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Radioactive Characterization of Leachates and Efflorescences in the Neighboring Areas of a Phosphogypsum Disposal Site as a Preliminary Step before its Restoration



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INTRODUCTION



After the recent closing of some phosphoric acid plants located in the South-West of Spain, it has been decided to restore a big extension (more than sixty hectares) of salt-marshes where historically it has been disposed some million tonnes of phosphogypsum.

This PG is characterized for containing high activity concentrations of several radionuclides from the uranium series, mainly ^{226}Ra , ^{210}Pb and ^{210}Po and to a lesser extent U-isotopes.

The PG disposal area can be considered as a potential source of radionuclides to their nearby environment, through the waters which percolates from them and through the efflorescences formed in their surroundings

A detailed radioactive characterization of the mentioned waters and efflorescences has been considered essential for a proper planification of the restoration plan to be applied in the near future in the zone.



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ENVIRONMENTAL IMPACT



They exist both direct and diffuse pollution points due to liquid effluents coming from the PG storage deposit (leachates), and generated by rainwater and the tidal influence.

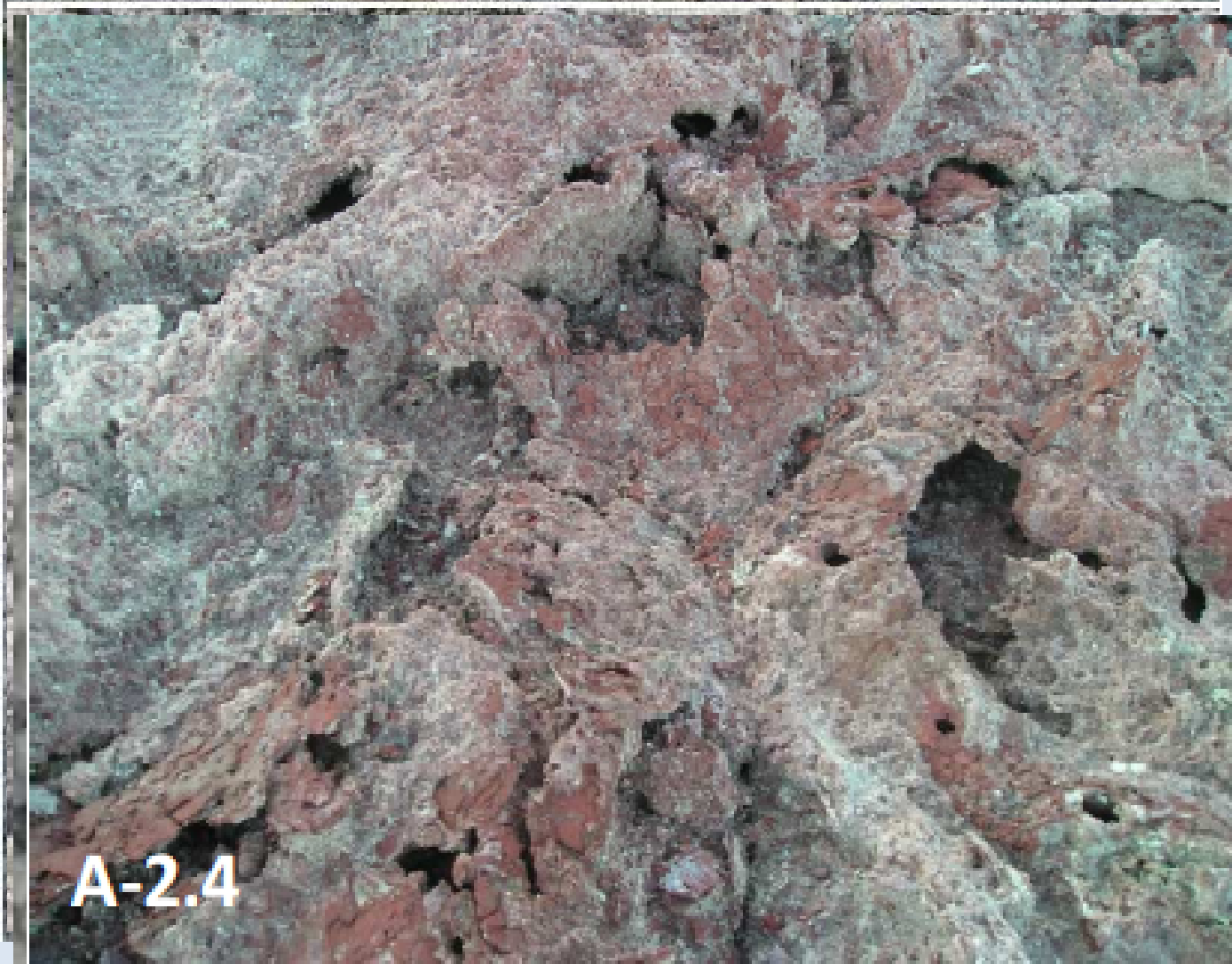
The landfill leachates are also the source of “efflorescences” (crystallized-precipitated salts in the water drainage coming from the phosphogypsum pile). In general, water soluble efflorescent salts are formed by the evaporation process of water during dry seasons, which produces a large variety of metal hydrosulphates

