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Landfill contamination problems: a general perspective and engineering geology aspects

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ABSTRACT. Using isotopic methods for the investigation of leachate contamination has produced good results mainly for the control of the interference between landfill activity processes and the underground groundwater environment. In this paper the results of study performed in several italian landfill sites are presented. The results obtained from the investigated areas show that the knowledge of background values of the environment is basic for pollution purpose. In particular, Oxigen-18, Deuterium and Tritium values have allowed groundwater residence times and location of recharge areas to be identified. After that, the use of tritium as pollution tracer provides information on the presence of leachate contamination earlier and better than chemical changes of the standard pollution parameters. Deuterium and Oxygen-18 values of leachate always appear to be somewhat different with respect to the groundwater values, but they can be used as contamination index only in cases of high fractions of leachate.

Key terms: Landfill, Monitoring, Tritium, Environmental isotopes, Leachate, Groundwater, Pollution, Rainwater, Waste, Recharge, Leakage, Aquifer, Chemistry.

Introduction

Groundwater and surface water contamination by sanitary landfills has been monitored since 1989 in Italy by using isotope techniques combined with chemical analyses. The results obtained are considered mostly satisfactory for identifying sources of contaminants and predicting the behavior (PELLEGRINI et alii, 1999). We present in this work, as examples, the results of chemical and isotopic measurements performed on rainwater, surface water and groundwater samples, with the aim of investigating the fate of contaminants released from several landfills located near Ancona, Florence (Central Italy) and Parma (Northern Italy, FIG. 1). This short note deals with the application of isotope techniques in surveys on landfills and the control of their impact with groundwater resources. In fact, there are a number of case studies about landfills in Italy, Austria and Germany which show the efficiency of isotope techniques for the evaluation of pollution sources, for following leachate movement and controlling hydraulic permeability of artificial barriers (FRITZ et alii, 1976; RANK et alii, 1992; TAZIOLI, 1993; WARD, 1997).

Sanitary landfills

A monitored landfill is a waste disposal system widely used in Italy and Europe at present. Landfills for waste disposal are generally to be considered as sites in which wastes are stored, designed to minimize the effects of waste disposal (MILLER, 1980). A standard monitored landfill is like a reactor fed with solid waste, rainwater, leachate and biogas. The qualitative and quantitative characteristics of the fluxes of leachate and biogas are related to the solid waste and to the processes taking place in the landfill. An impact on the underground environment is generally caused by exit-fluxes. Landfill usually can contain nonhazardous industrial waste, or nonhazardous municipal waste, or hazardous waste.

As the water infiltrates along the waste, leachate is formed from the liquids found in the waste as well as by

FIG. 1. Location of the investigated landfills.

leaching of the solid waste by rainwater; so leachate is the product of the liquid content of the waste, infiltrating precipitation, and groundwater (if the waste is below the water table). These liquids mix with the waste and dissolve both inorganic and organic constituents (FETTER, 1999). The pH is generally in the range of 6.5 to 8, due to the buffering offered by the generation of large amounts of CO_2 which dissociates to HCO_3^-.

Landfills have proven to be an important source of groundwater contamination in many different geological terrains, climates, and hydrogeological settings.

Method

The hazard assessment of industrial activities and landfills on groundwater resources should be made with prevention studies during the preliminary design phase. After their construction, it will be important to control and monitor the impact of the works on groundwater and surface water. In all these cases, the use of isotope techniques is really basic. By control, it is possible to evaluate the source of the pollutants and their dynamics in the underground environment. With artificial tracers the main hydrological parameters are evaluated in small areas, i.e. hydraulic rock conductivity (K) and groundwater effective velocity (u); these parameters are necessary to define the "safeguard transit time" for groundwater to protect sites of human activities and to evaluate aquifer vulnerability (TAZIOLI, 1993). These studies allow a hydrogeological characterization of the site in which an industrial structure has to be installed, with the aim to evaluate the potential hazard due to the industrial activity. The knowledge of groundwater transit time of a site under study gives the possibility to estimate the hazard degree of the industrial activity for the local groundwater environment. During the project phase, it is also necessary to make the geochemical and isotope characterization of a groundwater environment in order to verify the local natural background of the main parameters which could indicate pollution phenomena during the activity of the industry.

However, in the monitoring phase, and when we have to establish whether a landfill contaminates the surrounding environment, the pollution phenomena should always be investigated by means of isotopic techniques, and methods related to the behavior of tritium in the groundwater, leachate and rainwater.

An abbreviated theory of isotopes

It is known that the use of environmental isotopes allows the determination of groundwater origin and residence time, the recharge area, and the source of chemical elements.

