Radon 222 and Tritium in the prevention of sea water intrusion and oil pollution in a coastal karst aquifer

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Abstract. The partitioning properties of $^{222}\text{Rn}$ with organic liquids have been used efficiently to distinguish the presence of contaminations for hydrocarbons in the ground waters of a coastal karst aquifer of Northwestern Cuba. In presence of sea water intrusion $^{222}\text{Rn}$ and Tritium shows a different behavior. This property is used as an additional advantage for the identification of different sources of Chloride in groundwater; v.gr, sea water intrusion, formation waters or cooling waters used at the gas plants for electric generation. Isolated events of advance (and/or retreat) of sea water intrusion as well as oil spills from pipelines or during oil well completion were detected measuring the activity of the $^{222}\text{Rn}$ systematically in the ground waters.

1. INTRODUCTION

Northwestern Cuba is, basically, a karst territory (Fig. 1). The carbonate rocks of Neogene-Quaternary age forms a large and narrow coastal fringe of about 150 km length and not more than 5 km in its wider part. The karstified carbonate rocks reach a thickness of up to 300 m and different local and subregional flow systems are developed inserted in the so-called Karst Region of Northern of Havana-Matanzas [1]. The region is a coastal holokarst and the hydrogeological conceptual of great part of the territory was updated recently by [2] after the discovery of the the aquifer capacity of the underlying Cretaceous-Paleogene vulcanites and ophiolites that occasionally outcrops in some places at the South of the calcareous fringe.

FIG. 1. General location map

The most important problems affecting groundwater quality are due to the differentiated propagation of the marine intrusion and those derived of an intense onshore oil exploration and production of petroleum carried out since 1970 and enhanced in the last decade. The coast line is also systematically threatened by the presence of hydrocarbons in the marine waters coming from the cleaning of the bilges of ships circulating by international waters.
The hydrogeologic structure of the karst territory is particularly complex. Semi-confined and unconfined aquifer systems differently interrelated alternates with several superimposed permanent and seasonally hydrologic active cave levels as well as different kinds of paleokarsts. The alternation of cave levels leads the development of different conduit and diffuse flow areas that differentiate the development of the local flow systems of fresh, salted and brackish ground water and complicate the mixture of waters from different aquifer horizons and among them and the sea and with the scarce fluvial currents that furrow the territory of North to South.

The presence of waters contaminated by petroleum in semiconfined sectors of the aquifer and detached of the current oil exploration and exploitation, the high volume of formation waters reinjected to the system or disposed in surface but, at the same time, without constituting true oil brines and the locally intensive exploitation of ground waters that, because of the different uses they have allows the extraction of waters with different salinity have complicated the general sketch of the distribution of regional mineralization and the extension of the plumes of hydrocarbons.

For the efficient application of the Cuban national environmental regulations, to order the land and water use and to define the liability of the oil operators, the tourist companies, cattlemen and farmers on the ground waters quality it was required the identification of the different sources of chlorides and, in general, of the ground water salinity, to distinguish the main areas of concentrated natural recharge, to define the area of influence of the formation water injection wells and to map the extension of the oil contamination plume in the regional flow system.

An excellent approach to the solution of such problems was achieved by means of the application of isotope hydrology techniques taking advantage, especially, of the capacity of the Radon 222 as a partition tracer of hydrocarbons and of the Tritium indicator of the residence time of ground waters in the aquifer system. This contribution describes the methodological aspects and the main results obtained. Part of this paper was submitted to the II HISPANIC-LATIN AMERICAN SEMINAR ON CURRENT TOPICS OF GROUNDWATER HYDROLOGY AND IV ARGENTINEAN CONGRESS OF HYDROGEOLOGY and some results have been previously published [3-5].

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2. CONCEPTUAL MODEL

The partitioning properties of $^{222}$Rn with organic liquids established by previous authors [6-11] have been used efficiently to distinguish the presence of contaminations for hydrocarbons in the ground waters of a coastal karst aquifer of Northwestern Cuba. In presence of sea water intrusion $^{222}$Rn and Tritium shows a different behavior. This property is used as an additional advantage for the identification of different sources of Chloride in groundwater; v.gr, sea water intrusion, formation waters or cooling waters used at the gas plants for electric generation. Isolated events of advance (and/or retreat) of sea water intrusion as well as oil spills from pipelines or during oil well completion were detected measuring the activity of the $^{222}$Rn systematically in the ground waters. Tritium activity in groundwater allowed determining the
liability of different oil operators in ground and surface waters. Modeling of groundwater residence time has allowed dating different sources of oil pollution in the deep semiconfined aquifer.