The efflorescent salts sequester acidity, metals and radionuclides temporarily and release them later during rain or melting events



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EFFLORESCENCE SAMPLES





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SAMPLING (I)





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SAMPLING (II)





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RADIOMETRIC TECHNIQUES



- a) Determination in waters and efflorescences of ^{234}U , ^{235}U , ^{238}U , ^{230}Th , ^{210}Po and ^{232}Th by alpha-particle spectrometry with PIPS detectors. Sequential isolation of U, Th and Po using ion-chromatographic resins (UTEVA)



- b) Determination of ^{210}Pb , ^{234}Th , ^{226}Ra and ^{40}K in the efflorescences by low-background gamma-ray spectrometry with Ge detectors.

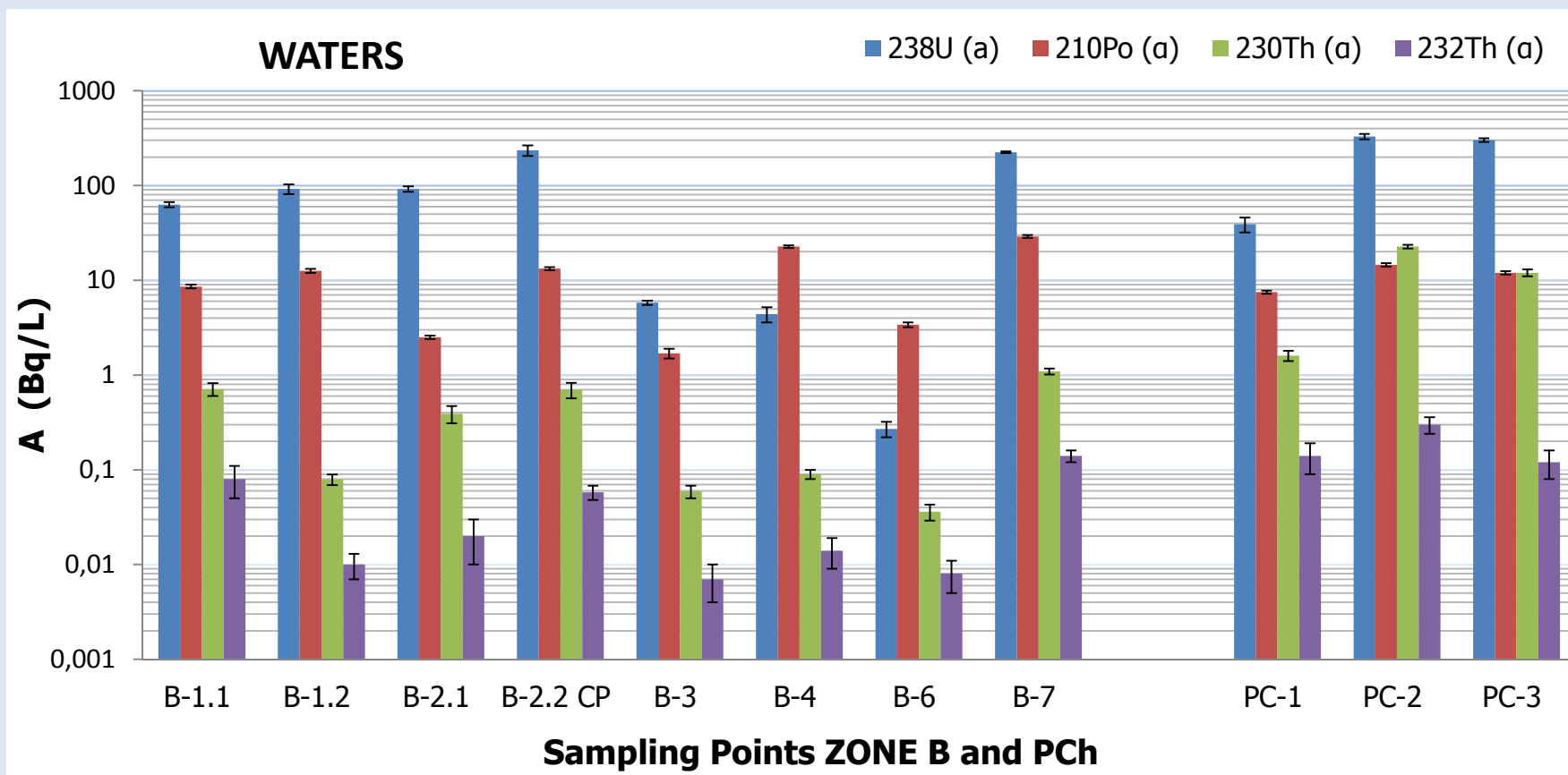


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RESULTS



WATERS : ZONE B including Perimetral Channel



$U > {}^{210}\text{Po} > {}^{230}\text{Th} > {}^{232}\text{Th}$



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RESULTS



WATERS : ZONE B including Perimetral Channel

In the waters $U > {}^{210}\text{Po} > {}^{230}\text{Th} > {}^{232}\text{Th}$

In PG ${}^{210}\text{Po} > {}^{230}\text{Th} > U > {}^{232}\text{Th}$

Concentrations of ${}^{234}\text{U}$ and ${}^{238}\text{U}$ as high as 300 Bq/l

Concentrations of ${}^{210}\text{Po}$ as high as 10 Bq/l

${}^{230}\text{Th}/{}^{232}\text{Th}$ activity ratio = 100

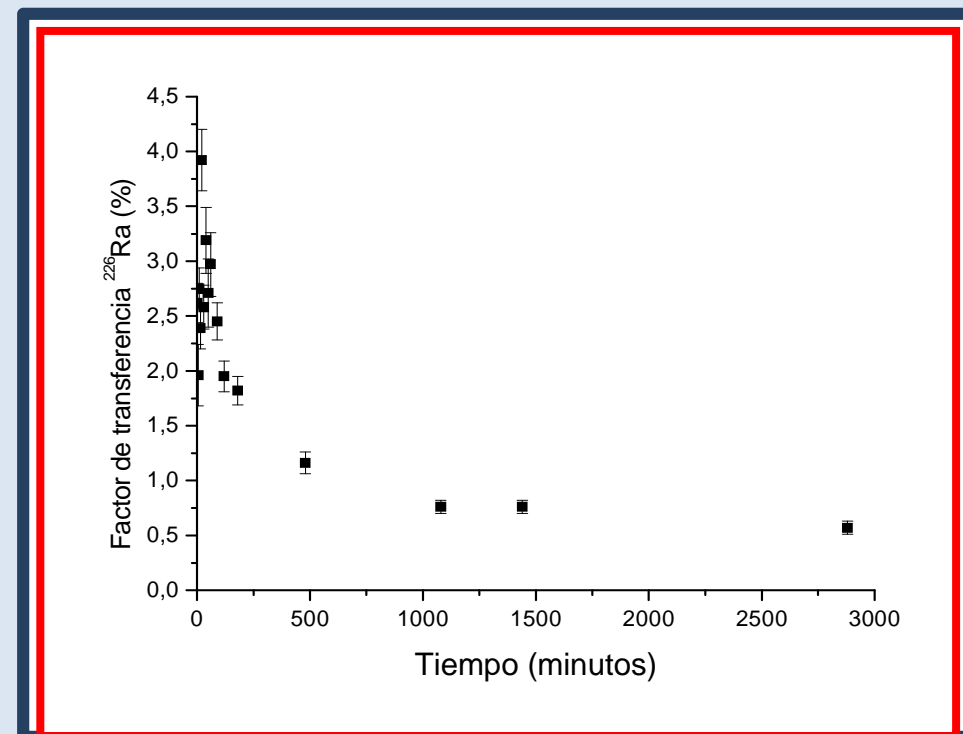
Concentrations of natural radionuclides in the waters
intimately related with their bounding form to the PG grains