The values of stable isotopes $\delta^{18}O$ and $\delta^{2}H$ are indicated as deviation from the international standard VSMOW (Vienna Standard Mean Ocean Water), which is referred to the mean value of the ratio $^{18}O/^{16}O$ or $^{2}H/^{1}H$ of the Ocean water, while Tritium activities are expressed as Tritium Unit = TU, where one TU represents a $^{3}H/^{1}H$ ratio of 10-18. The values of stable isotopes are influenced by latitude and altitude of precipitation; local studies allow us to determine the ?18O and ?2H altitude gradient with which it is possible to obtain information on recharge areas and in some cases on seasonal recharge.

At present, the values of 3H in precipitation are around 4-14 TU and the same values of tritium activities are measured in those groundwater which are characterized by a rapid renewal. In the past, however, due to thermonuclear experiments with bomb explosions in the atmosphere, the tritium background values in rainwater reached higher values than the present ones. Tritium is absent in confined aquifers with groundwater resident time higher than 40-50 years. Very high values of ^{3}H have been measured in leachate of landfills, as much as 100 times the 3H values of current precipitations (TAZIOLI & TACCONI, 1991; RANK et alii, 1992; TAZIOLI, 1993).

Other isotopes used for prevention and control of groundwater pollution are $\delta^{13}C$ and $\delta^{18}O$ of total dissolved carbon and $\delta^{34}S$ and $\delta^{18}O$ of dissolved sulphates. The values of $\delta^{13}C$ of dissolved carbonates, bicarbonates and CO_2 are around -15‰ and -10‰ referred to the standard PDB (Pee Dee Belemnite), while for surface waters $\delta^{13}C$ values are less high. For the atmospheric CO2, $\delta^{13}C$ is about -8‰; in soil-gas CO2, $\delta^{13}C$ has values ranging from -25‰ to -15‰. In marine carbonates, $\delta^{13}C \sim 0$‰ (LETOLLE & OLIVE, 1983). Under reduction conditions (similar to those present in landfill environments), $\delta^{13}C$ is highly enriched ($\delta^{13}C > +15$‰); so, if compared to the values measured in groundwater, there is a chance to verify the existence of pollution coming from landfills into the surrounding environment.

The values of $\delta^{34}S$ and $\delta^{18}O$ of dissolved sulphates, referred to the standards CDT (Canyon Diablo Troilite) and VSMOW, allow to us control the presence of pollution due to sulphur compounds generally different from those which characterize the natural environment. To best use $\delta^{34}S$, a knowledge of the values which characterize the local background is required, because $\delta^{34}S$ can greatly change in the presence of bacterial reductions in the underground environment. Bacteria preferentially utilize the light isotope inducing an enrichment on sulphates and a decreasing of $\delta^{34}S$ of sulphites of new formation. The values of $\delta^{34}S$ in rainwater range between -8‰ and -4‰, while $\delta^{34}S$ of $SO_4^=$ of evaporitic origin are between +10‰ and +22‰. Negative values of $\delta^{34}S$ have been recorded in materials transformed by bacterial reductions.

Also the nitrogen isotopes may give useful information about the origin of NH_4^+, NO_2^- and NO_3^- produced by agricultural activities or by urban sewage or animal manure.

Methods of isotope utilization

First, it is essential to perfectly investigate the background level of chemistry and isotope values inside the landfill

(leachate) and in the surrounding environment (groundwater and surface water); the objective of these investigations is to establish the base concentration level of the main environmental parameters related to contamination, and obtain indications about the source and residence time (age) of groundwater in the landfill proximity (TAZIOLI A. et alii, 2003). In particular, the methods used for detecting groundwater and/or surface waters contamination derived from the landfill, are based on the occurrence of tritium activity anomalies and chemical concentration changes. These methods are based on the environmental tritium content and isotopic composition of water in the region, and the subsequent evaluation of the data measured in the groundwater inside (leachate) and outside landfill. In fact, several studies in Italy, Austria, Poland and Germany have shown in the last years the existence of very high 3H content in leachates (100÷2000 TU), although the source of this weak radioactivity is not yet well clarified; it is possible that tritium derives from fluorescent screens, deals or pigments, quartz clocks, hospital wastes. An important role is played by evaporation mechanisms, which concentrate the tritium in the leachate, as we are trying to demonstrate by means of laboratory tests we are currently performing. These isotopes (2H, 3H, 18O) are ideal groundwater tracers and allow us to follow the leachate dynamics in the landfill and to monitor the interference with the groundwater environment even over long periods of time.

