

NORM IX

The 9th INTERNATIONAL SYMPOSIUM on NORM

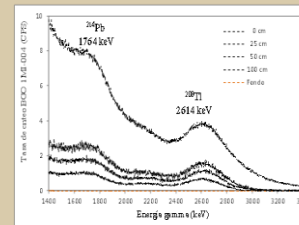


TENORM accumulation in oil tube's radioactive scales at Venezuelan Industry: characterization and proposed solutions.

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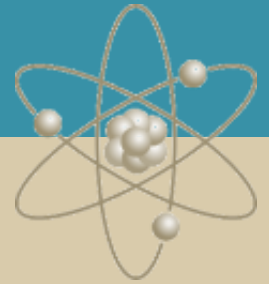


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Introduction and context

At national level, there are several large scale storage places for used oil production tubes and other equipment (in the picture Site 2)



Introduction and context

Each facility have hundreds of thousands tubes and other used equipment to be monitored



Oil production tubes

Oil production since 1914

Oil activity spread over large areas

Several large and medium scale repositories of used oil production tubes and other materials



Objectives



- Identify the presence of NORM in the oil industry in Venezuela
- Characterize the scales found in used oil production tubes and other equipment
- Determine if these are NORM/TENORM
- Assess the external dose for the workers
- Assess if there is risk of contamination

Methodology



- Three large deposits were monitored at national level
- Direct monitoring of dose rates (GM and alpha counters)
- *In situ* gamma spectrometry (BGO)
- Sampling for Lab. Analyses (XRD, SEM, TXRF, HPGe, etc.)



**Highly
Radioactive
Mineral Scales**

Results and discussion

➤ In situ dose rate over the deposit

Average dose rate measured directly on the extreme of the tubes. In the first place 30 thousand of tubes were scanned with a GM, while in the second place more than 100 thousand were monitored. The 3rd place was totally clean of radioactivity.

The external dose rates in most of the working places is as low as the background, so that it is not necessary to restrict access to the area. However, in the presence of contaminated tubes, a prudential distance (1-2 meters) of pipes contaminated with TENORM is required.

In a small proportion (less than 1%, but still hundreds of tubes) there are contaminated tubes, where the presence of high dose rates in their both ends is relevant, even at 30 cm of the tubes dose rates reached values that exceed limit for public. These areas should be marked as restricted until the pipes are cleaned.

Maximum dose rate values reach up to 35 $\mu\text{Sv/h}$ in contact with contaminated tubes

Accepted limits (derived dose rate limits)*

Occupationally Exposed Workers: 10 $\mu\text{Sv/h}$ (2000 working hours per year)

General public: 0,1 $\mu\text{Sv/h}$ (365 days x 24 hours)

*After international recommendations (ICRP / IAEA) and Venezuelan national regulations (COVENIN)

Results and discussion

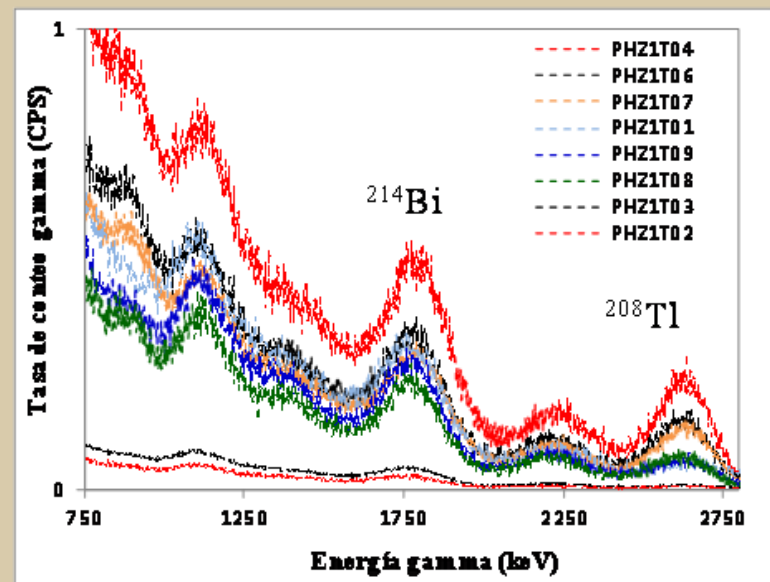
➤ In situ dose rate over the deposit

It is categorically recommended to tightly close all contaminated pipes, to reduce the risks of occupational and environmental pollution that these materials pose with high concentrations of radioactivity.

