

The groundwater pollution in Lombardy (North Italy) caused by organo-halogenated compounds

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Summary. - This paper deals with the phenomenon of the presence of organo-halogenated compounds in groundwaters of the Lombardy Region (North Italy). The regionwide study evidenced the magnitude of the phenomenon, since these compounds are employed in all productive and household activities. The main cause of groundwater contamination is the infiltration of industrial wastewater: in the Province of Mantova, for example, organic chlorinated solvents have their origin in the NaOCl wastewater treatment for ammonia removal. Organic halogenated compounds in waters intended for human consumption in Lombardy are present in 510 wells over 92 townships, affecting a population of 1,934,133 equivalent to 20% of the total resident population (1991 data). Maximum observed concentrations are related to trielin and tetrachloroethylene. Water treatment was achieved through aeration (stripping) and activated carbon or resin adsorption; in a few instances, also hydraulic interventions were implemented.

Key words: groundwater pollution, organo-halogenated compounds, remedial measures.

Riassunto (*La contaminazione delle acque sotterranee in Lombardia causata da composti organoalogenati*). - La ricerca, estesa a tutta la Regione, ha messo in evidenza la diffusione del fenomeno nelle aree fortemente industrializzate e densamente urbanizzate, poiché questi composti sono diffusi in tutte le attività produttive ed in quelle domestiche. La causa principale della contaminazione delle acque sotterranee è l'infiltrazione di acque di origine industriale: in provincia di Mantova i solventi organoclorati trovano origine nel trattamento delle acque NaOCl per la rimozione dell'ammoniaca. I composti organoalogenati nelle acque destinate al consumo umano in Lombardia interessano (dati 1991) 510 pozzi distribuiti su 92 comuni ed una popolazione di 1.934.133 abitanti, pari al 20% del totale. Le concentrazioni massime riportate si riferiscono alla trielina e al tetracloroetilene. Il trattamento delle acque è stato realizzato mediante aerazione (strippaggio) e mediante adsorbimento su carboni attivi o su resine ed in alcuni casi si è provveduto anche con interventi idraulici.

Parole chiave: contaminazione acque sotterranee, composti organoalogenati, misure di bonifica.

Introduction

Organo-halogens were first detected in Italian drinking water in 1975 when triand tetrachloroethylene were found in some water supply zones of Milan. Since then it has been shown that this problem is widespread particularly in heavily industrialized and densely urbanized areas. This present study examines the situation in the Lombardy Region from 1975 to the present time and describes the protective and remedial measures introduced since then.

Nature and classification of organo-halogenated compounds

In 1982 the European Economic Community [1] set a list of 129 substances that can be potentially hazardous to aquatic environment and human health. Among such chemicals there are 118 organic substances which include several organo-halogenated compounds.

This class comprises a large number of chemicals which represent more than 50% of the water pollutants detected in a study in an industrial area in Lombardy. They are responsible for perhaps as much as 90% of this pollution in terms of concentration [2].

There is no universally accepted classification of organo-halogenated compounds, however they can be divided into some general classes:

- on the basis of their physico-chemical properties (molecular weight, volatility, solubility, biological degradability) and of their interaction within the soil and the aquifer they contaminate, as suggested by the UK Department of the Environment [3];

- on the basis of extraction and concentration systems (purge-and-trap, AOX, EOX, etc.) and considering detection and metering methods;

- on the basis of their toxicity (carcinogenicity). For several compounds an assessment of carcinogenesis is available, made by various agencies and international organisations, for example EEC, WHO, CCTN (National Consultative Commission on Toxicology in Italy), EPA;

there are also evaluations of carcinogenic hazard (Reference Risk) made by WHO and EPA and maximum acceptable concentrations and guideline values prepared by Germany, United Kingdom, WHO and EPA [4];

- on the basis of their nature and chemical composition, as suggested by the Authors (Table 1):

1) aliphatic halogenated hydrocarbons: they are essentially halogenated compounds of methane, ethane, propane and ethylene;

2) aromatic halogenated hydrocarbons: the very common compounds are the chlorinated benzenes;

3) other halogenated compounds (for example: pesticides).

