Radiological environmental impact of the phosphate industry in Spain: a regulatory perspective

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INTRODUCTION

The phosphate industry is regarded as strategic and therefore it is present in almost all countries around the world. Its production is mainly targeted to the manufacture of fertilizers and, in less extent, in a variety of products including animal feed supplements and laundry detergents. Owing to the nature and the large quantities of raw materials and intermediate products involved, the whole chain of processing, storage and transport is liable to cause adverse environmental impacts. Atmospheric emissions, waste waters and solid wastes contain potentially hazardous substances, such as fluorine, heavy metals or natural radionuclides, at concentrations highly dependent on the particular process and on the origin of raw material.

There are currently two phosphoric acid production plants in Spain, holding an overall capacity of 550 kilotonnes P_2O_5. They are both located in Huelva (SW Spain) and apply the wet production route through digestion of the phosphate rock by sulphuric acid. This process generates as a co-product large amounts of gypsum (around 4-5 tonnes for every tonne of P_2O_5) which are stored in nearby stacks. Phosphogypsum (PG) management is an environmental challenge because of the large volumes of PG produced and the type and level of impurities it contains. Few more plants manufacture compound fertilizers by the mixed acid route, but this type of industries generally produce little solid waste and limited effluents.

No specific radiological constraints on effluent monitoring and release or on waste disposal have been yet imposed on NORM industries in Spain. However, other environmental regulations, and particularly Council Directive 96/61/EC concerning integrated pollution prevention and control (IPPC), have resulted in a substantial reduction on the phosphate industry emissions. Nonetheless, more than a century of industrial exploitation in Spain has left a heavy radiological legacy. Although nowadays all plants in the EU practice land disposal, in the past, discharges to the sea or to nearby watercourses were allowed, polluting reservoirs, estuaries and coastal waters. Absent or malfunctioning isolation of stockpiles in former active plants or at abandoned sites may as well have resulted in soil or water contamination. Three examples of environmental contamination, presently being abated, are given: (i) the Huelva estuary; (ii) the Flix reservoir at the Ebro basin, and; (iii) industrial soil near Cartagena.

ENVIRONMENTAL POLICY AFFECTING THE SPANISH INDUSTRY

The IPPC Directive introduced a comprehensive approach to industrial regulation in the EU. Under the terms of the Directive, Member States[1] must ensure that permits for the concerned industrial processes – which installations must obtain and comply with to be allowed to operate – include emission limit values based on the use of the best available technologies

[1] In Spain, this obligation has been further delegated to regional authorities (the Autonomous Communities).

(BAT). But the Directive relies on a combined approach, and requires, additionally, that permits do not lead to breaches of the environmental quality standards established in other legal instruments, as the Ambient Air Quality Directive (96/62) or the Water Framework Directive (2000/60). It also supports the achievement of the Waste Framework Directive (2006/12) and of the future Soil Framework Directive by requiring waste avoidance and preventing or minimising emissions to soil. List of hazardous substances in this legislation do not explicitly include radionuclides, although all carcinogens are generically concerned.

On the other hand, radioactivity issues of NORM practices are governed by Title VII of the Basic Safety Standards Directive. This title applies to "*work activities [...] within which the presence of natural radiation sources leads to a significant increase in the exposure of workers or of members of the public which cannot be disregarded from the radiation protection point of view*". No quantitative standards are specified therein, but further related regulatory guidance has been issued in other documents. In particular, for members of the public, doses due to existing or past NORM practices must be as low as reasonably achievable below a dose limit of 300 μSv y^{-1}.

All those legal instruments contribute to achieving the objectives of relevant international agreements (see Fig. 1). In particular, Spain has signed the two international Conventions on marine pollution concerning its coastal waters. These are: the OSPAR Convention, covering the north-east Atlantic north of Gibraltar, and west to Greenland and the mid-Atlantic Ridge; and the Barcelona Convention covering the Mediterranean Sea. Both imply the commitment by the Parties to phase out discharges, emissions and losses of hazardous substances deriving from land-based sources and activities (Sintra Statement, 1998; Syracuse Protocol, 1996).