Alpha particle count rates (several thousand counts per minute in the case of the most contaminated tubes) confirm that radioactive inlays contain natural radioisotopes. This implies that in case of inhalation of fine dust the radiological risk is very high.

Gamma spectra taken in the field (BGO) shows only the emission lines of the natural sub-chains of ^{226}Ra and ^{228}Ra . This observation is vital, since the management of artificial radioactive sources has considerably different implications.

Spectra were also used to better determine the external dose from TENORM scales.



Results and discussion

- Main mineral composition: 3 main kind of incrustations



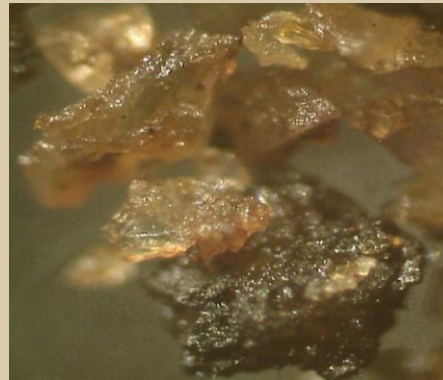
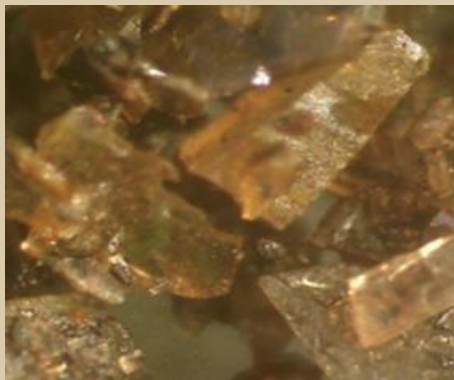
Barite and crude
24 $\mu\text{Sv/h}$



Barite and iron oxide
3-6 $\mu\text{Sv/h}$



Calcite
dose rate: background

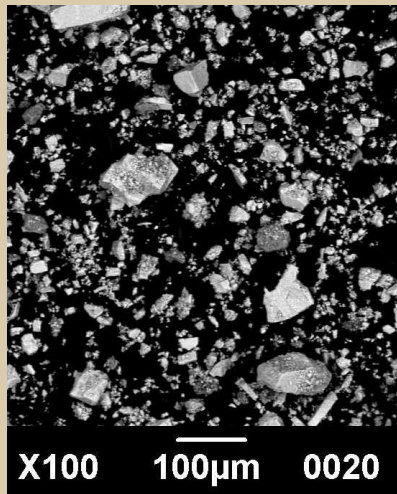


General appearance of the radioactive scales. (top: pictures frame 1 cm), and their minerals after grinding (bottom: pictures frame 1 mm).

Results and discussion

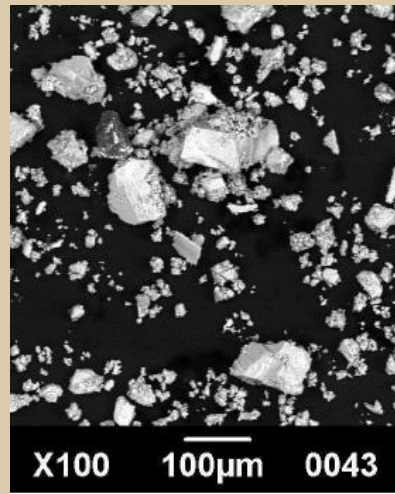
Back scattering electrons image (map of atomic number Z)

