ENVIRONMENTAL ISOTOPES AND ARTIFICIAL TRACER TECHNIQUES FOR INVESTIGATION AND MONITORING OF HETEROGENEOUS AQUIFERS

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SUMMARY

The paper presents the results of the use of artificial tracers and of geochemical and isotope analysis carried out in an area of the alluvial plain of the river Esino near the town of Ancona (Italy), polluted by Cr-VI from an industrial plant, and in three monitored sanitary landfills located in the upper alluvial plain of the river Po near the town of Parma (Italy) and near the town of Firenze (Italy). The values of the local background pollution parameters have allowed the interference of the landfill with the surface environment to be verified. The method utilized to control the interference of the landfill with underground environment, on defined hydrogeological situations, concerned a chemical and isotope control on surface water, groundwater, leachate and biogas. With the use of artificial tracers it was possible to know the transfer time of pollution to groundwater. High level of tritium were measured in the landfill leachate and in the polluted groundwater, and no values of tritium was detected in two confined groundwater.

KEYWORDS: pollution, artificial tracers, landfill, leachate, groundwater, environmental isotopes, aquifer.

1. INTRODUCTION

In this paper two methods for the study of pollution propagation in groundwater are illustrated; these methods have given good results in Italy concerning to several areas polluted by sanitary landfills and aquifers contaminated by uncontrolled industrial activity .

The first method consists of the utilization of artificial tracers to establish the main character of the aquifers and to calculate effective velocity (u) and pollutants propagation in groundwater.

The second method is based on comparison between chemical and isotope analysis of clean groundwater and contaminated leachate or groundwater.

The pollution risk is mainly due to industrial process and sanitary and hazardous landfills. It's known that the hazard increases in presence of heterogeneous aquifers, due to their generally high permeability.

The pollution control on these aquifers can be made by environmental isotope analysis, and by measures of groundwater dynamics carried out by means of artificial tracers. With these techniques the "Point dilution method"(q) and the "Transit time method"(?) were utilized.

For investigations on contaminated aquifers it is necessary to perfectly know the lithological – stratigraphic characters of the area to be controlled, and the hydrogeological parameters of the aquifers.

In the geologically heterogeneous aquifers it is difficult to calculate the hydrogeological parameters, so, in these cases, it is impossible to use the traditional technology for measuring the hydrogeological parameters and the groundwater effective velocity (?). It is necessary to take in consideration the artificial tracers techniques.

The obtained results are useful to evaluate the pollution risk especially on the areas where there is a collection of groundwater (Tazioli, 1993). In fact, with groundwater velocity values it is possible to calculate the "safety time" to protect the water plug works for drinking uses.

The use of isotope analysis on groundwater and surface waters gives good results for the control of the interference of leachate and the surface water. The study of the landfills in Italy with isotope techniques have been made until 1989.

The isotopes used for the control of sanitary landfills activity have produced very good results; the most commonly utilized are:

- $\delta^{18}O$, δD and tritium (^3H) of water molecule
- $\delta^{13}C$ of dissolved carbonate, bicarbonate and CO_2
- $\delta^{34}S$ and $\delta^{18}O$ of dissolved sulphates
- $\delta^{13}C$, $\delta^{18}O$, δD for biogas CH_4 and CO_2

The utilization of water molecule isotopes can furnish useful indications about leachate to groundwater transfer. They can flow through silty and clayey soils having a near to zero – variation, whereas main chemical parameters are strongly reduced by clayey materials.

In any low permeability aquifers tritium values inside and outside a landfill have been verified with little variations. The values of $\delta^{18}O$ and δD have given information for the area of the recharge of groundwater in relation to the landfill leachate.

For example, in two landfills near Parma (northern Italy) the natural tritium (^3H) values during 1995-2000, varied from 8 to 16 TU for clean groundwater and surface water, while for the landfill leachate the tritium values was about 100 times higher than the tritium levels of the rainfall. Actually the natural tritium of the rainwater in Italy is 5-14 TU.

