



Review

Presence of radionuclides in sludge from conventional drinking water treatment plants. A review

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ABSTRACT

The analysis of sludge samples generated during water treatment processes show that different radioisotopes of uranium, thorium and radium, among others can accumulate in that kind of samples, even the good removal rates obtained in the aqueous phase (by comparison of influent and effluent water concentrations). In consequence, drinking water treatment plants are included in the group of Naturally Occurring Radioactive Material (NORM) industries. The accumulation of radionuclides can be a serious problem especially when this sludge is going to be reused, so more exhaustive information is required to prevent the possible radiological impact of these samples in the environment and also on the people.

The main aim of this review is to outline the current situation regarding the different studies reported in the literature up to date focused on the analysis of the radiological content of these sludge samples from drinking water treatment plants. In this sense, special attention is given to the recent approaches for their determination. Another important aim is to discuss about the final disposal of these samples and in this regard, sludge reuse (including for example direct agricultural application or also as building materials) are together with landfilling the main reported strategies.

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1. Introduction

The radiological characteristics of the sludge samples generated in drinking water treatment plants (DWTP) are dependent on different factors as the source water type, and in this sense the geology and also the industrial activities of the areas where these plants are located are key parameters. Another important factor which can influence is the treatment that is applied over water samples (coagulation, flocculation, decantation and sand filtration). In the literature, there are different studies focused in the radiological characterisation of this kind of samples and this is of special importance since in most cases a common practice in DWTP facilities is the landfilling disposal of the sludge generated during the process, and in this case the health of the workers against the dangers arising from ionizing radiation has to be controlled (Lytle et al., 2014). However, in the last decades, an increasing trend is the reuse of the sludge by means of different alternative strategies as for agricultural purposes or as for building materials, but also in

this case it is extremely important to know the radiological content of these samples to minimize the associated risks.

DWTP facilities normally collect superficial water or ground-water to produce water that is suitable for human consumption. The raw water follow a fairly standard sequence of processes, which essentially consists in solids separation using physical processes as filtration, and chemical processes such as coagulation and disinfection. As a consequence of this treatment, the different pollutants present in the raw water as radionuclides can concentrate in the sludge generated. For this reason, this industry is included in the group called as naturally occurring radioactive materials (NORM) industries.

The presence of NORM can lead to radiation doses that not are insignificant from a radiation protection point of view. For this, within the European Union, Council Directive 96/29/Euratom paid important attention to natural sources of radiation. From that moment, EU Member States are obliged to identify work activities that can involve a radiological risk and for that, most of them have implemented regulations devoted to natural sources of radiation in their own national legislations. So since 1996 the European Commission has moved ahead publishing on regular basis, technical support guidance and recommendations on NORM issues. In this

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sense, in 2001 the European Commission published recommendations dealing with exemption and clearance levels for NORM residues. All these recommendations have provided member states with criteria and a technical framework to help in the establishment of national regulations for NORM (European Commission, 2001). In particular the clearance levels established for natural radionuclides from the ^{238}U was 1000 Bq/kg, the same activity concentration for natural radionuclides for the ^{228}Th and 10,000 Bq/kg for ^{40}K . However, in order to harmonize, promote and consolidate the recommendation established in the Radiation protection 122, the European Union established in 2013 a new Council Directive laying down basic safety standards (BSS) for the protection against the danger arising from exposure to natural sources of ionizing radiation, in this EU-BSS the clearance level for natural radionuclides from ^{238}U and the ^{228}Th established in 1000 Bq/kg (Euratom BSS, 2013).

The aim of this review is to discuss from the radiological point of view, different studies focused on the characterization of the sludge generated in DWTPs, giving special emphasis on the recent studies about the new methodologies used for the determination of the main radionuclides which can be present in this kind of samples. Finally, we also point out the main strategies followed for the disposal of this sludge, and in particular we highlight the recent trend followed in its reuse and the main consequences that can be derived for that.