Barite and crude



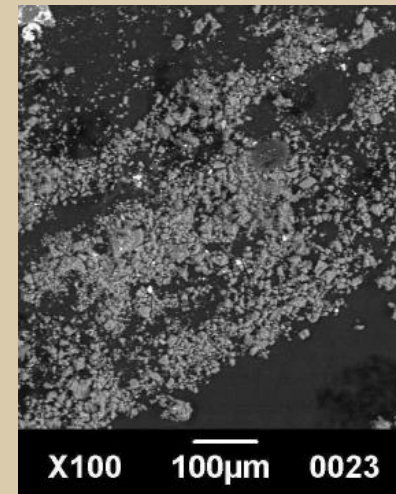
Many bright grains: barite

Barite and iron oxide



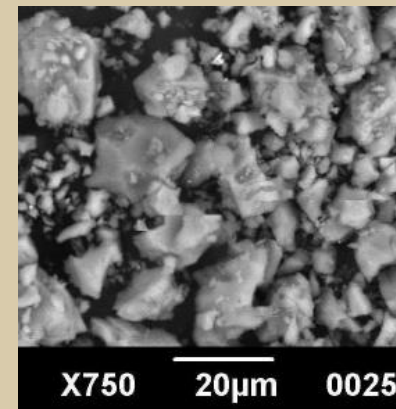
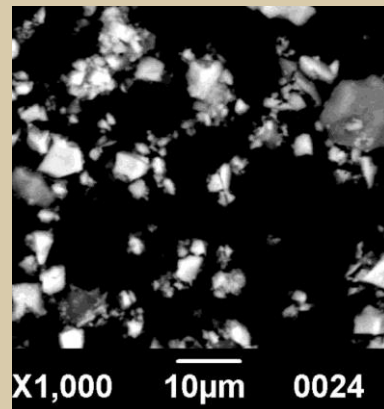
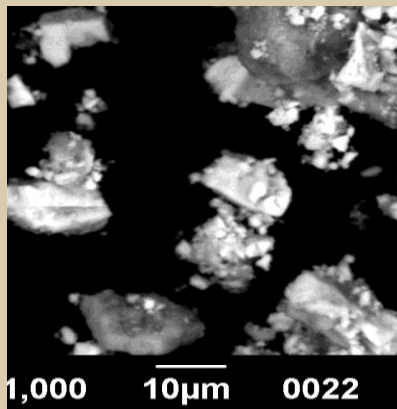
Many as well

Calcite



Very few barite grains
(mainly grey calcite grains, low

Z)



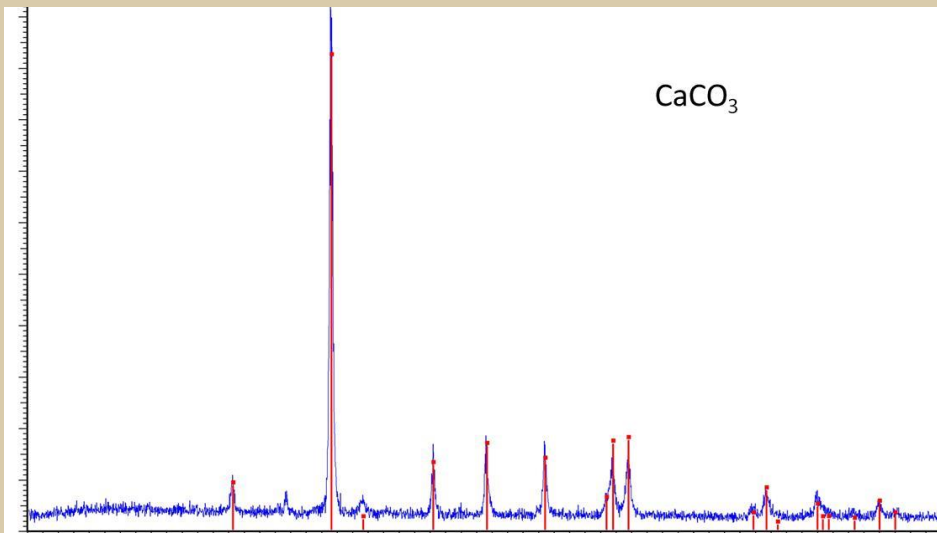
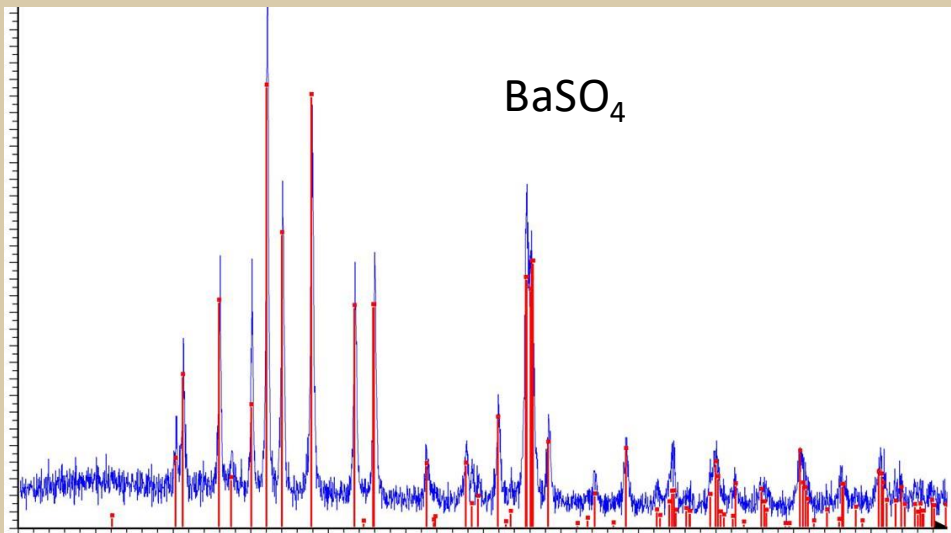
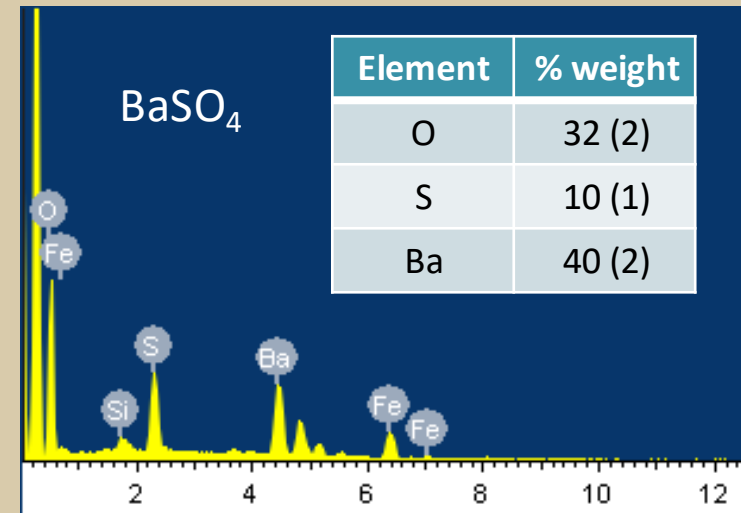
Results and discussion