When it is possible to provided the local background parameters of the surface water and groundwater, it is necessary to verify if there is pollution from a sanitary landfill. Some landfills, located to north and center Italy have indicated values of the pollution of leachate and in same cases the transfer out off landfills where on the silty-clay there was high tritium of the leachate but a very low variation of COD and BOD_5.

2. CASE STUDIES ON THE USE OF ARTIFICIAL TRACERS

A great number of substances can be used as artificial tracer; they have to be chosen in relation to their utilization for any aims to be reached. However, the main artificial tracers which have been used in several situations are the followings:

^{51}Cs – Cr-EDTA Chromium EDTA

^{131}I – NaI Sodium iodide

^{82}Br – NH_4Br Hammonium bromide

$C_{20}H_{10}O_5Na_2$ - Uranine

$C_{20}H_{31}O_3Na_3Cl$ - Rhodamine WT

NaCl – Sodium chloride

NH_4Cl – Hammonium Chloride

NaI – Sodium Iodide

These elements have been utilized in a lot of cases around the Italic peninsula. In this paper it will be presented a case of groundwater pollution in an area of river Esino near Ancona at central Italy, and in an area of Po valley in northern Italy.

2.1Alluvial aquifer contaminated by Cr-VI

In a coastal area of Marche region at central Italy an Industry of galvanic treatments has contaminated the groundwater with Cr-VI till a distance from the industry of about 12-13 km. The thickness of the polluted groundwater was 15-20 meters (Fig. 1). The maximum concentration of Cr-VI in the groundwater was 150 mg/l.

From a geological point of view this area is made with sand, gravel and silty-clay of Holocene and with a terraced alluvial deposit (Middle and Upper Pleistocene), as indicated at hydrogeologic section of Fig. 2.

The industry of galvanic treatments is located on the terraced alluvial deposits characterised by a high permeability (Fig. 3). An impervious diaphragm was made as a barrier for the area of the industry, inserted from the ground surface till the silty-clay of

Fig. 1. Geological map of the area near Ancona at the valley of river Esino polluted with Cr-VI. 1, alluvial deposits made up of sand, gravel and silty-clay of Holocene; 2. terraced alluvial deposits of Middle and Upper Pleistocene; 3, trace of the hydrogeological section of Fig. 2.

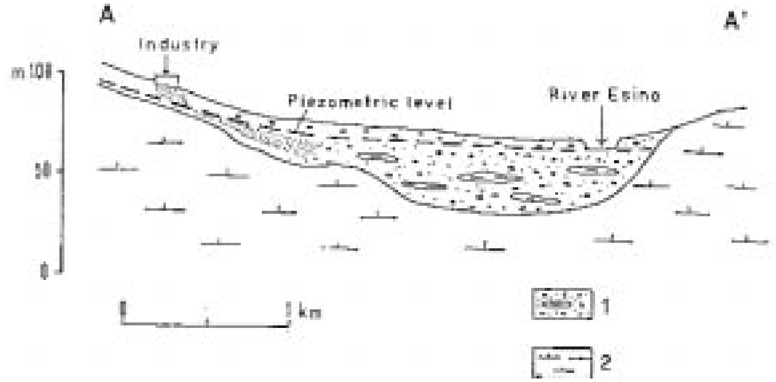


Fig. 2 - Schematic hydrogeological section of the polluted area. 1, alluvial deposits; 2, marly blue clay with silty-clay of marine origin.

Holocene at 8-9 meters depth, as indicated in the section of Fig. 3. The impervious barrier was made with bentonite and cement.