2. Influence of the treatment in drinking water treatment plants

As we have mentioned before in the Introduction section, DWTP facilities treat raw water to obtain water with a high quality to fulfil the legal requirements for water intended to human consumption. There are different studies focused in the determination of different radioisotopes in water samples from DWTP, in particular the ingoing (raw water) and outgoing (drinking water) water samples from this kind of plants, being the activity concentration found in the analysed samples in the levels of mBq/L (Ajayi and Owolabi, 2008; Baeza et al., 2012; De Oliveira et al., 2001; Jankovic et al., 2012; Palomo et al., 2010a; Walsh et al., 2014; Warwick and Croudace, 2013). For example, it is worth to mention the study of Palomo et al. (2010a) who focused their study in a DWTP located in the south of Catalonia (Spain) during a period of six years. In particular, the authors evaluated the capacity of this plant to remove several natural and anthropogenic radionuclides. For that purpose, first gross alpha, gross beta, residual gross beta and tritium activities were determined in the ingoing and outgoing water samples. Among all the parameters studied, gross alpha activity was the most critical because the values obtained were near to the gross alpha normative limit in Spain (0.1 Bq/L) (Spanish Government, 2003). The authors also evaluated the removal capacity of the plant and they confirm that the elimination rate was around 20% and 5% for alpha and beta emitters, respectively, and they related these results with the treatment followed in the plant that uses FeCl_3 as coagulant, and this favours the actinide elimination as has been previously reported (Baeza et al., 2006; Gáfvert et al., 2002). Moreover, in this study, in the sludge samples generated some gamma emitters could be quantified: natural (as for example ^{40}K or ^{214}Pb) and artificial (^{60}Co , ^{137}Cs and $^{110\text{m}}\text{Ag}$), these last ones could be attributed to the presence of nuclear power plant (NPP) located upstream of the DWTP on the Ebro River. From all the results obtained for the six years monitoring period the authors concluded that sludge samples can concentrate the radioisotopes which can be present in the raw water and that the major contribution to the radioactivity in the sludge samples was from natural sources (97% of total activity) whereas the contribution of artificial radionuclides was only around 3%.

In different studies special emphasis is given to the relationship between the removal of some isotopes which can be present in the ingoing water samples of DWTP with the used treatment: for example Gáfvert et al. (2002) and Baeza et al. (2006) demonstrated that the activity due to some radionuclides from uranium and thorium decay chains which are associated with particles are removed from the water by sedimentation after the addition of an iron coagulant. As an example, Baeza et al. (2006) using a pilot plant tested the effect of four different coagulants (two iron based and two aluminium based) to remove uranium and radium from a natural water. In that study it was also found that apart of the influence of the coagulant, the removal of the different evaluated isotopes was also very sensitive to the pH value. In that sense, the authors demonstrated that the best efficiency for uranium removal was obtained in all cases at pH 6, independently of the coagulant used, and the removal for uranium was around 80% for all the tested coagulants. On the other hand, in the case of radium, the highest removal for this isotope was obtained at basic pH values, but in this case, the use of different coagulants has a different influence over the removal of radium. For the iron coagulants the removal rate for radium was around 50% whereas for aluminium coagulants it was lower with values below 30%.

More recently, Baeza et al. (2012) reported another study in which the optimum treatment conditions (iron coagulant and pH) found in their previous study using a pilot plant (Baeza et al., 2006) were evaluated for a conventional DWTP located in Extremadura (South West of Spain). The authors conclude that by changing the working conditions of the plant, the gross alpha activity was reduced from 0.155 to 0.058 Bq/L and in the case of uranium the removal was in the range 53–83%. The differences observed from this study and the previously performed with a pilot plant in which they obtain higher removal rates for uranium was justified through the greater contact time between the water and the flocs in the conventional DWTP. In the case of radium, due to the low content of this isotope in the treated groundwater, the elimination rate could not be calculated.

Huikuri and Salonen (2000) also focused in the removal capacity of uranium. In particular they focused their study in drinking water treatment plants from Finland. In this case the authors evaluated the use of commercial ion exchange filters based on strong base anions and the removal of uranium was found to be over 95% using this resin, thus demonstrating the good performance of this kind of treatment in the removal of radioisotopes. In this study, water samples of different physicochemical characteristics were analysed and for all of them the results were similar.

In recent years, membrane technology has become a valuable tool in water treatment in order to produce a drinking water with a high quality. Even that this field has not been very exploited in the removal of radionuclides, there are recent studies in the literature that are focused in testing the use of these membranes, and in particular, in the use of reverse osmosis membranes by means of pilot plants (Montaña et al., 2013b; Nieto et al., 2013; Pérez-González et al., 2012). In these studies the authors demonstrated the benefits of these new technology in the removal of alpha and beta emitting isotopes, by measuring gross alpha and gross beta activities obtaining an average removal of around 90% in the case of alpha isotopes, whereas for gross beta the obtained results for both published papers were quite different and ranged between 35% for Nieto et al. (2013) and 93% for Montaña et al. (2013b).

Apart of these studies mainly focused in the removal for different natural isotopes by following different treatments, other studies evaluate the removal for artificial radionuclides. For example, Gáfvert et al. (2002) evaluate the use of different coagulant reagents (iron or aluminium coagulants) to remove different natural and artificial radionuclides of uranium, thorium, plutonium,

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