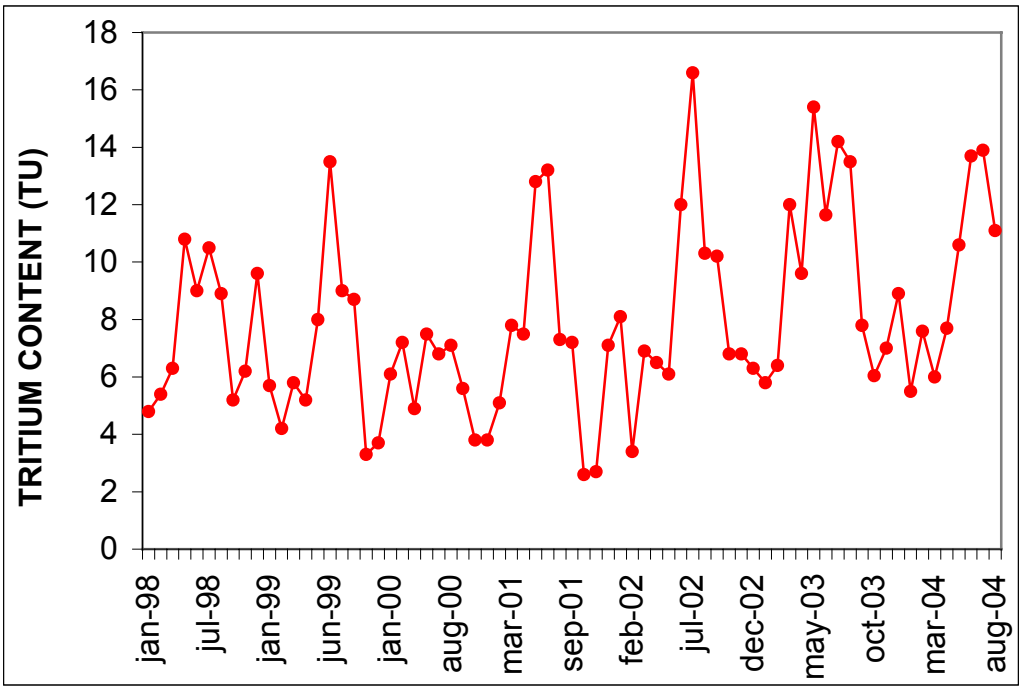

FIG. 2. Seasonal variation of rainwater tritium content in rainfall on Central Apennines (about 1000 m a.s.l.).

In order to estimate the regional base level of environmental tritium in shallow groundwater, the tritium content of monthly rainwater samples collected in some stations on the Apennines was measured. The tritium concentration ranged from 3 to 6 TU in winter months (October to April), and reached maximum values (up to 14 TU) in summer months (FIG. 2).

Monitoring

Chemical and isotope monitoring techniques were usually utilized for landfill investigations, with sampling of surface water, groundwater and landfill leachate; also the major chemical elements were measured, water temperature, pH, electric conductivity (EC, µS/cm), BOD, COD, redox potential (Eh), NH_4^+, CO_2, CH_4, H_2S, as well as the environmental isotopes, ?18O, ?2H and 3H for water molecules, $\delta^{34}S$ and $\delta^{18}O$ for dissolved sulphates and ?13C for dissolved bicarbonates, carbonates and CO_2.

Case Histories

Several examples of the application of isotope techniques and tritium content evaluation method are presented in this section. The utilization of these methods has allowed us to verify the absence of leakage from a landfill to the groundwater, or in other cases to find a source of pollution in the surrounding environment even before chemical parameters in groundwater changed. Some case histories come from past investigations and works (TAZIOLI & TACCONI, 1991; TAZIOLI, 1993; TAZIOLI, 1996; PELLEGRINI et alii, 1999; CALESTANI et alii, 1999), the latest ones concern a work still in progress (TAZIOLI A. et alii, 2003).

Collecchio landfill (Emilia-Romagna)

The Collecchio landfill is located by the River Taro, 22 km south of Parma; the area of the sanitary landfill is characterized by a large aquifer used for water-supply purposes. The presence of pollution due to biogas and leachate migration was recognized especially by means of tritium analyses which gave leachate values much higher than those of rainfall water (TAZIOLI, 1993; PELLEGRINI et alii, 1998; CALESTANI et alii, 1999). The Collecchio landfill area is at 80-100 m a.s.l. (FIG. 3) and is characterized by an alternation of alluvial Holocene clay and silty-sandy clay from the ground level to 10-13 m in depth and by an alternation of alluvial Holocene and Pleistocene gravel-sandy and silty-clay sediment from 13 to over 40 m in depth. The gravel-sandy sediment contains phreatic groundwater with a 20-25 m deep water table. Hydraulic conductivity of the aquifer is about 10-3 cm/s; the superficial silty-clay bed has 10^{-6} to 10^{-4} cm/s. The aquifer is fed by local rainfall and rivers; the recharge area is between 100 to 800 m a.s.l. and the groundwater stays for only a short time in the aquifer. The landfill activity took place from 1980 to 1995.