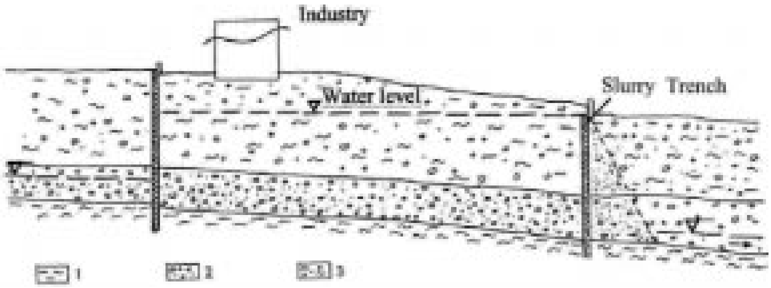
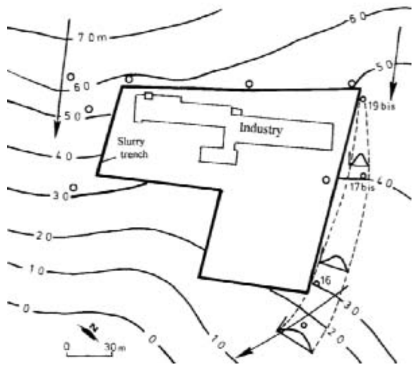


Fig. 3. Area of the cromature industry closed with slurry trenches inserted till the clay at 8-9 m depth. The section indicate the overflow of the polluted water of the industrial till groundwater layer. 1, marly-clay; 2, gravel; 3, clay-mud.

Close to the industry there are some wells for drinking water, so it is necessary to know the exact times in which the Cr-VI will arrive to these points; in this case it is necessary to use the artificial tracer to calculate the values of effective velocity (u). At the external side of the industry were made artificial tracers measures with Rhodamine WT and Hammonium Chloride (NH_4Cl), as indicated at Fig. 4 and 5.



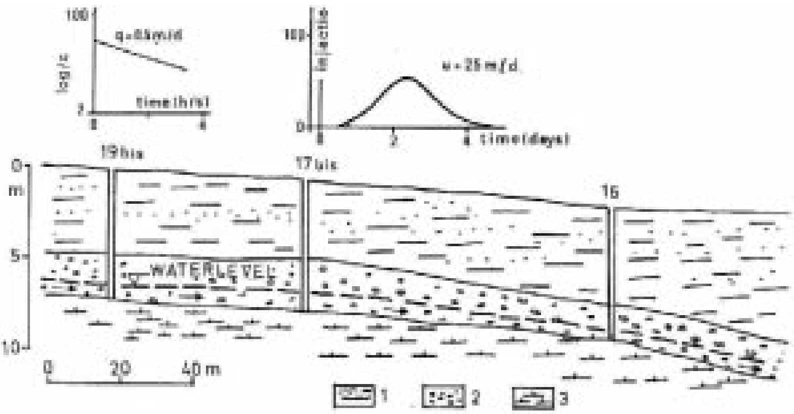


Fig. 4 . Groundwater contour lines and measurement of the specific discharge (q) and the effective velocity () of groundwater with the use of artificial tracers in the area outside of the industry.

The obtained results were:

q	=	0.5 m/d	specific discharge
ū	=	25 m/d	effective velocity
i	=	0.02	hydraulic gradient
k	=	10^{-2} cm/s	permeability

With these values it is possible to verify the transfer times of pollution to groundwater, even till the centre of the fluvial valley where there are the wells for drinking water (Fig. 4).

2.2 Aquifer in Po river valley

Other measures of effective velocity (u) were made on alluvial heterogeneous aquifers either in natural flow or perturbed flow. These measures concern areas located in northern Italy near the town of Modena (Fig. 5).

Fig. 5. Location of the "Fossa Spezzano", in which the variations of the channel discharge have been measured in different points.

In particular it has been studied the polluted water dispersion from artificial channels to groundwater; the artificial channels were contaminated by urban outflows.

The geology of the area is made by gravely sands with clayey silts layers, which contains a groundwater layer located about 10 mts from ground table. The aquifer permeability is very high. The artificial tracer used is Rodhamine WT. The channel discharge was from 100 to 1800 l/s, with leaks of about 10-150 l/s (Fig. 6).