Studying water pollution, it is usual to refer either to "volatile halogenated hydrocarbons" (VOX) or to "volatile organic compounds" (VOC) which have their boiling point between 40 °C and 120 °C. They are generally employed as solvents and are often called "organo-chlorine solvents" [5]. Methane derivatives are named "trihalomethanes" (THM).

All these substances belong to an homogeneous group of liquid non-mixable, refractory, non-retained in permeable soils and toxic compounds.

The major chemical and chemico-physical properties of these compounds are summarized in Table 2.

Industrial use of organo-halogenated compounds

Various organo-halogenated substances are produced by the chemical industry and employed in industrial, small industrial, agricultural and domestic activities.

The Italian consumption of the most significant organo-halogenated compounds is summarized in Table 3, with the main uses [6].

The main uses are:

- the chemical and pharmaceutical industries employ such compounds as solvents in extractive processes;
- the pickling of metals in which trichloroethylene, perchloroethylene and trichloroethane are used; their consumption in Italy amounts to 56 t/y;

Table 1. - The commonest organo-halogenated compounds

Parameter	EEC number	Formula
Halogenated aliphatic hydrocarbons		
<i>Derivative C₁</i>		
Dichloromethane (methylchloride)	62	CH ₂ Cl ₂
Chloroform (trichloromethane)	23	CHCl ₃
Bromoform (tribromomethane)		CHBr ₃
Bromodichloromethane THM		CHBrCl ₂
Dibromochloromethane		CHBr ₂ Cl
Carbon tetrachloride (tetrachloromethane)	13	CCl ₄
Freon 11 (fluorotrichloromethane)		CCl ₃ F
Freon 12 (Difluorodichloromethane)		CCl ₂ F ₂
<i>Derivative C₂</i>		
1,1 dichloroethane	58	CHCl ₂ -CH ₃
1,2 dichloroethane	59	CH ₂ Cl-CH ₂ Cl
1,1,1 trichloroethane (methylchloroform)	119	CH ₃ -CCl ₃
1,1,2 trichloroethane	120	CH ₂ Cl-CH ₂ Cl
Vinylchloride	128	CHCl=CH ₂
1,2 ethylenedichloride	61	CHCl=CHCl
Trichloroethylene	121	CHCl=CCl ₂
Tetrachloroethylene	111	CCl ₂ =CCl ₂
<i>Derivative C₃</i>		
1,2 dichloropropane	65	CH ₂ Cl.CHCl.CH ₃
1,3 dichloropropane	66	CH ₂ Cl.CH ₂ .CH ₂ Cl
Aromatic halogenated hydrocarbons		
Chlorobenzene	20	C ₆ H ₅ Cl
1,4 dichlorobenzene	55	C ₆ H ₄ Cl ₂
1,2,4 trichlorobenzene	118	C ₆ H ₃ Cl ₃

Table 2. - Main chemical and physico-chemical characteristics of organo-halogenated compounds

