MEASURED TRITIUM IN GROUNDWATER RELATED TO ATMOSPHERIC RELEASES FROM THE MARCOULE NUCLEAR SITE

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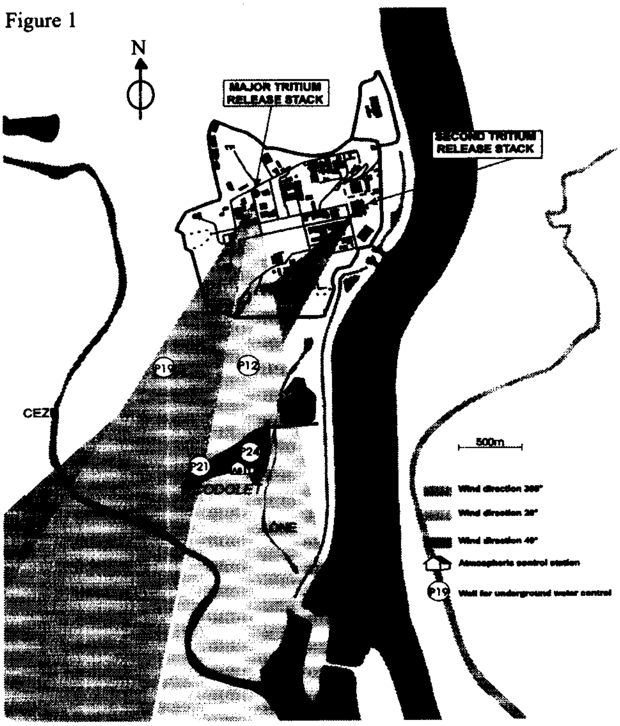
1 - Introduction

Tritium is released into the atmosphere during normal operation from the industrial facilities operated by COGEMA at Marcoule; over a 15-year period covered by this study (1979-1994) the quantities ranged from 4940 to 520 $TBq \cdot yr^{-1}$. Atmospheric release in rainy weather results in tritium migration into the groundwater by a series of mechanisms associated with the water cycle. COGEMA monitors the groundwater by means of boreholes, four of which are shown here (Figure 1). Atmospheric monitoring is also routinely performed; data on the tritium activity concentration in the air and rainwater are available for the same time period. A simplified observation suggests a relation between the atmospheric tritium release and the groundwater radioactivity .In 1994, the activity ranged from 100 to 200 $Bq \cdot l^{-1}$ in the boreholes located 1 km and 2 km downwind from the point of release, diminishing with the distance to less than 20 $Bq \cdot l^{-1}$ at about 3 km.

Figure 1



The authors attempted to model two types of transfers: atmospheric transfer from the release chimney to the borehole, and transfer in the alluvial groundwater. The aquifer comprises the alluvial deposits forming the Codolet plain extending to the south of Marcoule, downwind from the point of atmospheric tritium release. The hydrogeology of the entire Marcoule site has been described in previous studies by the French bureau of geological and mineralogical research (BRGM) and ANTEA.

2 - Modeling Atmospheric Transfer

COGEMA possesses a site model for Marcoule based on a wind-tunnel mockup representing the local relief, and used to calculate the Atmospheric Transfer Coefficient (ATC). Given the known annual tritium releases, weighted by the frequency of tritiated vapor (HTO) conditions, the ATC values are used to calculate the activity concentration in the air in rainy weather, and to determine the concentration in the rainwater:

$$Ar = Aa.\frac{1}{\rho}.f$$

where Ar : radioactivity in rainwater ($Bq \cdot l^{-1}$)

Aa : radioactivity concentration in air ($Bq \cdot m^{-3}$)

ρ : specific activity of water vapor in rainy weather (12.10^{-3} $kg \cdot m^{-3}$)

f : factor characterizing tritiated water vapor ⇄ rainwater exchange[1] depending notably on distance and intensity of rainfall.

The authors determined an experimental value for f using the weekly measured values in the rainwater and air at the atmospheric monitoring station (AS1): $f = 0.15$. The measured rainwater activity at AS1 correlates satisfactorily with the value calculated by the model (Figure 2). The model may therefore be used to calculate the rainwater activity contributing to the groundwater activity at any point on the plain.

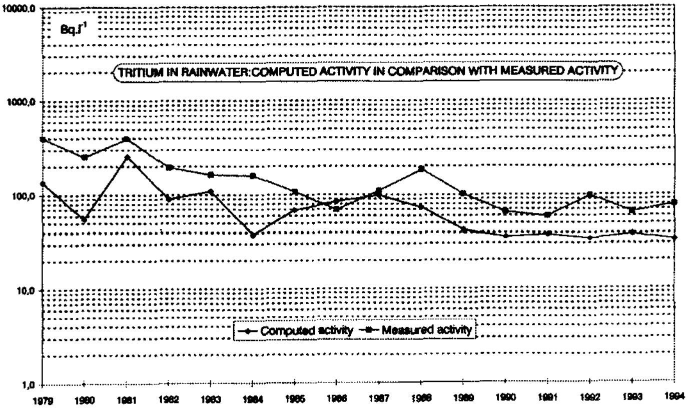


Figure 2

3 - Modeling Rainwater – Groundwater Transfers

When tritium-laden rainwater reaches the ground, the complexity of the subsequent pathways depends on the nature of the soil and its moisture content:

- A negligible fraction enters the surface hydrographic system directly in lakes or waterways.