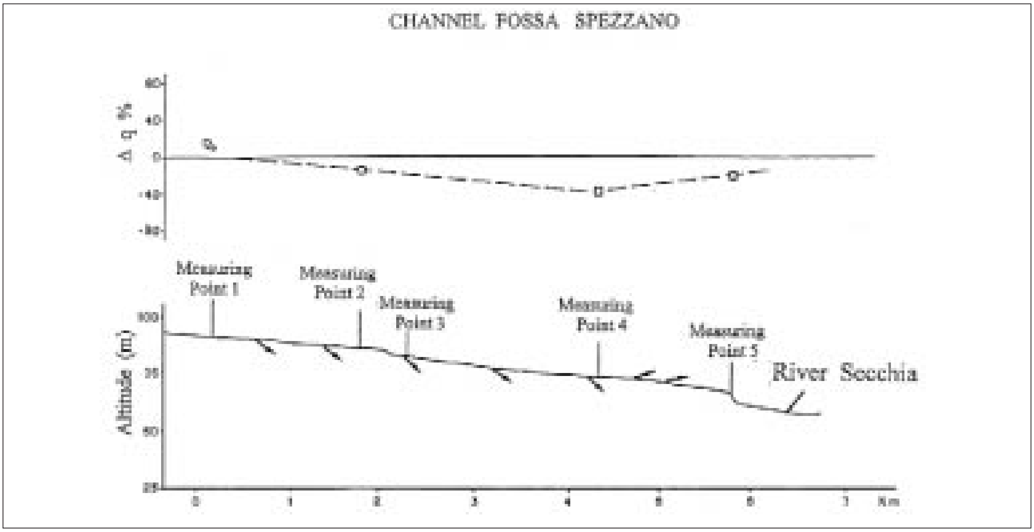


Fig. 6. Variation of the channel "Fossa Spezzano" from Spezzano to river Secchia.

With this artificial tracer it has been possible to establish how many pollutants were going to the aquifer during a certain period.

3. CHEMICAL AND ISOTOPE METHODS FOR THE CONTROL OF THREE SANITARY LANDFILLS IN ITALY

A study was carried out on three sanitary landfills near the town of Parma, (northern Italy) and near the town of Firenze, central Italy (Fig. 7), from 1989 to 2000. The landfills of Parma are located in the upper alluvial plain of the river Po valley (Fig. 8). The Ravadese Landfill is located at north of Parma over an alternance of alluvial Quaternary clay and silty-clay from the ground surface up 6 to 40 meters depth. This area is about 50 meters a.s.l..

The level of the confined groundwater is of 2 to 4 below the ground surface, while the level of a little unconfined groundwater layer is generally lower than that of the confined groundwater (Fig. 9) (Calestani et al., 1999).

