one should do with waste that contain slightly lower than limit radioactivity content, in case of radium, for example, 9 kBq/kg. Expected effects on environment caused by waste with 10 and 9 kBq/kg of radium should not be significantly different. Actually, the proposed clearance level for natural radionuclides from uranium and thorium decay series have been set at the level of 1 kBq/kg. So, even if one wants to follow the existing regulation quickly will meet the big gap between 1 and 10 kBq/kg.

After deposition TTW in environment, enhanced concentration of natural radionuclides first of all, results in enhanced exposure of biota to external gamma radiation. So, accurate measure of the total radionuclides concentration in these waste materials is crucial to asses the potential radiological risk at a dump site. However, if one managed to gather data about physical presence of each particular radionuclide, this information would give only a part of the knowledge necessary to evaluate its harmful potential. The radiological hazards can be increased by migration process of mobile fraction of these radionuclides to the vicinity of a depository (Sheppard et al. 2005). Being released into an ecosystem, they can enhance the gamma radiation doses to biota. That is the reason why it is crucial to know the mobility of radionuclides (Martinez-Aguirre et al. 1995). The mobility and environmental behaviour of every element depend on their speciation in certain waste material (Zhongwen et al. 2002). The speciation of a radionuclide is generally related to its physical and chemical forms existing, that is, simple and complex ions in interstitial solution, exchangeable ions associated with waste material organic fractions occluded or co-precipitated with metal oxides, carbonates, sulphates and other secondary minerals.

But the exposure to external gamma radiation is only a tip of an iceberg. One should remember that in thorium and uranium decay series there are 7 and 12 alpha particles respectively (additional 12 are in actinium series). Even when one consider decay series starting from radium (^{226}Ra or ^{228}Ra) what is very common in TTW waste the number of alphas decreases only to 7 and 9, respectively. It is hardly ever taken into consideration but in environment, in case of plants, especially plant roots, the exposure to external alpha radiation is as important as exposure to alpha radiation emitted by incorporated emitters. Also one should remember about betas emitted by natural radionuclides. Actually the weighting factors for alpha and beta radiation established for human being can not be directly applied for plants but there are no rational reasons why they should be significantly lower. Direct measurements showed that absorbed dose resulted from alpha radiation can reach the same level as doses from gamma radiation (Michalik 2008).

Besides the problems caused by activity concentration, radionuclides speciation and migration, the evolution of the related risk must be taken into consideration. Namely, the most common radionuclides responsible for risk creation are radium isotopes. In case where source of TTW are formation waters (oil and gas industry, underground mining) radium isotopes, both from uranium and thorium series, ^{226}Ra and ^{228}Ra are dominant. It means that, significant disequilibrium in natural decay series exists. At the beginning, just after deposition, there are almost pure radium as the only contaminant (besides possible other, non radioactive ones, of course). Radium isotopes are usually weakly mobile and not bio-available in environment. As it was proven radium creates not soluble radium-barium sulphate or its atoms are strongly bonded with fine clay minerals. The bio-available part usually does not exceed 1 % of total activity (Leopold et al. 2007). So, the environmental risk is limited only to the exposure to external gamma radiation, or as it was shown above, alpha radiation and finally could be partially limited by a cover from an inert material. Moreover, the part of exposure derived from radium ^{228}Ra will relatively quickly decreases (the half live of ^{228}Ra is 5.7 years, so not supported by long-lived parent radionuclides ^{228}Ra will quickly disappear). The only difficult situation is that, in the decay series started with not-supported radium ^{228}Ra, the activity concentration of decay products i.e. ^{224}Ra can exceed the activity concentration of radium after few years. It can cause troubles during measurements and appropriate actual dose assessment.

Quite different situation exists in case of radium ^{226}Ra. It decays slowly. Not only from the point of view of a human being but also form point of view of the environment it lasts eternally. 1600 years is long enough to be eye witness of changes going on in an ecosystem.

As it was already mentioned, the bio-availability of radium is very weak, but such comfortable situation with immobilised radium does not last long. Taking into consideration the relationships inside the uranium decay chain one should expect not so quickly, but permanent growth of long lived radium daughters as lead ^{210}Pb and polonium ^{210}Po. Both these radionuclides are very well known as easy migrating nuclides in environment and available to biota. After decay, the nuclide of radium is released from the barium-sulphate or clay mineral cage. Moreover, between radium and lead and polonium, there is radon (or thoron respectively) occurring in gaseous form. So, after one hundred years one should expect in the site where TTW with radium had been dumped exactly the same activity of easy migrating and easy bio-available lead and polonium isotopes. Besides specific chemical properties, they are beta and alpha emitters respectively and both of them are chemiotoxic elements.

In summary, the proper land reclamation of sites affected by TTW is an interesting challenge.

Remediation trials

Among noticed approaches to the reclamation of land affected by TTW, the simplest one, compliant with radiation protection rules, is to treat such waste literally as radioactive ones and apply all required restrictions. But for many reasons, it is applicable in limited cases (Michalik 2008,2). Usually, only parts of dewatering systems from crude oil and natural gas industry (Varskog 2007) or gangue from mineral sand processing (Hutchinson and Toussaint 1998) are disposed to especially prepared repositories. Sediments had been created in surface tailings sometimes are treated in special way (Al-Masri and Suman 2003) but usually, they are left without any action. Some promising efforts to wash out radium from oil sludge were made in Egypt (Afifi et al. 2009). Also applications of biotechnology, in order to make radium more mobile from uranium milling waste were noticed (Muñoz et al. 1995). But any of them have been applied in technical scale. In uranium mining and milling industry, where implementation of strict rules of radiation protection has long history, usually a kind of hard remediation is applied. Land-filing and covering with an inert matter is sufficient to limit the exposure to external radiation as well as further radon exhalation (Krizman 1995), (Juhász et al. 2001). But this approach is not sufficient enough to stop radium and further polonium migration.