XRD & SEM with XRF probe

Barite and crude and Calcite
Barite and iron oxide

Barite mineral shows traces of quartz. In the second sample no mineral compounds of Fe were found, so the Fe oxides are amorphous (and mainly originated from the tube corrosion). Calcite scales are almost pure.

The ratio Ba:S as 4:1 confirms that micro crystals are made of barite, where Ra substitute for Ba



Results and discussion

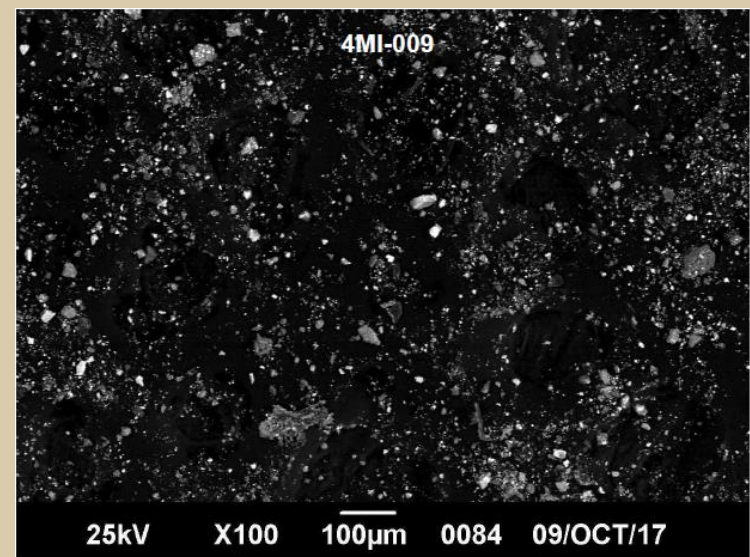
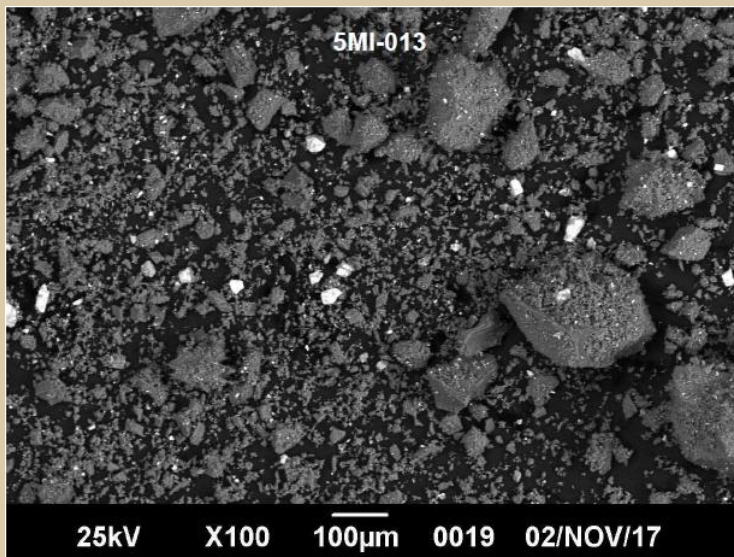
SEM in backscattering mode