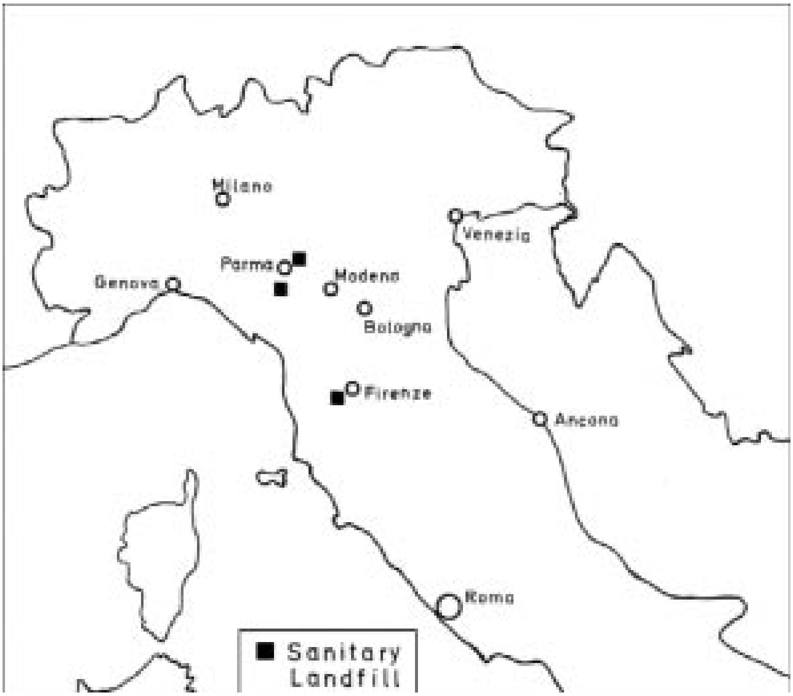


Fig. 7. Sanitary landfills located near Parma at northern Italy and near the town of Firenze at central Italy.

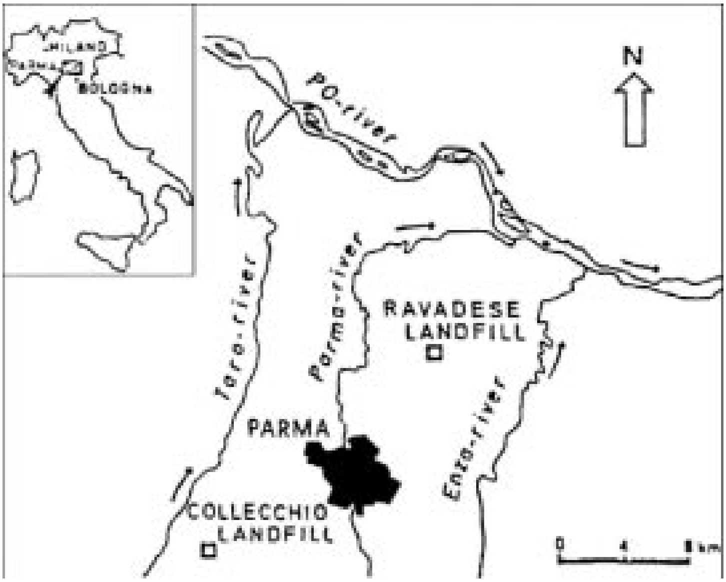


Fig. 8. Location of Ravadese and Collecchio Sanitary Landfills.

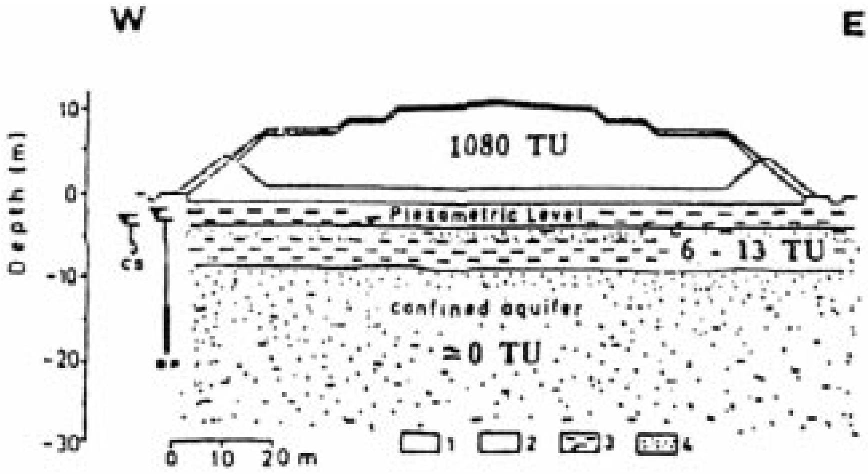


Fig. 9. Hydrogeological transversal cross-section at Ravadese Landfill area. 1, waste; 2, compacted natural clay; 3, silty layer with silty-sandy thin beds; 4, sandy and sandy-silts sediment and values of tritium activity on landfill leachate and on confined and unconfined groundwater (Pellegrini et al., 1998).