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LEACHING EXPERIMENTS

FRESH PG

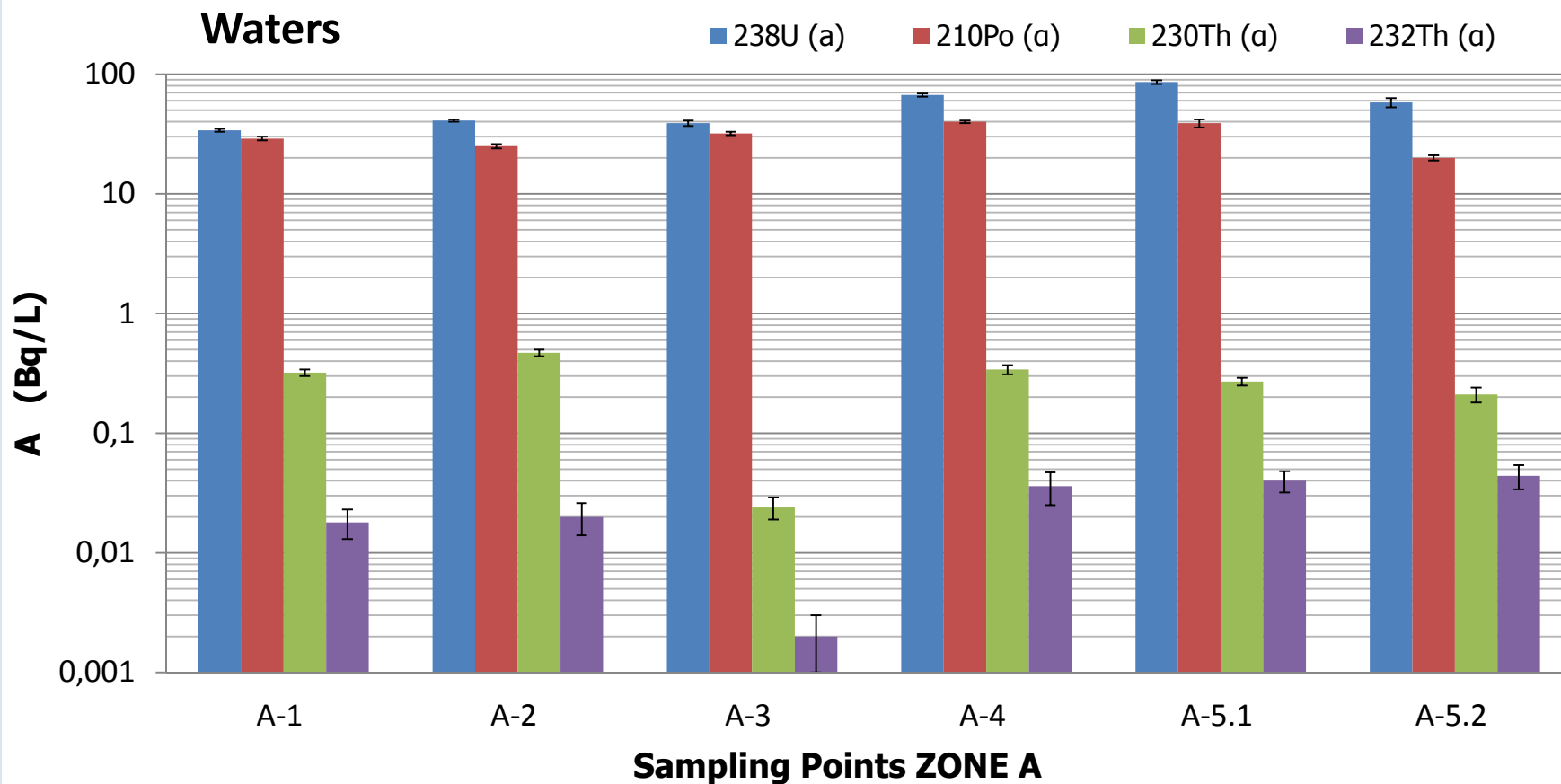




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RESULTS

WATERS : ZONE A



$U > {}^{210}\text{Po} > {}^{230}\text{Th} > {}^{232}\text{Th}$



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RESULTS

WATERS : ZONE A



In the waters $U > {}^{210}\text{Po} > {}^{230}\text{Th} > {}^{232}\text{Th}$

In PG ${}^{210}\text{Po} > {}^{230}\text{Th} > U > {}^{232}\text{Th}$

The U concentrations in waters from this zone are lower than the U concentrations in zone A and PCh \Rightarrow Old PG previously washed in U.