Table 1 – Isotopic values of groundwater and leachate in landfill areas (S. Martino a Maiano).

	$\delta^{18}O$ ‰ vSMOW	$\delta^{2}H$ ‰ vSMOW	^{3}H TU	$\delta^{13}C$ ‰ V-PDB	$\delta^{15}N$ ‰ (NH_3) v-NBS	$\delta^{34}S$ ‰ ($SO_4^{=}$) v-CDT	$\delta^{18}O$ ($SO_4^{=}$) vSMOW
Ground-water	-7.16 ÷ -6.5	-44.8 ÷ -41.6	8-15 (phreatic) < 1 (confined)	-18.52 ÷ -10.65	+5.09 ÷ +16.13	-21.3 ÷ -3.0	-0.21 ÷ +11.4
Leachate	-6.33	- - -	980	+24.0	+13.19	+8.0	+27.0

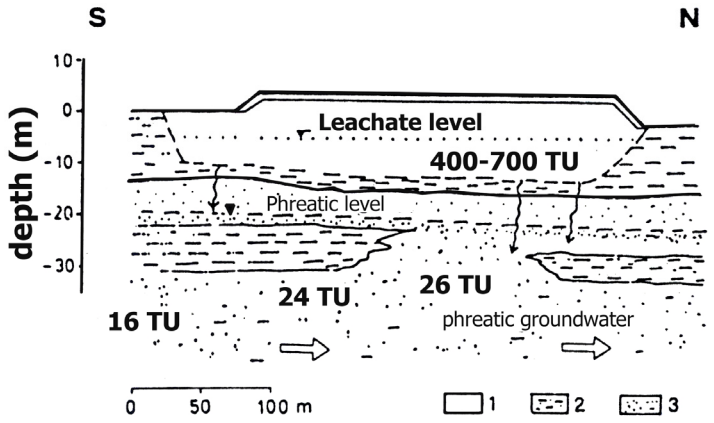

FIG. 3. Hydrogeological cross section of the Collecchio landfill area, with tritium values of leachate and polluted water (values referred to 1997). LEGENDA: 1. waste deposits. 2. silty and sandy clayey layer. 3. alternance of gravel and sandy sediments (after PELLEGRINI et alii, 1999).

In the Collecchio landfill area the geochemistry of unconfined groundwater is of the calcium bicarbonate type, with electric conductivity values between 700 to 1200 µS/cm, with a negative Redox potential (-20 to -30 mV) and a mean water temperature of 13 to 14 °C. The landfill leachate concentration is very variable; the values of COD are 850 to 9500 mg/l, electric conductivity is between 2560 to 22000 µS/cm. The values of leachate temperature are between 20 and 23°C. The groundwater tritium activity allowed leachate migration to be monitored along the S-N groundwater flow direction as far as 2.5 km from the landfill. Considering the local tritium background, tritium values of polluted groundwater are very low, ranging between 18 TU and 26 TU (FIG. 3). For this kind of groundwater pollution, caused by leachate migration from sanitary landfill, the chemistry values indicate a variation in some chemical parameters only close to the landfill, whereas the inhomogeneous tritium values are distributed a long distance downstream of the landfill. These results show that tritium content monitoring may give information on the presence of leachate pollution long before the occurrence of chemical changes in standard chemical pollution parameters.

S. Martino a Maiano landfill (Tuscany)

The aim of the investigation was to verify the presence of a leakage beneath the natural barrier - made of stratified clay - constituting the landfill, and the groundwater environment.

The landfill is located in a small catchment area on marine blue clays with levels of fine sand of Pliocene age, and was monitored between 1989 and 1991.

The aquifer sandy levels, whose thickness reaches 4 m, contain confined groundwater with high concentrations of NH_4^+ (25 ppm) and $SO_4^=$ (800 - 1400 ppm) that led us to suppose a presence of pollution phenomena from the landfill to the surrounding environment.

The values of $\delta^{18}O$ and δ^2H showed a meteoric origin of the groundwater which is recharged by local precipitations. The tritium activities indicated long residence times for the confined aquifer and a rapid renewal for the phreatic one, which had the same tritium activity as rainwater, while the leachate displayed values of tritium activities reaching 980-1000 TU (TABLE 1 AND FIG. 4).