Barite mineral is present in a broad range of grain sizes, ranging down to micro-meter and sub-micron sizes, this is mainly caused by the fragility of this mineral. This fact and the low solubility of the barite causes the accumulation of fine dust with barite whitening the tubes and in the contaminated soils.

Then, it is necessary to avoid the loss of scales fragments to the soil

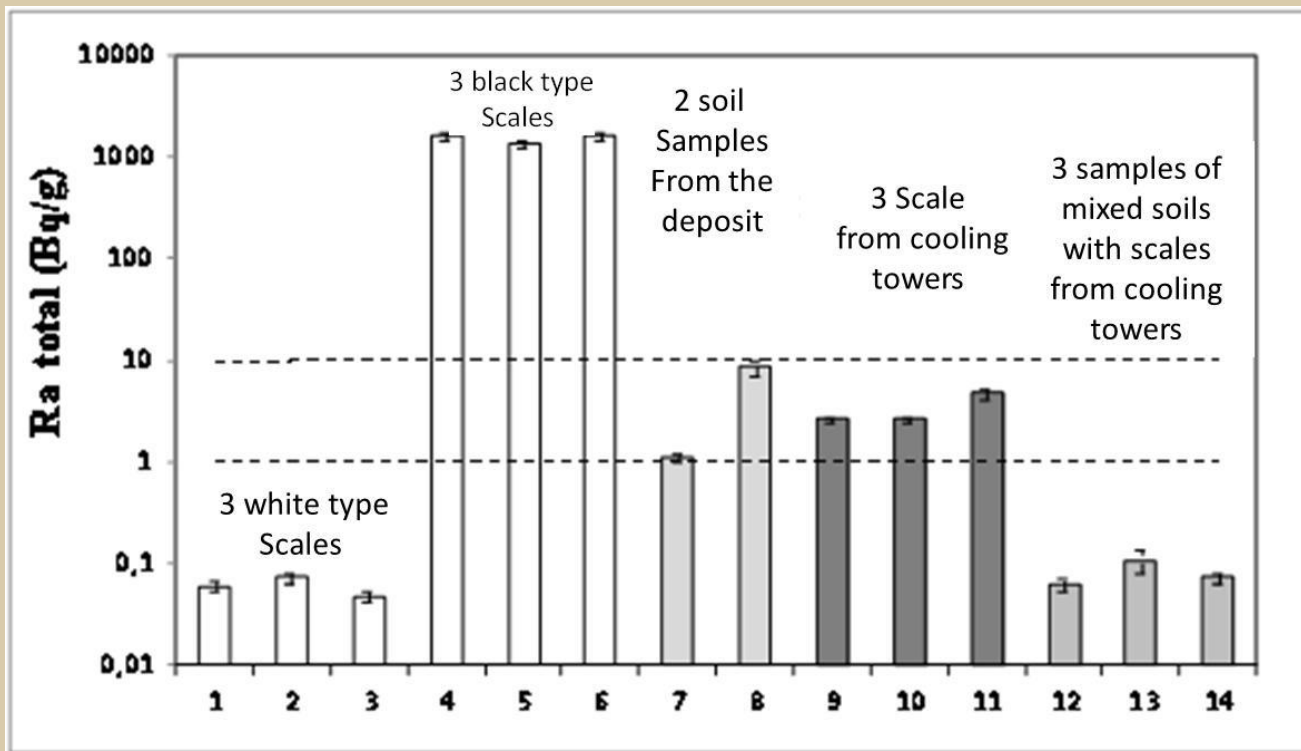
Also, it will be necessary in the future to assess the corresponding AMAD for those TENORM and for the contaminated soils.