The ${}^{210}\text{Po}$ concentrations in the waters of zone B clearly higher than the ${}^{210}\text{Po}$ concentrations in zone A and PCh \Rightarrow ${}^{210}\text{Po}$ more “available” in the old PG areas, fact that can be related with microbial activity.

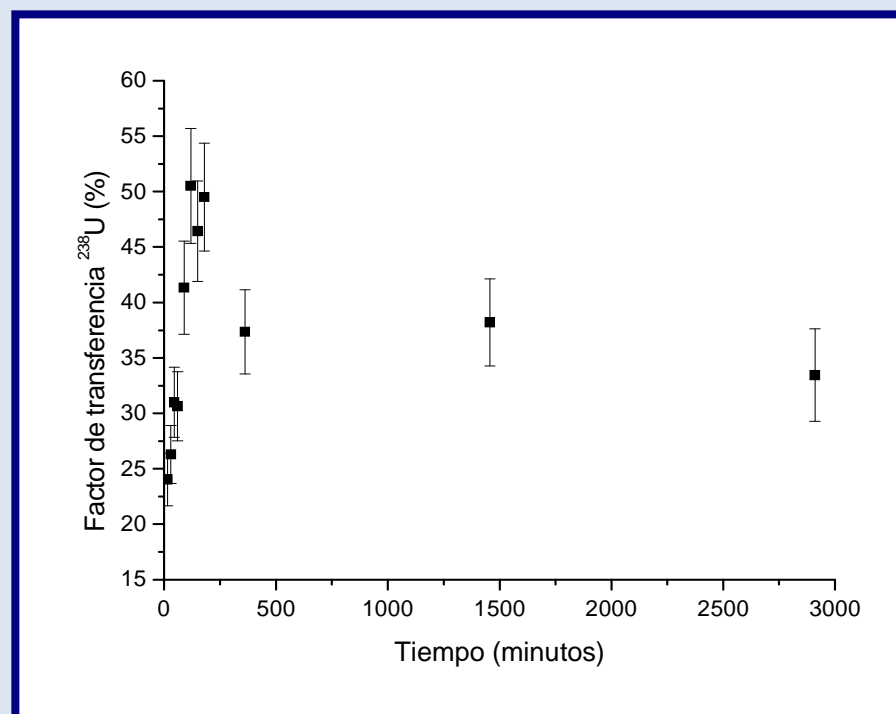
${}^{230}\text{Th}/{}^{232}\text{Th}$ activity ratio = 100