Geochemical and isotope analyses on groundwater and leachate showed that NH_4^+ was naturally produced by organic matter located in the sandy layers and that the dissolved $SO_4^=$ came from nodules and crystals of gypsum belonging to the clay formation (TAZIOLI, 1996).

The presence of high values of tritium in the leachate offers a useful tool to monitor possible leakages from the landfill to the aquifer, because tritium normally displays a good behavior in the underground environment, so that information on groundwater pollution phenomena may be obtained even long before the pollutants have been measured.

The hydrogeological section of Fig. 4, which shows the distribution of tritium in groundwater and leachate, and therefore excludes a leakage from the landfill to groundwater. The results obtained from isotope determinations are listed in Table 1; the most useful information derives from $\delta^{13}C$ of dissolved carbon and $\delta^{34}S$ and $\delta^{18}O$ of dissolved sulphates (TABLE 1). The isotopic data of groundwater are very different from those of leachate, so this fact allows us to distinguish pollution in groundwater caused by leachate, and to assure that there is no active connection between the landfill and the surrounding environment.

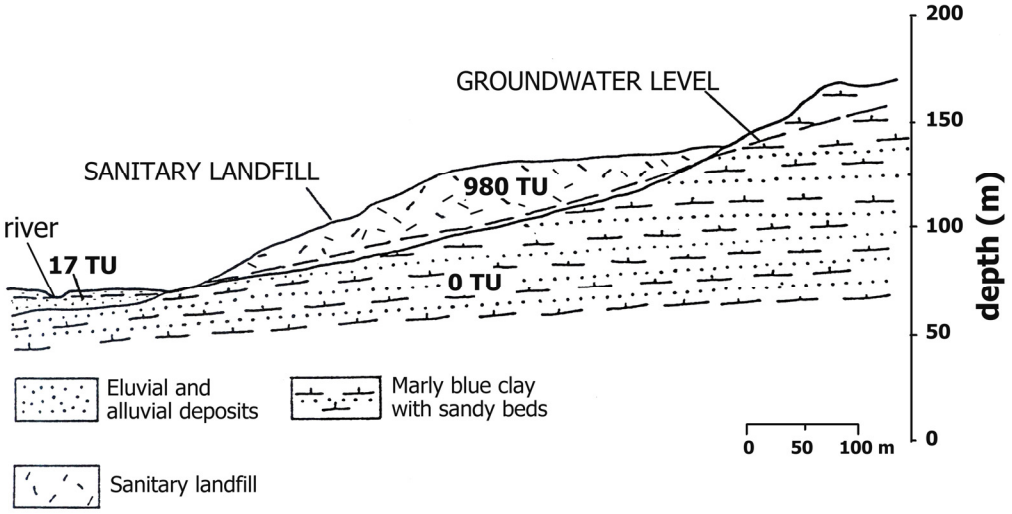

FIG. 4. Geological section and setting of San Martino a Maiano landfill, with tritium content in leachate and groundwater (values referred to 1989; after TAZIOLI & TACCONI, 1991).

Monte Umbriano landfill (Ancona)

The landfill is located in a hilly area near the Adriatic Sea, made up of stratified marly blue clay of Plio-Pleistocene age, interbedded with strata of silty sand. Eluvial and colluvial clayey deposits, affected by landslides, cover the marine formation with a thickness between 2 and 17 m. The hydraulic permeability of the stratified marly blue clay gives values from 10^{-8} to 10^{-6} cm/s, while in eluvial and colluvial deposits the permeability is higher. Groundwater levels vary from -5 m from the ground surface - in the upper part of the area - to 0.5-1 m a.g.s., in the lower part.

Groundwater chemistry displays water of calcium bicarbonate type with high concentration of $SO_4^=$ (1000 ppm) and Cl^-. The landfill was active over a period of 18 years, during which it caused considerable contamination of the soil surface and groundwater environment in the area located downstream of it. In this case, too, the high tritium content of the leachate was utilized as a tracer of groundwater pollution coming from the landfill.

This was possible because the measured tritium activity on the leachate displayed values ranging from 300 to 500 TU, while the activity measured in groundwater was around 10-16 TU (TABLE 2). Measured values of $\delta^{18}O$, δ^2H and ^3H in groundwater of eluvial and colluvial deposits indicate a recharge from local precipitation and a rapid renewal of groundwater. The values of $\delta^{13}C$ in the groundwater are very different from those measured in the leachate (TABLE 2).

Table 2 - Isotopic values of groundwater and leachate in landfill areas (Monte Umbriano landfill).