In the pictures, the white particles correspond to highly radioactive barite in non-radioactive matrices



Results and discussion

➤ Radioactivity concentration: Site 1



White calcite scales are not radioactive

The major radioactivity levels correspond to the black type scales: barite with crude. Up to 1650 Bq/g ($^{226}\text{Ra} + ^{228}\text{Ra}$)

The soil samples taken near the most radioactive tubes are slightly contaminated probably due to fragments of the scales

The cooling towers produces lower radioactivity concentrations (1 to 10 Bq/g)

The dilution of this scales with local soils reduce the radioactivity down below 0,5 Bq/g

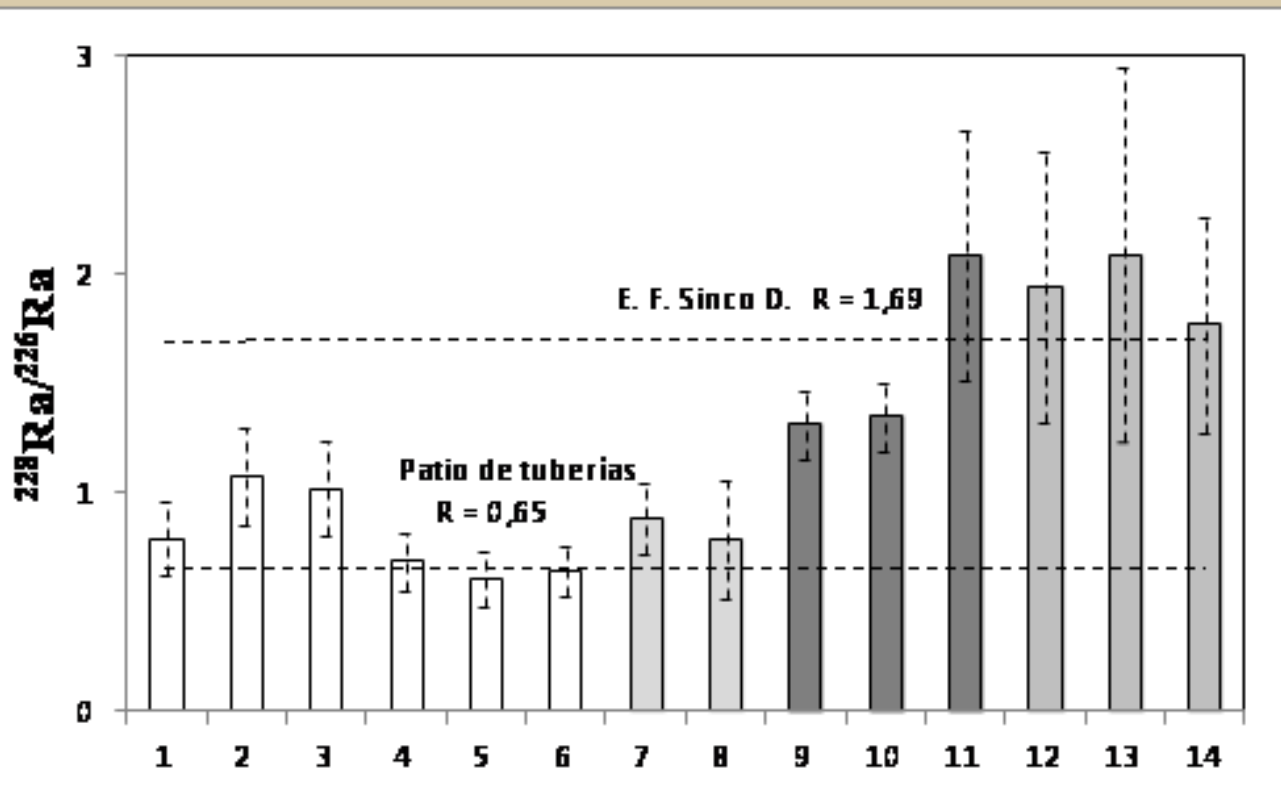
Results and discussion

➤ Radioactivity Ratios: Site 1

^{226}Ra is in larger proportion than ^{228}Ra in the most polluted samples, the calcite samples and the contaminated soils