The Collecchio Landfill located at south of Parma is at 80-100 meters a.s.l. and is characterized by an alternance of alluvial Holocene clay and silty-sand from the ground surface to 10-13 meters in depth. There is also an alternance of alluvial Holocene and Pleistocene gravelly-sand and silty-clay sediment that is from 13 to 14 meters depth. The phreatique groundwater layer is at 20-25 meters in depth (Fig. 10).

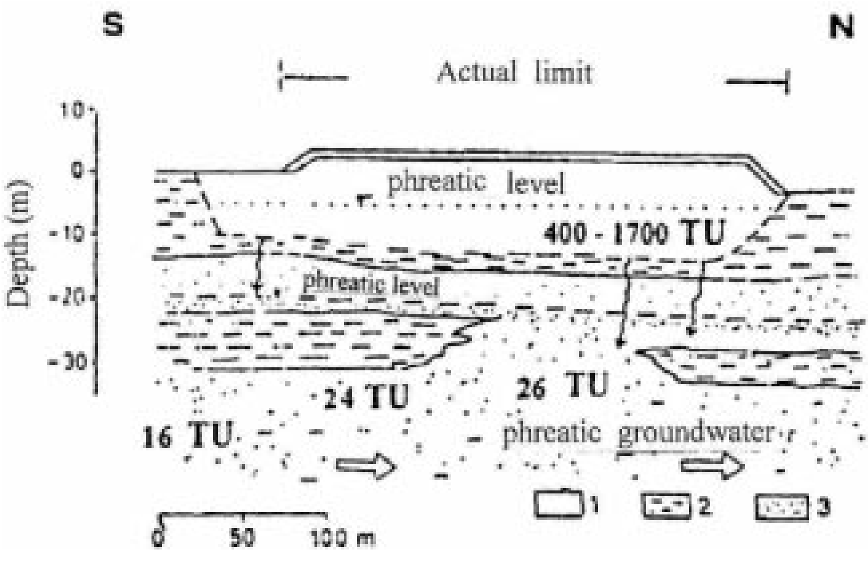


Fig. 10. Hydrogeological transversal cross-section at Collecchio Landfill area. 1, waste; 2, silty and sandy-clayey layer; 3, alternance of gravel and sand; values of tritium activity (Pellegrini et al., 1998).

The area of sanitary landfill of San Martino a Maiano near Firenze has lithological character given by blue Pliocene clays which contain gypsum and beds of sand, whose thickness is between a few centimetres to 4 meters.

Some sandy layers contain organic matter and concentration of NH4+ varying from a few ppm to 25 ppm. The sandy layer contains confined groundwater that, in some place, is in pressure.

The chemistry of clean groundwater shows a sodium chloride facies with high concentrations of sulfates. The dissolved soild content is around 2 – 3 g/l.

Chemical and isotope analysis of surface water and groundwater have concerned the major chemical elements, on pH, redox potential, on NO_3^- - NO_2^- - NH_4^+ - TCO - BOD_5, water temperature and isotope analysis of $\delta^{18}O$, δD and tritium of water molecule, of $\delta^{13}C$ for dissolved bicarbonates - carbonates and CO_2. To check the polluted biogas, chemical analysis for O_2 - N_2 - CO_2 - CH_4 - H_2S and isotope analysis for $\delta^{13}C$, $\delta^{18}O$, δD for CO_2 and CH_4 have been made.

The phreatique aquifer of landfills area is generally fed by local rainfall and irrigation.

In the Radavese Landfill (Fig. 9) and in the San Martino a Maiano Landfill (Fig. 11 and 12) there was no pollution, while on Collecchio Landfill at south of Parma (Fig. 10) there was a pollution in leachate and biogas.

The tritium values of the Ravadese Landfill leachate and of San Martino a Maiano leachate were 1080 and 980 TU, while the values of unconfined groundwater was 6 – 13 TU, comparable to the values of the local rainfall water. The tritium values of confined clean groundwater were about 0 TU.