	$\delta^{18}O$ ‰ vSMOW	$\delta^{2}H$ ‰ vSMOW	^{3}H TU	$\delta^{13}C$ ‰ V-PDB
Ground-water	-6.5 ÷ -6.0	-43.3	8-16	-13.97 ÷ -10.03
Leachate	- - -	- - -	300-500	+15.59

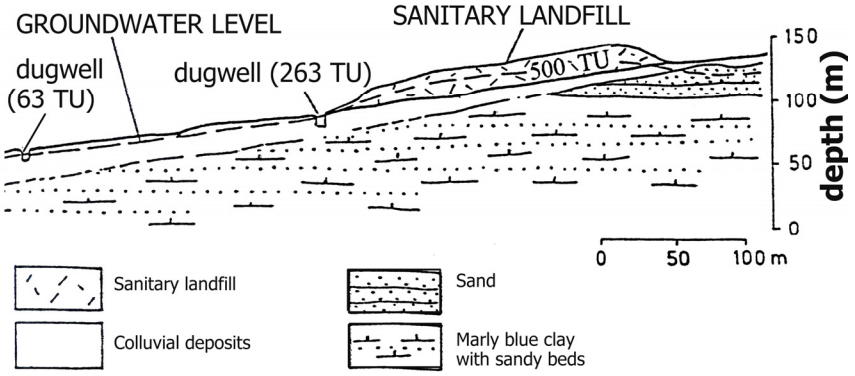

FIG. 5. Sanitary landfill of Monte Umbriano, near Ancona. Tritium distribution in landfill and dugwells downstream (after TAZIOLI, 1993).

A one-year control of the landfill allowed us to verify that the pollution was due to the surface runoff of the leachate during periods of higher rain intensity (winter and spring seasons), while during the period of low precipitation there was no pollution in the area downstream of the landfill. Due to the nature of the sediments and their low permeability, most of the pollutants are absorbed by clay material, and only high tritium activity of leachate was measured in the wells dug downstream of the landfill (FIG. 5), indicating the occurrence of pollution.

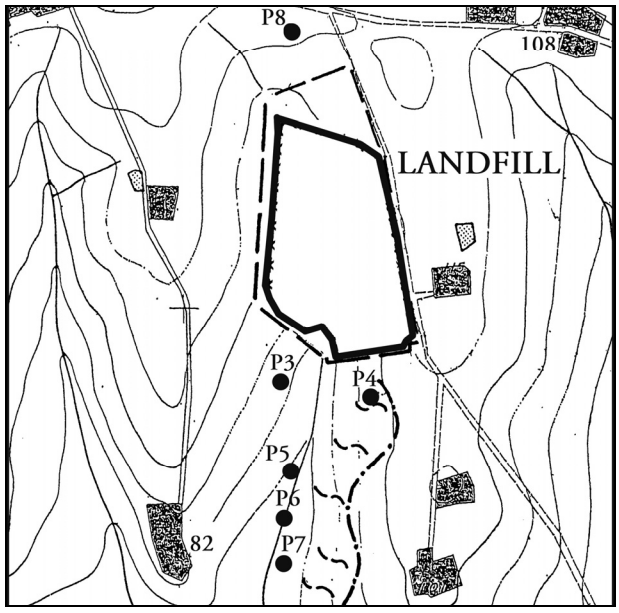

FIG. 6 a. Location of landfill n. 1 (Ancona Province) and wells (after Tazioli A. et alii, 2003).

Landfills in the Ancona Province

Chemical and isotopic measurements performed on rainwater, surface water and groundwater samples, with the aim of investigating the fate of contaminants released from some landfills located near Ancona, Central Italy, are presented here. These landfills have been monitored since 2001. The isotope determinations included $\delta^{18}O$, $\delta^{2}H$ and tritium (^{3}H). The investigations of groundwater and surface