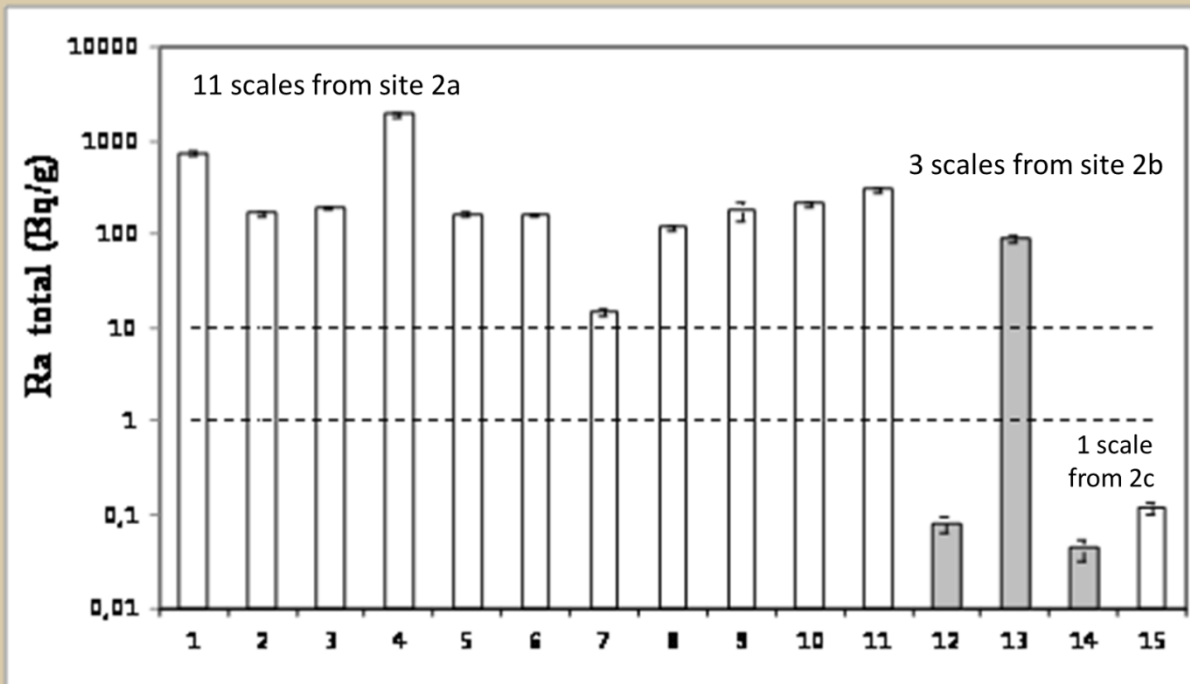
However, the scales from the cooling towers and the local soils contains more ^{228}Ra instead.

The $^{228}\text{Ra}/^{226}\text{Ra}$ isotopic ratio allows the grouping of the different tubes, which have characteristic ratios for each group