Compound	Formula	Molecular weight	Density (g/cm ³)	Boiling point (°C)	Vapor pressure (mm Hg)	Octanol-water coefficient (log K _{oc})	Henry's constant (conc. in air/ conc. in water)	Threshold odor concentration (mg/l)
Dichloromethane (Methylchloride)	CH ₂ Cl ₂	85	1.32	40.1	440			
Chloroform (Trichloromethane)	CHCl ₃	137	1.48	61.7	200	2.53	2.53	
Bromoform (Tribromomethane)	CHBr ₃	253		149.5	5.6			
Bromodichloromethane THM	CHBrCl ₂		1.98	90				
Carbon tetrachloride	CCl ₄	154	1.59	76.7	99 (20 °C)		1.2	
1,1 dichloroethane	CHCl ₂ -CH ₃	99	1.25					2,000
1,2 dichloroethane	CH ₂ Cl-CH ₂ Cl	99	1.24	83.5	82		0.05	
1,1,1 trichloroethane (Methylchloroform)	CH ₃ -CCl ₃	133	1.34	74.1	100 (20 °C)	2.49	0.17	
1,1,2 trichloroethane	CH ₂ Cl-CHCl ₂	133	1.44	113.7	19 (20 °C)			
1,2 dichloropropane	CH ₂ Cl-CHCl-CH ₃	113	1.15	95.5	40			
1,3 dichloropropane	CH ₂ Cl-CH ₂ -CH ₂ Cl							
Chloroethene (Vinylchloride)	C ₂ H ₃ Cl	62	0.92	-14	2,660		50	
1,1 dichloroethylene	CHCl = CHCl	97	1.22		495			
Trichloroethylene	CHCl = CCl ₂	132	1.46	86.7	74	2.29	0.48	500
Tetrachloroethylene	CCl ₂ = CCl ₂	166	1.62	121	14 (20 °C)	2.6	1.1	300

Table 3. - The Italian market (1983-1985) for the most important organo-halogenated compounds in different segments

Compounds	t/y	%
Dichloromethane		
Aerosols (co-solvent or co-propellant)	14,850	33
Process solvent in chemical and pharmaceutical industries	13,500	30
Paint industries	4,050	9
Special resins solvent	12,600	28
Total	45,000	100
Perchloroethylene		
Dry cleaning (also considering industrial degreasing and degreasing hides)	30,000	70
Pickling of metals etc.	12,000	27.5
Solvent for rubbers and special components	1,000	2.5
Total	43,000	100
1,2 dichloropropane, 1,2 dichloroethane and similar		
Paint industries, denaturation of solvents	24,000	80
Stain-removers (for civil and industrial use)	3,000	10
Pharmaceutical and phytopharmaceutical industries	3,000	10
Total	30,000	100
Trichloroethylene		
Pickling of metals etc.	20,000	80
Dry cleaning (see above)	3,000	12
Solvents for extractive processes	2,000	8
Total	25,000	100
1,1,1 trichloroethane		
Hot pickling of metals	8,400	35
Cold pickling of metals	12,000	50
Formulation solvent for adhesives, tanners, inks, textiles etc.	3,600	15
Total	24,000	100
Chloroform		
Extractive processes in pharmaceutical industry; laboratory uses	1,000	100
Carbon tetrachloride		
Pharmaceutical and chemical laboratory uses; for insecticides and fumigants formulation	250	100
1,2,2 trichloroethane, 2,1,1 trifluoroethane (Freon 113)		
Pickling in electronic industry	2,000	100
TOTAL	170,000	

- the paint industry where dichloropropane, trichloroethylene, perchloroethylene, trichloroethane and dichloromethane are used as denaturing agents, thinners and paint removers;

- domestic and industrial stain removers consist of 2/3 dichloropropane and dichloroethane, 1/3 trichloroethylene. The total market is 4,000 t/y.;

- dry cleaning of clothes and textile materials where the most commonly used solvent is perchloroethylene, with 30,000 t/y, against 1,300 t/y of trichloroethylene; the latter together with Freon 113, is also employed to degrease raw hides before tanning;

- aerosols which use about 15,000 t/y of solvents, especially dichloromethane. The Montreal protocol is an international agreement to reduce chlorofluorocarbons in aerosols, mostly CFC compounds.

Source and movement of pollution

Volatile halogenated hydrocarbons are found widely in industrialized nations; and they can be considered ubiquitous groundwater pollutants.

There are two main causes of groundwater pollution caused by these compounds. The first is the seepage of industrial wastewater, in particular the direct disposal of spent pickling liquor in pits [7].

The scientific basis needed to understand the behaviour of halogenated-hydrocarbons in aquifers is essentially hydraulic and chemical. The factors affecting their movement are: the nature and the quantity of pollutant substances, the aquifer characteristics, the polluted area, etc. [8]. The mechanism of adsorption, dispersion and degradation is particularly important [9].