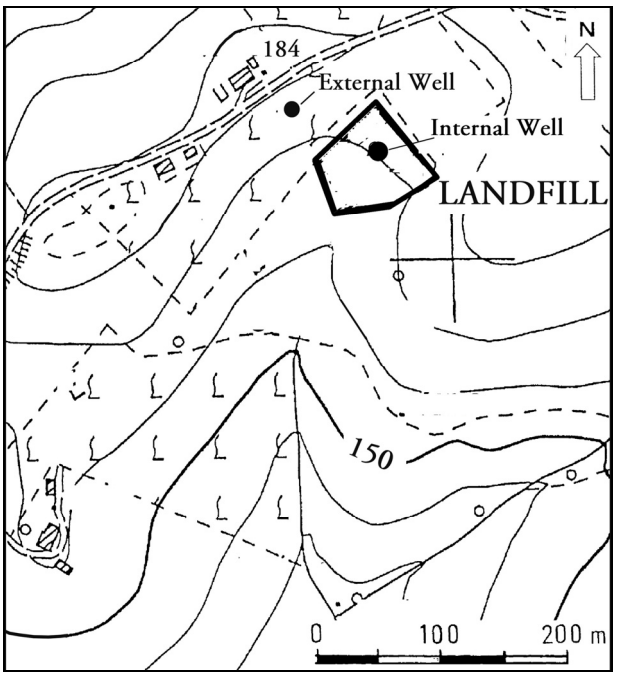

FIG. 6 b. Location of landfill n. 2 (Ancona Province) and wells (after Tazioli A. et alii, 2003).

water contamination were undertaken on landfills abandoned between 1986 and 1999, and located in a hilly zone, 20-30 km north west of Ancona, characterized by clayey and silty soils, and a water table at -5 ÷ -8 m a.g.s. The isotopic and chemical monitoring was started two years ago and was carried out on leachates, surface waters and groundwater (the last-mentioned being sampled in several downstream wells). The isotope determinations are reported in Table 3.

The tritium concentration in leachates can be very high, due to a still active tritium release from the landfill. In principle, therefore, it is possible to identify the contamination of groundwater from the tritium increase with respect to the base level, and also estimate the mixing ratio with the landfill leachate (FRÖLICH, 1994; CHILTON et alii, 1998).In the younger Landfill 1 (FIG. 6 a), which was abandoned in 1999, the leachate tritium concentration is still high (over 250 TU). However, only the downstream wells P4 and P5 display a tritium content above the natural base level, thus showing that groundwater may be currently contaminated by the landfill to a significant degree (leachate fraction 2-3%). It should be noted also that groundwater in well P5 has a recharge source different from that of the other wells, as indicated by the stable isotopes. In particular, in Fig. 7 $\delta^{18}O$ anf $\delta^{2}H$ values for all these landfills are reported; it is notable that the leachates from groundwater and surface waters can be distinguished.

In fact, due to an evaporation process to which the liquid in the landfill is subjected, the isotopic values of the latter are more enriched with respect to those of the groundwater; moreover, the surface water displays an even more marked evaporation behavior.

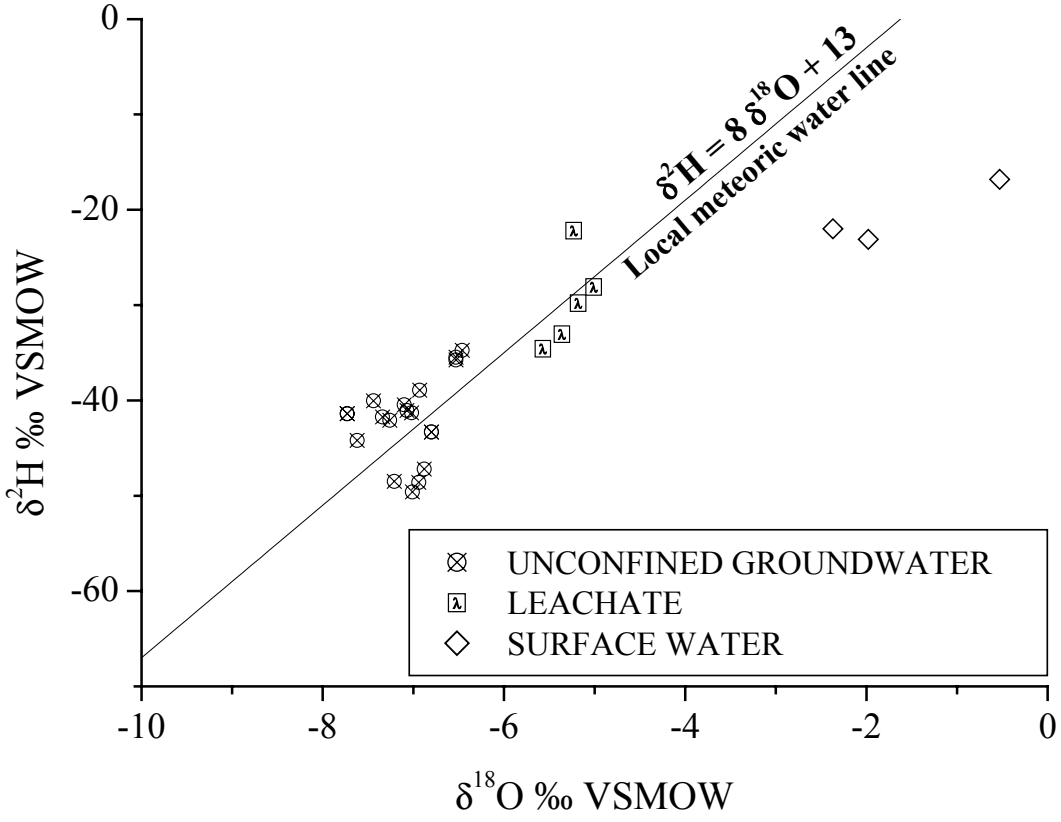

FIG. 7. $\delta^{18}O$ and $\delta^{2}H$ values in landfill area of Ancona Province. The values concern with phreatic groundwater, surface water and leachate.