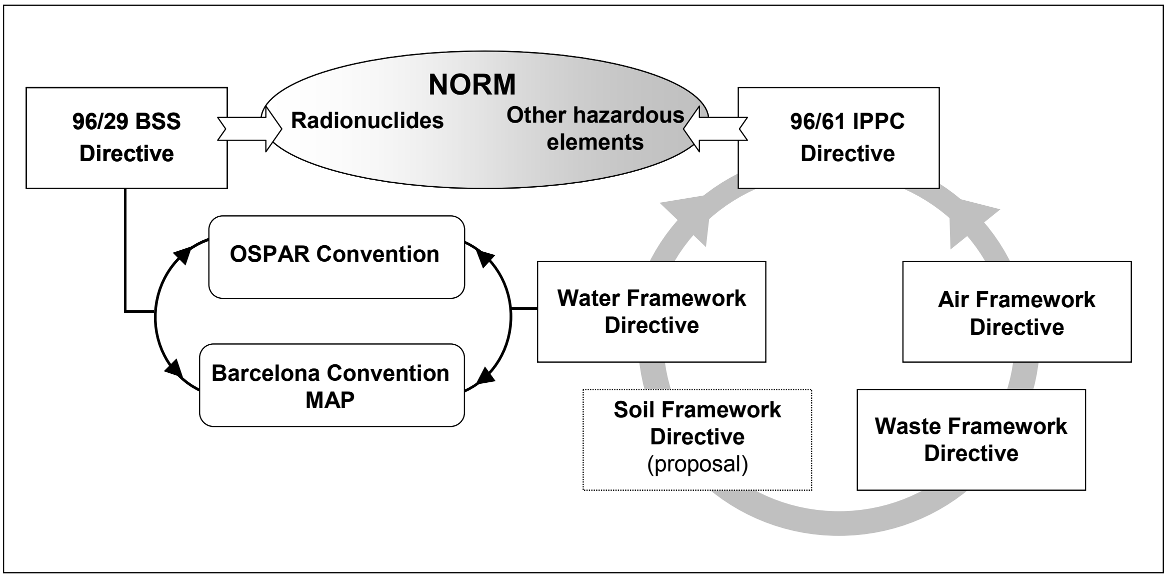

Figure 1: Interaction of selected legal instruments relevant to the phosphate industry.

PRESENT AND PAST ENVIRONMENTAL IMPACT

Radioactive pollution due to phosphate production or processing facilities currently in operation in Spain is negligible. Since liquid or solid wastes releases to the environment are prevented, particulate matter discharged into the atmosphere is the most important additional source of radioactivity. Data on annual particulate matter emissions are available from the EPER-Spain register. Assuming this matter has the same average radioactivity content as fertilizers, it can be verified that the impact is neither of radiological concern. Historic environmental pollution of the industry, on the contrary, remains at a number of places. The clean up of historic contamination is controlled by the regional authorities, to whom CSN has act as an advisor for radiological issues.

The Huelva estuary

The Huelva estuary, formed by the confluence of the Tinto and Odiel rivers, has been historically affected by industrial discharges from nearby factories, including two phosphoric acid production plants. An acid waste stream and 20% of the PG were dumped into the aquatic environment while the remaining 80% was disposed in stacks over salt marshes of the estuary. But since discharges were forbidden in 1997, all wastes are stored in the stacks and a closed circulation system for fluids has been established.

In the H_2SO_4 route, about 90% of the ^{226}Ra and the ^{210}Po and some 15% of the uranium isotopes from the ore are left in the PG (Bolivar et *al.*, 1996). The radioactive impact in the area has been uninterruptedly monitored since 1988. Radionuclide concentrations in the estuary are decreasing since the cessation of discharges to the aquatic environment. Radioactivity in water and surface sediments has returned to background levels except for the Odiel river, where activity in sediments close to the plant still remain in 2002 over one order of magnitude above the natural background levels of the area. (Villa et *al.*, 2004). Although different studies have shown that the radiological impact to public due to the stacks is not significant (Cancio et *al.*, 1998), they are being restored by a cover of clay, to reduce emanation, an overlying layer of soil and vegetation.

