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The presence of radionuclides in wastewater treatment plants in Spain and their effect on human health

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ABSTRACT

Wastewater treatment facilities can produce sludge containing measurable amounts of radioactivity. The radionuclides that are concentrated may originate from radionuclides present in the original source of water or from water treatment. This treated water ends up most frequently in the aquatic environment, but it may also receive further application, for instance, as irrigation water in agriculture. Therefore, evaluation of its quality is essential to both protect the health of the environment and also of humans.

The aim of the present work is to study the radioactivity in liquids and sludge generated at 11 wastewater treatment plants in Spain, working under a variety of conditions, as there is an increase in usage of wastewater treatment plants in Spain as a result of frequent drought periods.

Gross alpha and gross beta activities were determined in influent and effluent liquids and sludge by ZnS solid scintillation detectors and low-background gas-flow counters. Natural and artificial gamma radionuclides in the sludge were analyzed with a germanium semi-conductor detector.

The results indicate that the radiological characteristics of the effluents and sludge do not present a significant radiological risk and make them suitable for future applications.

1. Introduction

Wastewater treatment is an essential public service which will increase in importance in the future due to population growth, increasingly restrictive environmental regulations, and demand for wastewater re-use.

Currently, the wastewater treatment industry faces a number of challenges, including urban population growth, the need to treat wet weather flows, more stringent discharge regulations, and demand for water conservation through wastewater re-use. The EPA estimates that water and wastewater capacity will need to grow by 5–8% annually over the next decade.

There is also a rapid growth in the re-use of municipal wastewater for irrigation and ground water recharge, which necessitates enhanced treatment to remove nutrients (nitrogen and phosphorus), suspended solids, and other contaminants.

Sewage treatment is the process of removing contaminants from wastewater. It includes physical, chemical and biological processes to remove physical, chemical and biological contaminants. Its objective is to produce a disposable effluent and solid waste or sludge suitable for discharge or re-use back into the environment without causing harm to communities or pollution. A conventional wastewater treatment plant (WWTP) provides pre-treatment, primary and secondary treatment and sometimes a tertiary treatment. Pre-treatment removes materials that can be easily collected from the raw wastewater before it damages or clogs the pumps and skimmers of primary treatment clarifiers. Primary treatment involves physical settling of the raw wastewater and secondary treatment consists of a biological process that is followed by physical settling. The purpose of tertiary treatment is to provide a final treatment stage to raise the effluent quality using disinfection before it is discharged to the receiving environment. Common methods of disinfection include ozone, chlorine, ultraviolet light, or sodium hypochlorite.

Sludge processing is complex and can consist of a variety of operations including: sludge thickening, sludge stabilization by lime addition or digestion (either aerobic or anaerobic), sludge de-watering, and, ultimately, disposal by landfill, composting, land application, or incineration. In most plants, primary and secondary sludge are combined, thickened by sedimentation or flotation, stabilized, and de-watered by the use of a belt filter press or centrifuge.

In Spain 90% of the wastewater generated is treated in wastewater treatment plants and hydrological planning foresees the

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construction of numerous new plants by 2014 to provide service to all towns with fewer than 2000 inhabitants. This treated water ends up most frequently within the aquatic environment (river or sea), but it may also be further used, for instance, as irrigation water in agriculture. Evaluation of its quality is therefore essential to protect human health and the environment.

Levels of naturally occurring radionuclides of U-238 series, the Th-232 series and K-40 are present in soil and water used for drinking and industrial processes. These isotopes may be introduced into the sewerage system from ground and surface water as well as from potential industrial discharges. Another possible source of radioactivity at WWTPs is the authorized release of man-made radionuclides into the system. In most regions, the level of radionuclide contamination in wastewater results from weapons testing, accidents, industrial applications, research institutions and medical uses. The radionuclides most commonly used in biotechnology, hospital and medical facilities are ^{3}H, ^{99}Tc and radio iodines.

On the other hand, ^{7}Be (of a cosmogenic origin) and $^{210}Pb_u$ (radon progeny) are natural radionuclides present in surface airborne particulate matter and are subject to wet and dry deposition onto the ground.

Wastewater is transported to the sewage plant where solids and pollutants are removed using a simultaneous biological-chemical precipitation method. Radioactive elements represent a special case of inorganic pollutants and they show similar environmental behaviour to their stable chemical isotopes. Radionuclides released into the sewage systems may be concentrated in digested sludge or released to the recipient surface waters. It has been shown previously that sewage sludge is a very sensitive indicator for radioactive materials released from hospitals and an indicator of radionuclides in general (Sundell-Bergman et al., 2008). During the wastewater treatment process, radionuclides are concentrated in the sewage sludge in a similar way to solids and toxic metals.