In the older Landfill 2, abandoned in 1986 (FIG. 6 b), the tritium content of leachate is nowadays only about 30 TU, as most tritium has been washed out by the percolating precipitation. However, in the deep well located in the perimeter of the landfill, the tritium activity is higher than the present natural level, probably indicating contamination from the landfill deposits above. The stable isotopes indicate that the recharge of this groundwater derives from precipitation at an elevation of 500-600 m a.s.l. In fact, other wells in the area which are recharged by local precipitation, have a $\delta^{18}O$ value close to -5‰ (TAZIOLI A. et alii, 2003).

Several other landfills have been investigated in the zone, showing no significant contamination of the

surrounding groundwater, whose tritium content is within the limits of the natural base levels (4-6 TU). In these landfills, also the leachate tritium values are no higher than 15-20 TU. The investigations in them is ongoing with periodical measurements of electrical conductivity, pH, temperature (T °C), and isotopic and tritium content of groundwater and leachates.

Table 3 $\delta^{18}O$, $\delta^{2}H$ and Tritium in landfill areas (Ancona Provinces).

LANDFILL	Sample	$\delta^{18}O$ ‰ VSMOW	$\delta^{2}H$ ‰ VSMOW	^{3}H (TU ± 1)
Landfill n. 1	Leachate 19/11/2002	-5.23	-22.2	291,2
	well P3 -5m	-6.53	-35.75	8,4
	well P4 -5m	-6.46	-34.75	10,4
	well P5 -1m	-7.73	-41.4	12,7
	well P5 -7m	-	-	11,3
	well P6 -7m	-	-	10,2
	well P7 -7m	-6.80	-43.3	4,1
	well P8 6m	-5.60	-31.75	10,7
Landfill n. 2	Leachate 9/07/02	-7,55	-45,03	29,9
	Leachate 7/05/02	-	-	28,6
	External well	-7,02	-41,3	8,2
	Internal well -13m	-7,34	-41,7	20,2
	Internal well -9m	-	-	14,1
	Internal well 26/03/02	-	-	11,3
Landfill n. 3	Leachate 21/10/01	-5,57	-34,58	13,2
	Leachate 26/03/03	-6,89	-40,71	14,6
	Leachate 7/05/03	-	-	11,1
	Leachate 9/07/03	-	-	12,0
	upper well 4 31/10/01	-8,42	-54,24	8,9
	upper well 3 7/11/01	-	-	12,1
	upper well 1 31/10/02	-6,47	-41,1	8,2
	upper well 2 7/11/02	-	-	11,5
	well P1 12/11/2002 -	-7,26	-42,08	14,6
	well P2 26/03/02	-6,92	-38,92	12,1
	well P2 7/05/02	-	-	10,6
	well P2 9/07/02	-	-	10,9
	well P3 19/11/2002 -	-7,07	-41,06	20,5
	well P3 9/07/03	-	-	16,5
Landfill n. 4	Leachate 26/03/02	-5,36	-33,06	10,2
	river	-5,83	-35,46	-
Landfill n. 5	well 1 31/10/01	-7,26	-42,09	8,8
	well 2 31/10/02	-6,93	-39,92	9,9

Concluding remarks

The use of chemical and isotopic methods has produced good results for the control of the interference between landfill activity processes and the underground groundwater environment. The results obtained from the investigated areas show that an accurate knowledge of the background values of the environment in which the landfill is located is basic and enables us to establish a safe monitoring of its activity.

Oxygen-18, deuterium and tritium data have allowed groundwater residence times and location of recharge areas to be identified. In particular, the use of tritium as a pollution tracer may provide information on the presence of landfill contamination much earlier than chemical changes of the standard pollution parameters. The deuterium and oxygen-18 of leachates always appear to be somewhat different with respect to the groundwater values, but they can be used as contamination index only in cases of high fractions of leachate. Tritium is usually much more useful as a contamination index, and enables us to clearly identify the contamination deriving from mixing with landfill leachate.

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