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LEACHING EXPERIMENTS

OLD PG

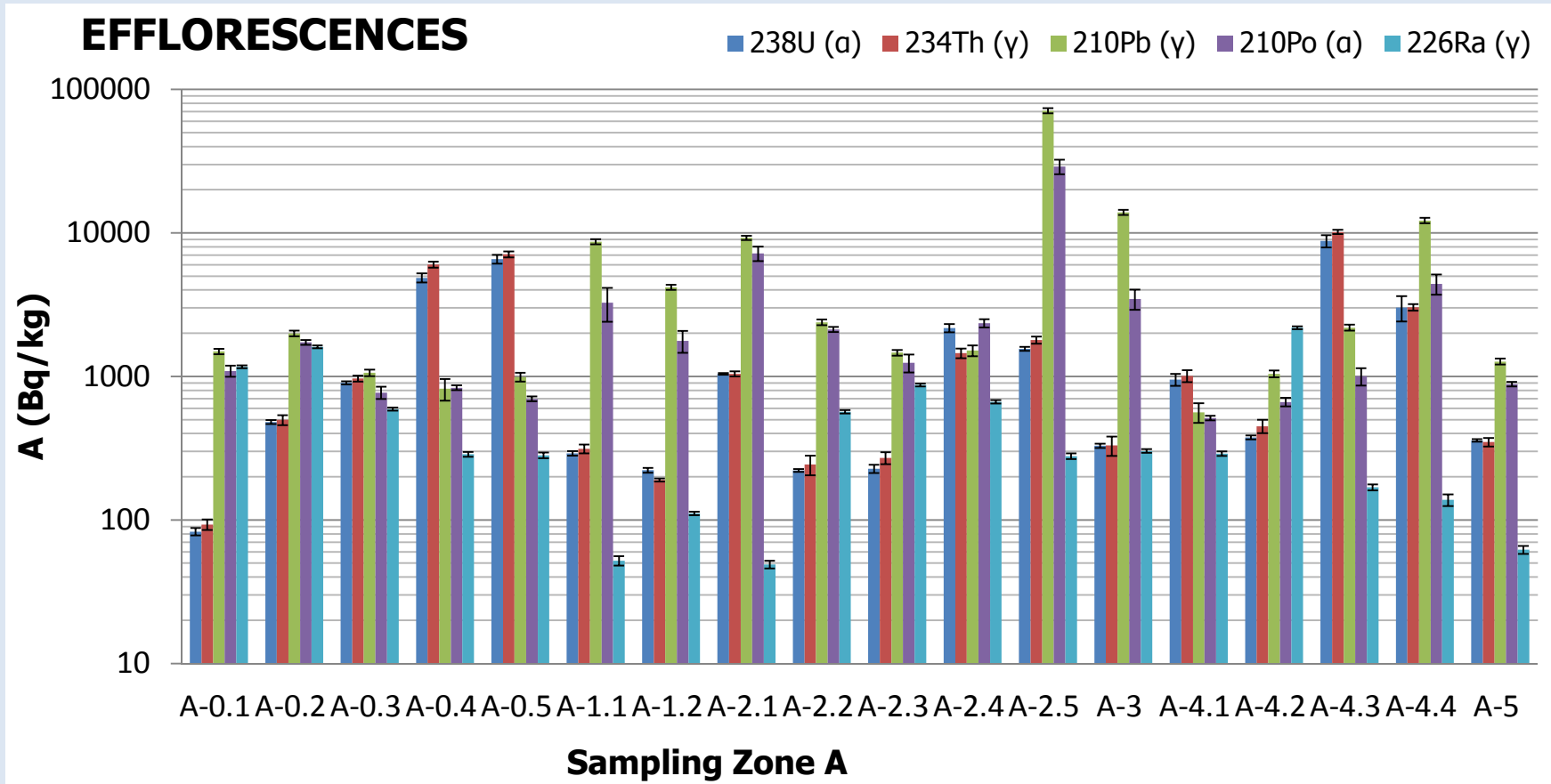




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RESULTS

EFFLORESCENCES ZONE A



TWO TYPES OF EFFLORESCENCES ?