For Collecchio Landfill area the tritium values were 400-1700 TU,

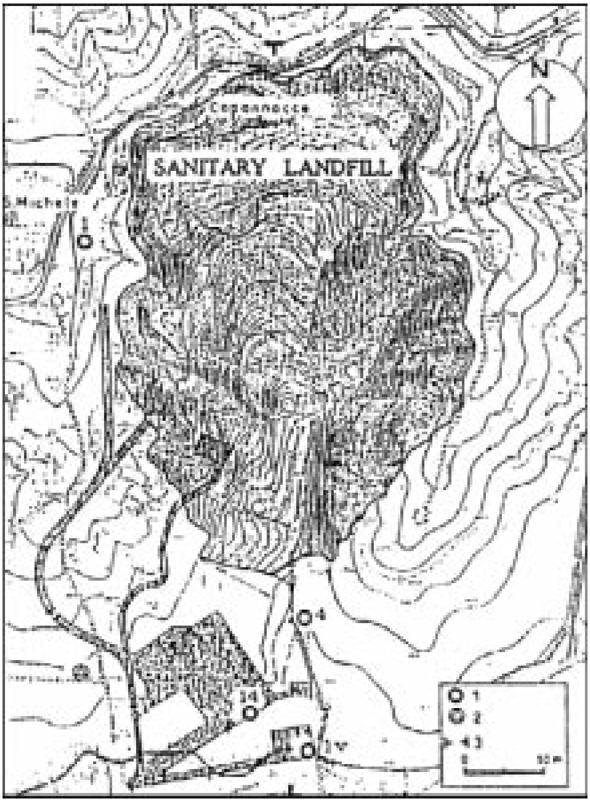


Fig. 11. Location of the landfill near Firenze, San Martino a Maiano. 1, borehole; 2, borehole equipped with piezometers at different depths; 3, trace of hydrogeological section of Fig. 12.

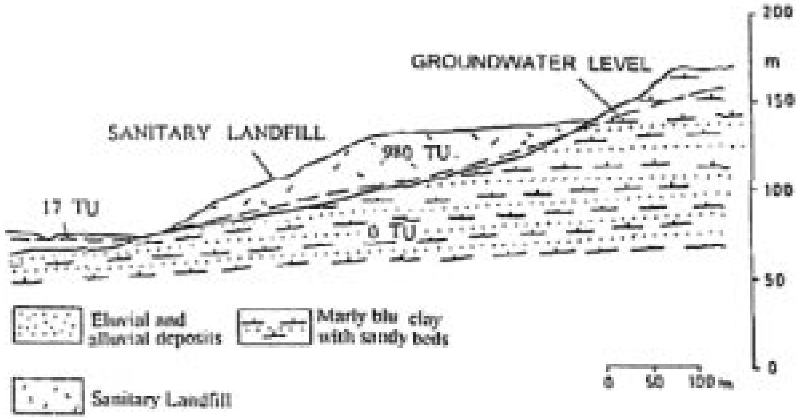


Fig. 12. Hydrogeological transversal cross-section of the S. Martino a Maiano (Firenze) landfill area, values of tritium activity on landfill leachate and on confined and unconfined groundwater (Tazioli et al., 1991).

while the values of the unconfined groundwater, not polluted, were 6-13 TU. The values of polluted groundwater were upper to 18 TU. The contamination arrived till 2,5 km from the landfill toward north, as indicated in fig. 13.