- Runoff water contributes to the surface water flow.

- Another fraction infiltrates directly into the soil and may enter the evapotranspiration cycle, constituting an available water supply in unsaturated zones, or may slowly percolate down to replenish the groundwater. The rainfall replenishment fraction is subject to specific hydrodynamic mechanisms in the aquifer, and migrates at a rate depending on the hydraulic gradient, the permeability and the storage coefficient in the case of transient variations.

Tritiated water migration in the aquifer is the result of three kinematic phenomena:

- convection, corresponding to the mean fluid displacement;

- kinematic dispersion, due to the heterogeneity of the microscopic flow rates;

- molecular diffusion through Brownian movement of water molecules (this phenomenon is negligible in a permeable aquifer).

3.1 -Calculating the Rainfall Replenishment

Climatological data on rainfall, temperature and insolation obtained from the French Weather Service for the period from 1979 to 1994 were used to determine the evapotranspiration potential (ETP) from the Turc and Penman formulas. Groundwater replenishment by rainfall was estimated using the GARDENIA hydrological model developed by the BRGM, in which a series of reservoirs simulate the principal mechanisms of the water cycle: rainfall, evapotranspiration, infiltration and flow. Transfers from one reservoir to another are governed by simple laws and are adjustable for each reservoir. The model calculates the theoretical water table from rainfall records and from the ETP. The model flow parameters are adjusted by comparison with measured groundwater levels.

The mean annual replenishment calculated in this way over the time period of this study was 145 mm for a mean annual rainfall of 740 mm. The ratio of the two terms, approximately 20%, is consistent with the values commonly observed for this type of aquifer.

3.2 - Hydrodynamic Flow Model

A hydrodynamic model is used to quantify the groundwater flows based on the calculated rainfall replenishment, the aquifer geometry and hydrogeological parameters (permeability, storage, etc.) and boundary conditions. The MARTHE code (BRGM) solves the diffusivity equation using a finite-differences algorithm. In the hydrogeological context of the site, a single-layer regular square mesh was used.

The model was validated for the Codolet plain by comparison with reference data representative of intermediate water conditions from a synchronous piezometric survey conducted in October 1995; under these conditions, the groundwater drains into the Lône and the Cèze.

3.3 - Transport Model

The objective is to account for the tritium activity values measured by piezometers in the Codolet plain based on the various groundwater migration mechanisms using the MARTHE code, which also integrates specific transport equations.

Tritium influx to the groundwater was estimated over monthly time steps from the calculated rainfall replenishment and from the radioactivity measured in the rain by COGEMA. These values were used as input for the model in the form of a mass infiltrated over the entire surface of the Codolet plain.

The model uses the hydrodispersive parameters (kinemattic porosity, longitudinal and transverse dispersion) estimated for the aquifer after several groundwater tracing campaigns carried out earlier on the Marcoule site. The simulations covered a 15-year period from 1979 to 1994. The model calculates the radioactivity of each grid square for each monthly time step to account for the measured variations and the iso-activity maps.

4 - Conclusion

The results show satisfactory agreement between the radioactivity values measured in the boreholes and the calculated values (Figure3). It appears that the tritium activity observed on the Codolet plain may therefore be explained by the contribution of rainfall, based on a deterministic approach taking into account all the physical phenomena involved in the transfer.

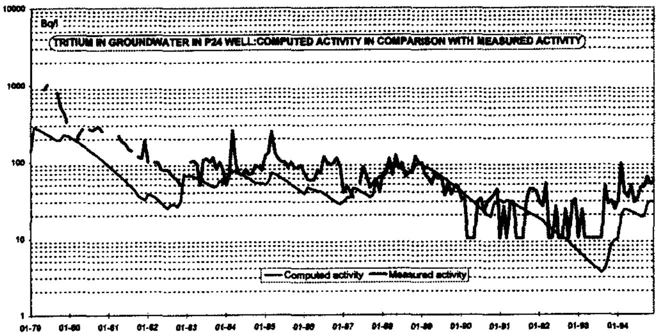


Figure 3

The combined use of several models makes it possible to predict the groundwater radioactivity according to the released activity and the average weather conditions.

References

1. Chamberlain and Eggleton, "Washout of Tritiated Vapor by Rain", *Int. J. Wat. Soil*, **8** (1964) pp. 135-145 .

2. Cortella *et al.*, "Surveillance du tritium dans l'environnement", *SFRP Journée Tritium*, Dijon (April 1986) pp. 273-279.

3. Yoshikazu Inove *et al.*, "Ecological Aspects of Atmospheric Discharged Tritium in the Vicinity of Nuclear Facilities in Japan", *IRPA*, **1** (May 1984) pp. 180-183.

4. Brissaud ,Alamy,Landreau and Thiery, "Wastewater infiltration-percolation of aquifer recharge or water reuse",*International symposium on ground water management:quantity and quality,2-5 October 1989 Spain*

5. Thiery D, "Modeling contaminant transport through unsatured zone in transient state with a random walk particles model",*Proceedings of the international conference on modeling groundwater flow and pollution ,Nanjing 1991, pp311-316*

6. Lecomte ,Crochet, Sauter and Vancon, "Feasibility of remediation of aquifers polluted by organics compounds; pumping strategy and field application in France" .*Contamined Soil 95 pp1057-1066,Kluver AcademicPubilsher(Netherlands)1995*