The Flix reservoir

Wastes from a dicalcium phosphate (DCP) plant were released to the Flix reservoir waters until 1986. The plant is associated to a chloro-alkali factory and uses the excess hydrochloric acid for the production of DCP. Although the chemical reaction is similar, the waste products from the HCl and the H_2SO_4 processes are quite different. The sludge deposits contain the major fractions of U and Th, while ^{226}Ra remains in solution and enters into the surface water as soluble compound ($^{226}RaCl_2$).

In 2003, very high levels of organochlorines, heavy metals and radionuclides were detected in sediments of the reservoir. No clear signal of NORM contamination in downstream riverbed sediments have been found but dissolved ^{226}Ra concentrations 6 times higher than background have been reported in river waters 1 km downstream. (Costa et *al.*, 2004) The remedial option selected is *ex-situ* treatment of more than 800.000 tons of contaminated sediments. They will be removed by dredging, treated in a plant built next to the extraction zone and transported to a controlled landfill.

Cartagena industrial area

Environmental contamination of this area is of minor concern compared to the previous two examples, but it has been selected for being representative of industrial land reclamation. The terrains of a former fertilizer factory (previously dedicated to the manufacture of explosives) are going to be restored due to ongoing Cartagena town expansion. Soils are contaminated not only by natural radionuclides but also by Pb, Zn, Cd and As, coming from phosphate production wastes and from pyrite ashes. Groundwater (an aquitard system) is affected, but at pollutant concentrations below the regulated limits.

Separate criteria have been set for radioactivity and heavy metals in order to grant authorization for residential use (see García-Talavera et *al.*, 2007). Remedial works to fulfill those criteria will imply the removal of wastes and contaminated soil that will be transported to a landfill.

CONCLUSIONS

The radiological environmental impact of phosphate production or processing facilities currently in operation in Spain is not significant. Areas affected by historic contamination are being clean up under the control of regional authorities, advised by CSN on radiological issues. Nevertheless, although Spain is moving towards a higher level of environmental protections, radioactivity and chemical hazards are considered separately (in the legal and practical sense), and a holistic framework needs to be developed. Selecting a remedial alternative for NORM contaminated sites requires evaluating the resulting ecological and human health benefits and risks. BAT and ALARA decision approaches are fundamentally similar, but different remedial options may be preferable regarding chemical and radioactive risks. Public risk perception is also a key factor in the decision process and, thus, it is essential to effectively communicate to the stakeholders the real benefits and risks of every remedial option.

REFERENCES

Bolivar, J.P., García-Tenorio, R., García-León, M., 1996. On the fractionation of natural radioactivity in the production of phosphoric acid by the wet acid method. J Radioan Nucl Chem, 214:77-88.

Cancio, D., Gutiérrez, J., Sáez, J.C., Palomares, J., 1998. Revisión de la situación radiológica en la zona de vertidos de la industria de fosfatos en Huelva. Informe del Ciemat para el CSN (in Spanish).

Costa, E.; Sanchez-Cabeza, J.; Garcia, J.; Masque, P., Grimalt, J., 2004. Environmental radiological impact by a fertilizer complex in the Ebro river (Spain). NORM IV Conference, Poland.

García-Talavera, M., Martínez, M., Martín-Matarranz, J.L., Salas, R. Serrano, I., Sanz, M.T., Ramos, L., 2007. Setting radon specific release criteria and demonstrating compliance for land affected by NORM. International Conference on Environmental Radioactivity: From Measurements and Assessments to Regulation. Viena.

Villa, M., Absi, A., Moreno, H., Garcia-Tenorio, R., Manjón, G, 2004. Natural Restoration of a Spanish Estuary Historically Affected By Anthropogenic Inputs of Natural Radionuclides. Norm IV Conference, Poland.