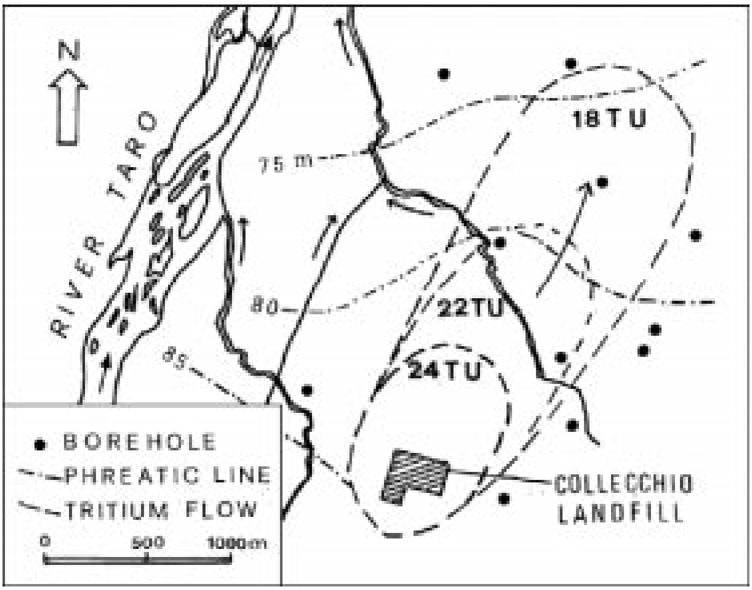


Fig. 13. Leachate migration from the Collecchio landfill, with the use of the measurement of tritium activity of groundwater.

With the analysis of tritium values it is possible to know and detect the pollution flow even at a great distance from the contaminant source; on the contrary, watching the chemical values only, it is not possible to do it with the same easiness and accuracy.

4. CONCLUSION

The use of artificial tracers and of chemical and isotope techniques has produced good results to know the transport time of the pollution to groundwater and for monitoring the interference of landfill activity processes with groundwater. The analysis carried out on water taken from the boreholes near to the landfill, have given the local background parameters which are necessary for the control of the landfill interference.

The control of the landfill leachate interference with the underground environment has been verified especially by means of tritium analysis which produced leachate values much higher than those of rainfall water, surface water and groundwater, as documented by several studies carried out in the past ten years on Italian landfills (Tazioli, 1993; Pellegrini et al., 1998).

REFERENCES

TAZIOLI G.S., TACCONI P.: "Application of Artificial Tracers in Pollution Studies". Adv. Group Meeting on: "Use of artificial tracers in Hydrology". IAEA, Vienna, 19-22 March, 1990, IAEA-TECDOC-601, 1991, 7-34.

TAZIOLI G.S.: "Isotope and Tracer Techniques Applied to studies of Sanitary Landfills and to on Industry for Galvanic Treatment". "Consultants' Meeting on Isotope Techniques in Groundwater Pollution Studies". Vienna, 6-9 December 1993. IAEA 1993, 36-48.

TAZIOLI G.S., BUCCI D., BENETTI E.: "On a Cr-6 Contamination of alluvial aquifer". IV Geoengineering International Congress on "Soil and groundwater protection". Torino, 10-11 March 1994. Associazione Mineraria Subalpina - IAEG Sez. Italiana. Dip. Georisorse e Territorio. Atti Proceedings 2, 1994, 541-547.

TAZIOLI G.S.: "A Simple Nuclear gauge for Suspended Sediment concentration of Natural Stream". Int. Symp. on Isotopes in Water Resources Management, Vienna, 20-24 March 1995, IAEA-SM-336/12P, 1, 1996, 257-258.

PELLEGRINI M., TAZIOLI G.S. & VANNUCCHI M. (1998): "The Use of Isotope Techniques on Sanitary Landfill Control and Pollution Monitoring: The Case of a Sanitary Landfill near Parma, Italy". 8^{th} International IAEG Congress. Vancouver, British Columbia, Canada, 21-25 September 1998, A.A. Balkema, Rotterdam, IV, 2321-2327, 1998.

CALESTANI G., MASSERANO M., PELLEGRINI M., TAZIOLI G.S., VANNUCCHI M. (1999): " On the methods for monitoring the interference of sanitary landfills activity processes with the underground environment. The example of two monitored landfills near the city of Parma, Italy". Seventh International Waste Management and Landfill Symposium, Cagliari, Italy 4-8 October 1999. Environmental Impact, Aftercare and Remediation of Landfills. Vol. IV. Grafiche Galeati, Imola, Italy, 245-252.