New studies show that volatile halogenated hydrocarbons are quickly leached into the groundwaters. Most of these compounds are highly refractory to chemical and biological hydrolysis, but, for some volatile halogenated hydrocarbons, biological transformation is very important, because they give origin to sulfur volatile compounds in anaerobic conditions [10].

Analytic techniques

Currently the most commonly used method of determining "chlorine solvents" is based on the technique of head-space analysis in standard vials. The gaseous phase sample is injected in a gas chromatograph equipped with an electron capture detector (ECD). The capillary glass column (Chromopack) is maintained at room-temperature (20 °C). The method was described by Cavallaro and Grassi [11] while a critical examination of the method in relation to the parameter no. 32 of DPR 236/88 was recently made by Milani [12].

Because of the relatively low sensitivity of the above method, especially for dichlorinated compounds, some analyses were carried out with the stripping method described by Grobe [13].

Organo-halogenated compounds in drinking water: the case of the Lombardy Region

Lombardy has an area of 23,857 km², 47% on the plane, 40.6% mountainous and 12.4 hills. The density of inhabitants is of 372/km², while the national average is of 189/km². The census, made in 1986, shows a regional population of 8,874,893 inhabitants. These people are 15.6% of the national total and they are distributed among 1,546 municipalities. About 45% of the Lombard population lives in the Milan District.

The first research carried out in the Lombardy Region on organo-halogenated compounds, was in 1975, when trichloroethene and tetrachloroethene were found in some wells of the Milanese aquifer [11].

This research was then extended to all treatment works of the aquifer, and enlarged to cover the determination not only of trichloroethylene and tetrachloroethylene, but also chloroform, 1,1,1 trichloroethane (methylchloroform) and carbon tetrachloride.

Because there were no standards for these individual solvents the Scientific Commission, which was established, fixed the maximum admissible concentration (MAC) at 250 µg/l of total solvents. The results showed that 15% of samples were over this limit value [14].

The increasing contamination found in wells of the Milan aquifer from 1979 to 1982 is summarized in Table 4.

In this time research on organic chlorinated volatile compounds was extended to all the District and so a very high pollution in the North of the city was found.

EC Directive number 778 (1980) does not give a MAC for parameter 32 (organo-halogenated compounds not included in parameter 55 pesticides). In 1988 Italy with statute DPR no. 236 set a MAC of 30 µg/l for this parameters to be met by 8 May 1991. At this date the limit was not met in many parts of the country. Indeed some regions granted derogations on the basis of special Ministerial Decrees to allow higher values than the above MAC. For the Lombardy Region a value of 50 µg/l was set as an annual average of at last six bimonthly samples (Decree of Ministry of Health, 8 May 1991).

It should also be noted that the National Health Council proposed limits for individual compounds non obligatory similar to other EC states. The standards for halogenated organic substances set by a number of EC and other countries are given in Table 5.

On the basis of fixing the maximum admissible value (MAV) at 50 µg/l, the situation in 1991 in Lombardy is summarized in Table 6.

The number of the Municipalities concerned by the presence of halogeno-derived compounds (chlorinated solvents) which exceed the set value is 92.

The affected population amounts to as much as 1,934,133 inhabitants, that is 20% of the total.

Table 7 summarizes the maximum values for single compounds in wells of Milan District, supplying more than 50% of population.

Table 4. - Trends of pollution in wells of the Milan aquifer (intervals of concentrations, ppb)