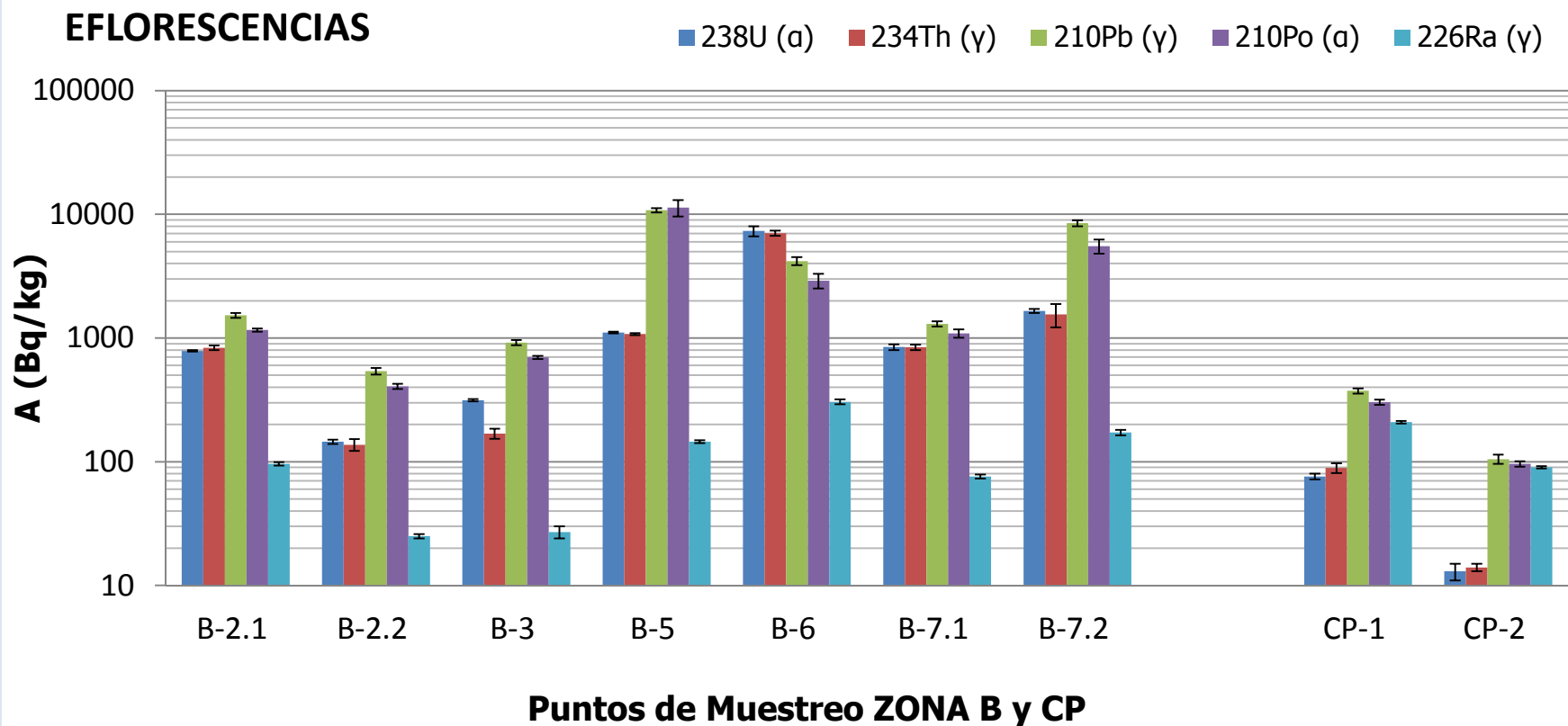


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RESULTS



EFFLORESCENCES ZONE B AND PCH





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RESULTS

EFFLORESCENCES



The radionuclide concentrations in the EF from the perimetral channel are clearly lower than the found ones in the EF collected in the surroundings of Zone A and B. Related with the different way of formation.

The Efflorescences can act as a delayed source of radionuclides, either to the nearby aquatic environment, if they dissolve in the waters that sporadically can cover them, or to the atmosphere if some efflorescence dry material is resuspended by the breezes affecting periodically the estuarine area.

The ^{226}Ra activity concentrations in all the EF are quite moderate in comparison with ^{210}Pb and U-isotopes, with values even lower than the found in the PG samples. These results allow to reject the hypothesis that the efflorescences are mainly formed for the PG grains of smaller grain size.



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ENVIRONMENTAL RADIOACTIVE IMPACT



Hydrological studies performed in the area allow evaluating the annual flow of waters leaking from both PG areas to the surroundings in 80000 m³/year.

This flow is quite low in comparison with the annual flow of the Tinto river, which according to the official hydrological series, has been evaluated in 90·10⁶ m³/year

This imply for example in the case of uranium, assuming its concentration in the leached waters three to four orders of magnitude higher than natural values in the estuary, that in the U input from the piles to the estuary can be in the order of the natural uranium supplied to the estuary.

The moderate radioactive impact of the PG leaked waters in the estuary, has been ratified, by independent experimental studies performed by analyzing the concentrations and fate of different radionuclides from the uranium series in different compartments of the estuary



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CONCLUSIONS



The acidic waters discharging from the phosphogypsum piles to the surrounding environment contains very high concentrations of radionuclides from the uranium series (3-4 orders of magnitude higher than in a sea water) particularly of the U-isotopes

The activity concentrations for the same radionuclides in the efflorescences, cover an ample range, with the activity concentrations of ^{210}Pb and ^{210}Po being in general very high and, in general, higher than other typical radionuclides of phosphogypsum as ^{226}Ra and ^{238}U .



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CONCLUSIONS



These facts clearly highlight the different trend of the radionuclides from the uranium series in the different processes occurring in the area and affecting their behaviour: dissolution in the acidic waters, incorporation to the efflorescences during their formation, etc.

However, the low weight of the outflows from the piles in comparison with the water flow in the estuary and the low density of existing efflorescences imply a moderate radioactive environmental impact in the estuary as a whole.

Thanks a Lot