The possibilities of application of different methods of land reclamation have been considered towards the sites contaminated by radium rich sediments originating from underground coal mining (Chałupnik et al. 2001), (Michalik 2004). The first based on application of phytotechnology was tested on an abandoned mining settling pond. Six-year-lasting observation of contaminated area let one noticed that the process of natural plants transgression was so effective and, even without any support, good enough to stop the physical propagation of contamination. The plants overgrowing the pond created a tight cover able to stop water and air land erosion. It supposes that, in case of controlled and supported propagation of selected plant species, it would be actually an effective and cheap method for immediate land reclamation of contaminated with radium sites.

The possibility of phytoextraction at this area was evaluated for two plant species *Cirsium vulgare* and *Calamagrostis epigejos*. The balance of radium ^{226}Ra in plant and sediment at the tested areas showed that one can expect only less than 0.01% of total amount of radium will be extracted during one vegetation season (Michalik et al. 2009). So it is by far too small for effective application of this method. Moreover, the experiment was done based on sediments in which radium was mainly adsorbed at clay minerals. Other experiments, done on sediments with radium-barium sulphate, a hardly soluble mineral, let one to expect even lower results. So, such approach does not solve the problem at all.

The recommended and usually followed approach to utilisation of sediments that had been gathered in underground workings is to put them into old galleries considered never been used again. From the radiation protection point of view such approach is optimal for safe disposal of sediments from surface settling ponds too. However, from technical point of view a lot of obstacles exist as limited capacity of empty underground spaces or distance from settling pond to the nearest shaft in use.

Finally, an approach based on dilution of radium waste was taken into consideration. In general, dilution of radioactive waste is totally forbidden. But in case of natural radioactivity, which is present in small amounts elsewhere, such approach to decrease derived risk sounds rationally. Especially in case of hard coal mining, where technological process creates fovorable circumstances to do it in economic way. During coal exploitation process, at least, half of mine spoil is waste rock and gangue. The radium activity concentration in thse waste do not differ significantly from average value taken as background for earth crust. So that it would be a good “solvent” for radioactive sediment. Actually, mechanical mixing of huge amount of mineral waste is complicated and expensive but again, coal exploitation process provides an opportunity to do it as a “by the way”. Namely, all excavated matter must pass through a coal treatment plant in order to clean coal from gangue and prepare expected fraction of coal by flotation. The total amount of flotation and coal cleaning waste is big enough to turn back radium activity concentration in sediments to background level after homogenous mixing.

The possibility to apply such approach was tested in a mine. The total amount of mine spoil: coal and all types of created waste were balanced against total amount of radioactive sediments gathered in water galleries at all mine levels. It have been done year-by-year since 2005 (table 1). Codes were given based on rules of European waste catalogue (Michalik 2009).

Table 1. The balance of the excavation process

year	Mine productivity (t):					
	total	coal	Waste from coal cleaning process code: 010102	Waste from flotation code: 010481	Sediment from water galleries code: 190899	Sum of : 010202 and 010481
2005	6895262	3674000	3050585	170677	1740	3221262
2006	6760336	3703900	2894547	161889	1620	3056436
2007	7056571	3737600	3163933	155038	3668	3318971

The current and archive data concerning radium activity concentration in every kind of considered materials were used (table 2). Because there are no data about behaviour of such sediment during coal enrichment process, all possibilities were taken into account, it means: total amount of radium accumulated in particular kind of waste have been considered separately (table 3).

Table 2. Basic statistics of radioactivity in sediments and gangue

	code: 190899		code: 010202 & 010481	
	Ra-226	Ra-228	Ra-226	Ra-228
	Bq/kg			
average	705,6	364,4	79,7	73,1
median	409,0	246,0	73,0	79,0
Minimum	21,2	19,0	24	10
Maximum	8272,0	2880,0	189	112
Number of samples	39		15	

Table 3. Radium activity concentration in mixed waste

nuclide	year	190899 + 010202 + 010481		190899 + 010102		190899 + 010481	
		maximum	*average*	*maximum*	*average*	*maximum*	*average*
Ra-226 [Bq/kg]	2005	193,36	80,07	193,61	80,09	270,57	86,05
	2006	193,28	80,06	193,52	80,08	269,08	85,93
	2007	197,92	80,42	198,36	80,45	375,81	94,19
Ra-228 [Bq/kg]	2005	113,49	73,22	113,58	73,23	139,93	76,01
	2006	113,47	73,22	113,55	73,23	139,42	75,95
	2007	115,06	73,39	115,21	73,40	175,97	79,80

Obtained results seem promising. The average activity concentrations in all end-products of the process are slightly increased in comparison to their original value. Moreover, considering the worst case scenario, it means the total amount of radium remains in the flotation waste, the smallest contributor to the total mass of concern, the activity concentration does not differ significantly from other ones and there are no limitations in their disposal at surface mine spoil bank and further use as i.e. aggregate.

Conclusion

In the light of different approaches to remediation of areas affected by waste with enhanced concentration of natural radionuclides, the dilution method with inert material or waste originating from industry of concern seems to be well justified from technical and economical point of view. The example from coal mining industry shows, based on the balance of waste rock and gangue produced by every mine, that there are enough capabilities to use this technology for safe disposals of radium-rich sediments that had been gathered in surface settling pond due to either former or current mining activity. However, such approach needs to be approved by appropriate regulation.

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