The use of $^{222}\text{Rn}$ is based in its partition property with organic fluids. This property states that when groundwater containing $^{222}\text{Rn}$ in equilibrium state of emission-disintegration migrates to a zone contaminated by NAPLs the activity of $^{222}\text{Rn}$ decreases. When groundwater abandon the polluted site the $^{222}\text{Rn}$ activity should return to the initial equilibrium value. The mathematical model of partition is thoroughly described in the literature [6-11]. The behavior of Tritium and Cl is also well described in the literature and the properties useful in sea water intrusion studies have been summarized by [12].

The main inconveniences of the combined application of these techniques are, however, the short life time of Radon 222, and the low activity of Tritium in the atmosphere and, in turn, in many of the sampled ground waters.

3. $^{222}\text{Rn}$, $^3\text{H}$ and Cl Patterns in Terrestrial Waters

The study was conducted to assess the patterns of seasonal behavior of $^{222}\text{Rn}$, $^3\text{H}$ and Cl in the following cases:

- Fresh groundwater (not affected by sea water intrusion)
- Fresh groundwater affected by isolated events of sea water intrusion.
- Sea water intrusion
- Groundwater affected by oil produced waters spill.
- Groundwater affected by an oil spill.

3.1. Fresh groundwater (not affected by sea water intrusion)

Fig. 2 shows the normal patterns recorded in fresh ground waters close to the recharge zone. A very similar behavior between Cl and $^{222}\text{Rn}$ could be observed. The correspondence is acceptable with a linear correlation coefficient of 0.84 (95% of certainty). Fig. 3 shows the behavior in another station of the same aquifer, 700 m upstream. In this case, between $^3\text{H}$ and Cl, the correlation coefficient is high: 0.87 (95% of certainty).

3.2. Fresh groundwater affected by isolated events of sea water intrusion.

The same aquifer experienced an advance of sea water intrusion in April 2005 (Figs. 4-5) promoting a complete divergence in the relations $^3\text{H}=f(\text{Cl})$ y $^{222}\text{Rn}=f(\text{Cl})$ recorded in EWS 2 monitoring station.

3.3. Sea water intrusion

In this case the dependence Cl-$^{222}\text{Rn}$ (Fig. 5) is not absolutely concluding; nevertheless the relations $\text{Rn}^3\text{H}$ y $^3\text{H}/\text{Cl}$ (Figs. 6-7) shows well differentiated patterns to identify the presence of sea water into the aquifer.

3.4. Oil produced water spill

As stated above, in presence of a produced water spill, Radon decreases its activity not only because of its partition property but because its divergence under conditions of high salinity (Fig. 8).

3.5. Groundwater affected by an oil spill

Figs. 9 clearly shows the partition properties of $^{222}\text{Rn}$ where the migration of the contaminated plume and the aquifer recovery are perfectly recorded in the change of activity of $^{222}\text{Rn}$. 
4. CONCLUDING REMARKS
The partition properties of $^{222}$Rn with organic liquids could be successfully used to distinguish the presence of oil pollution in groundwater. The different correspondence that $^3$H shows in the presence of sea water intrusion is an additional advantage to isolate the presence of Chlorides coming from sea water from those coming from produced waters in oil fields. Isolated events of sea water intrusion advance inland could be successfully detected measuring systematically the activity of $^{222}$Rn in ground waters in a monitoring network.

On the other hand, once more time it should be stressed that the main inconveniences of the combined application of these techniques are, however, the short life time of Radon 222, and the low activity of Tritium in the atmosphere and, in turn, in many of the sampled ground waters. Nevertheless, the easy sampling, conservation and measurement of Radon and Tritium activities as well as Chlorides concentrations together with the low lab prices reinforces considerably the systematic use of these variables in the hydrogeological monitoring of complex coastal karst areas were fresh water resources are strongly threatened by sea water intrusion and oil exploration and production as described in this case study.

FIG. 2. Chloride and $^{222}$Rn patterns in a fresh water aquifer.

FIG. 3. Chloride and $^3$H patterns in a fresh water aquifer.
FIG. 4. Correspondence $3H$-$Cl$ in fresh ground waters in presence of a sea water intrusion event.

FIG. 5. Chloride-$^{222}$Rn relation in groundwater affected by sea water intrusion.

FIG. 6. Chloride-$^3H$ relation in groundwater affected by sea water intrusion.
FIG. 7. $3H-^{222}Rn$ relation in groundwater affected by sea water intrusion.

FIG. 8. Produced water spill

FIG. 9. Seasonal fluctuation of $^{222}Rn$ activity and contamination events of oil spills.

REFERENCES