Results and discussion

➤ Radioactivity concentration: Site 2

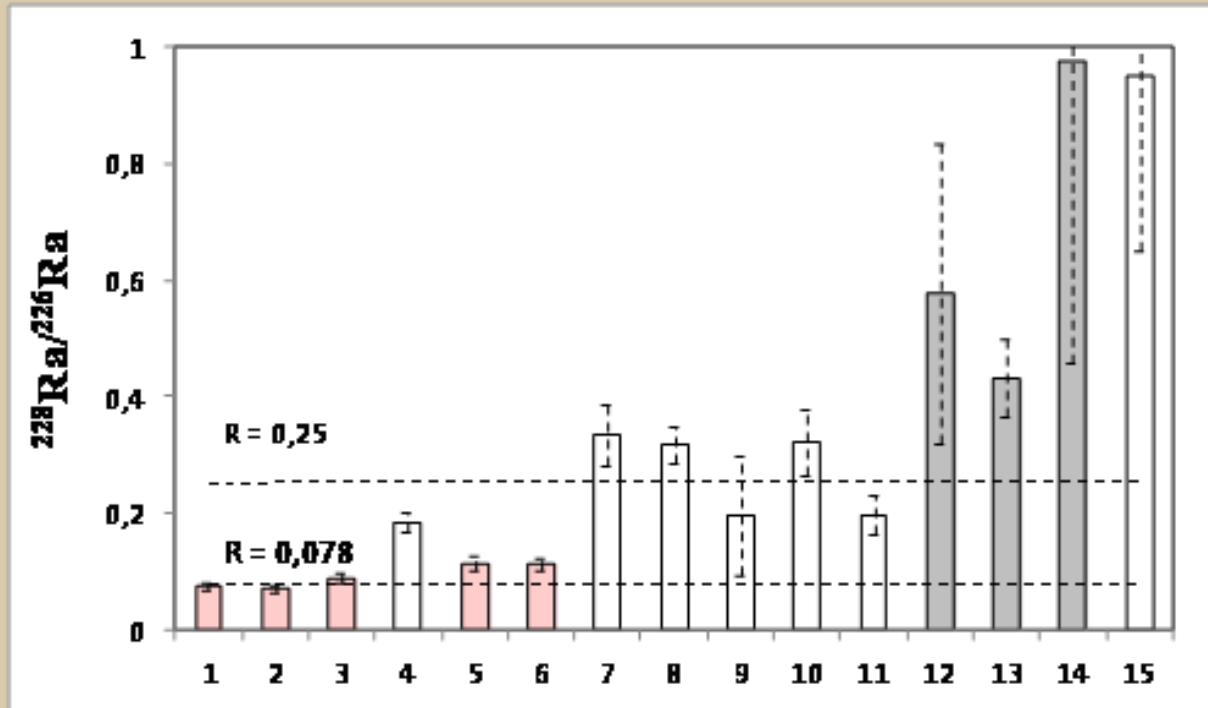


The majority of the identified tubes are contaminated with TENORM scales

Most contaminated tubes have barite scales within a range of 100 to 2000 Bq/g

Results and discussion

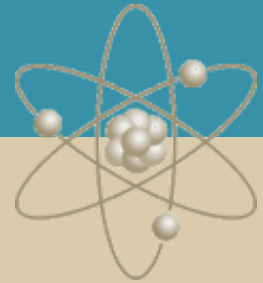
➤ Radioactivity Ratios: Site 2



^{226}Ra is in larger proportion than ^{228}Ra in all the samples

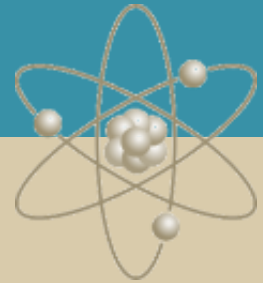
Again, the $^{228}\text{Ra}/^{226}\text{Ra}$ isotopic ratio allows the grouping of the different tubes, which appears to have different ratios for each group

Conclusions



- The vast majority of the pipes stored in the visited sites are free of NORM. However, there is a group of radioactive pipes with scales type TENORM with a considerable amount of radioactivity. There are about 500 contaminated pipes one site and more than a dozen at the other site, this represent more than 5 metric tones of highly radioactive waste.
- The most radioactive scales, which correspond to barite, are dark with small white and bright mineral crystals (translucent or transparent under a microscope). Its appearance is similar to that of a coarse-grained sandpaper, it is amalgamated with hydrocarbon remains (black)
- The external dose rates in most of the places are sufficiently low so that it is not necessary to restrict access to the area. However, near the polluted tubes a prudential distance (1 or 2 meters) of pipes contaminated with TENORM is advised

Conclusions



- The main risk for workers is the possible contamination. The wind can raise dust and transport radioactive particles that, if inhaled, may represent a high risk of contamination and internal radiation exposure, which is particularly important in these cases where there are several alpha emitters and high radioactivity concentrations
- The highest concentrations in each one of the contaminated sites are 1650 and 3000 Bq/g (2 millions of Bq per kilogram), these values are amongst the most radioactive ones at global scale
- In order to avoid the transport of such polluted tubes and to reduce the associated risk, there have been developed a system for *in situ* descaling and decontamination. It is a hermetically sealed portable closed circuit system that can be used to clean the tubes in the oil field or in the deposits. The radioactive wastes must be then stored in a bunker for temporary storage. We are now studying the feasibility of reinjection of these radioactive materials in the form of a mud, down in geologically safe strata.

Aknowledgements



Thanks you all for your kind attention

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- **INTEVEP - PDVSA**, Project: *Radiometric Survey in Storage Facilities / Management of Production Pipes and other TENORM in the National Petroleum Industry (OF-497.1-16, 2016-2017)*
- **IAEA**, National Technical Cooperation Project: *NORM/TENORM monitoring in industrial by-products: radiological & environmental protection and uses as building materials. (VEN/9/012, 2018-2020)*
- **LabNuclear Technical Services LLC**, Project: *Prototype system for in situ descaling and radioactive decontamination of oil production tubes (LNTS-RDI-012, 2017-2018)*