Direct discharge of radioactive material into the sewerage system in Spain is regulated by the Spanish Royal Decree 783/2001 (BOE, 2001). This law "Regulation on Health Protection against Ionizing Radiation" was approved as a result of the European Council Directive 96/29 (EURATOM, 1996).

It should be pointed out that the European Union has no regulations on the disposal of sludge samples generated in WWTPs, but this industry has been considered in the European Council Directive 96/29. This Directive applies to facilities such as installations with naturally occurring radioactive materials where human activity has increased the potential for exposure (NORM industries) (IAEA, 2003).

In Spain, NORM industries have come under subjected to regulation (transfer of European Council Directive 96/29) (EURATOM, 1996) in the publication of the Spanish Royal Decree 1439/2010 (BOE, 2010).

The regulation deals with "Natural Sources of Radiation" and the need to study activities in which workers or members of the public could be exposed to significant doses of radiation (EUR, 2002).

In the literature there are few interesting studies that evaluate the impact of radioactivity on potable water treatment and its elimination. For instance, there are methods to remove uranium, radium (Sorg, 1990; Baeza et al., 2006), cesium and strontium (Smith et al., 2001) and the studies by Gäfvert et al. (2002) and Jiménez and De la Montaña Rufo (2002) show the differing behavior of both naturally occurring and anthropogenic nuclides in the purification process at drinking water treatment plants. However, the number of wastewater studies that can be found in the literature is limited. Stetar et al. (1993) and Ipek et al. (2004) studied the removal of some artificial radionuclides in secondary treatment and radioactivity behavior in biological treatment. These studies have shown that radioactivity in wastewater is reduced by biological treatment and the authors conclude that radioactivity removal could occur by adsorption to the activated sludge. Rodriguez and collaborators (Rodriguez et al., 2009) recently completed a study which analyzed the removal efficiency of reverse osmosis in wastewater. The results indicate that reverse osmosis is able to reduce the concentration of gross alpha and gross beta activity and produce high quality water.

On the other hand, the occurrence of radionuclides in sludge samples for different WWTPs has been studied by some authors to evaluate their potential danger. Initially, the studies were focused on artificial radionuclides as a result of the Chernobyl accident and nuclear tests. A Swedish study by Erlandsson et al. (1983) analyzed radionuclides from global fallout and local sources in ground level air and sewage sludge. Imhoff et al. (1988) sampled sewage sludge from 10 WWTPs in Ruhrverband (Germany). The results indicate that a sudden increase of radioactivity was observed with an initially high fraction of I-131 due to the Chernobyl accident and the presence of long-life fission products.

Research was later focused on other potential sources such as medical and industrial waste and the water treatment process. Note that some of these sources may contain both natural and artificial radionuclides. Miller and collaborators analyzed sludge samples from 25 municipal WWTPs to determine if sewage effluents from nuclear facilities had levels of radioactivity above those expected from the environment and the sludge samples had maximum values of 2 Bq/Kg of Cs-137 and 3 Bq/Kg of Co-60 (Miller et al., 1996). Puhakainen (1998) found gamma activities between 24 and 250 Bq/Kg in sludge samples from a WWTP in Finland and detected their origin due to medical applications and industrial processes. Martin and Fenner (1997) measured radioactivity levels in sludge at one WWTP next to a medical complex which carries out radioiodine treatments with the aim of determining concentrations of I-131 and estimating radiation doses to employees and the public. A similar investigation it has been recently carried out by Jiménez and colleagues (Jiménez et al., 2010).

For instance, in the U.S., groundwater is generally used in water treatment processes to produce drinking water, and consequently, EPA has already published several technical reports on this topic. One of them summarizes data on the radioactive constituents and levels found in sewage sludge with the aim of evaluating any potential health concerns in the future together with recommendations on management of radioactive materials in sewage sludge (Bastian et al., 2005).

Within this context, the present work investigates the occurrence of radioactivity both in the inlet and outlet waters and sludge at 11 conventional municipal WWTPs in Spain with the objective of studying their levels of radioactivity in the resulting treated water and sludge, their removal in the treatment plant and the radiological risk for workers or members of the public.

2. Material and methods

2.1. Sample collection

A total of 22 water samples from the influent and effluent and 11 sewage sludge samples from 11 conventional municipal WWTPs in Spain were collected.

The plants were located in two different areas of Spain, in the north east (zone 1) and the north west of Spain (zone 2) and the selection was carried out by taking into account the fact that plants were in different geological areas and that they used different treatment processes. The wastewater included domestic water, fruit and vegetable washing water, hospital effluent, other varieties of wastewater and rainwater.

The sewage sludge samples correspond to a mix of primary and secondary sewage, which was anaerobically digested and then dehydrated using press filters. The conventional treatment for