Wells	< 50	50-100	100-150	150-200	200-250	250-500	500-1,000	1,000-1,500	> 1,500	Activated wells (no.)	Date of sample
no.	197	118	76	54	42	60	14	1	-	557	3-5-1979
%	35.4	20.3	13.7	9.7	7.5	10.8	2.5	0.2	-		
no.	200	109	83	56	36	60	14	1	-	559	1-6-1979
%	35.8	19.5	14.9	10	6.4	10.7	2.5	0.2	-		
no.	197	103	93	65	45	52	12	1	-	568	13-2-1980
%	34.7	18.1	16.4	11.4	7.9	9.2	2.1	0.2	-		
no.	198	104	96	66	49	49	10	2	1	575	30-6-1980
%	34.45	18.08	16.7	11.4	8.53	8.53	1.73	0.34	0.17		
no.	224	112	93	65	54	52	9	1	-	610	30-6-1981
%	36.9	18.5	15.2	10.5	8.85	8.4	1.5	0.15	-		
no.	237	123	90	79	65	17	8	-	2	621	30-6-1982
%	38	19.8	14.5	12.8	10.5	2.7	1.3	-	0.3		

Table 5. - Concentration limit values of main organo-halogenated compounds in different Countries ($\mu\text{g/l}$)

Compound	Italy (a)	Lombardy Region (b)	France (c)	United Kingdom (d)	Germany (d)	Belgium (d)	Ireland (d)	Luxemburg (d)	USA (e)	Switzerland (f)	Canada (g)
Parameter 32 80/EEC/788	30	50				20	100	50	30	25	
Carbon-tetrachloride			3	3	3				5		5
1,2 dichloro-ethane			10						100		
1,1 dichloro-ethylene									7		
trichloro-ethylene			30	30	25				5		
tetrachloro-ethylene			10	10	25						
trichloro-methane (chloroform)			30								
trihalo-methanes				100					100		350
1,1,1 trichloro-ethane					25				200		
tetrachloro-ethane					25				5		
dichloro-methane											50

(a) DPR no. 236/1988; (b) Decree of Ministry of Health 8-5-1991; (c) Circular 16-5-1989; (d) see [4]; (e) US Draft proposal of Drinking water standards 1988 (maximum contaminant level, mg/l); (f) Federal Office for Public Health, Berna, June 1981; (g) J. Hickmann *et al.*, *Drinking water treatments*, Oxford 1989.

Table 6. - Organo-halogenated compounds present by concentration, in wells of municipalities in Lombardy

District	no. of municipalities with levels above 50 µg/l	Activated wells	Controlled wells	no. of wells by interval of concentrations				
				<1	1-30	30-50	50-100	>100
BG	3	9	7	0	4	3	0	0
BS	5	14	14	0	3	7	2	2
CO	7	25	25	6	7	8	1	3
CR	-	-	-	-	-	-	-	-
LE	-	-	-	-	-	-	-	-
LO	1	-	-	-	-	-	-	-
MN (*)	14	30	30	0	30	0	0	0
MI	47	421	376	10	189	88	63	26
PV	2	5	5	0	3	0	2	0
SO	-	-	-	-	-	-	-	-
VA	13	53	53	2	31	14	2	4
Total	92	557	510	18	267	120	70	35

(*) Difficult to characterise because of mixing of supplies.

Table 7. - Maximum concentration values of organo-halogenated compounds present in Milan District wells

Compound	Maximum value (µg/l)
Tetrachloroethylene	1,112
Trichloroethylene	4,000
Chloroform	161
Carbon tetrachloride	60
1,1,1 trichloroethane (methylchloroform)	171

From Table 7 it is possible to observe that trichloroethylene and tetrachloroethylene, used both for metal pickling and dry cleaning, have the highest concentration, 4,000 and 1,112 µg/l respectively.

Aquifers protection and remedial treatment

The Lombardy Region planned a regulatory framework for groundwater protection.

This regulation system started with the issuing of the Regional Law of 10 September 1984, no. 53 "Urgent interventions about drinking water supply, for the reclamation and protection of groundwater" and it was integrated with the Regional Law of 27 May 1985, no. 62 "Regulation of urban wastewater and public sewage discharge. Protection of groundwater from pollution". This law proposes and improves the previous rules, making them more effective.

It is a complex rule set and it deals with the problem of the aquifer from two directions:

- 1) protection of waters from pollution;
- 2) their reclamation for drinking use (when they are not used for drinking waters because of the presence of pollutants).

The rules concerning the first aspect can be defined preventive, regulating urban and industrial wastewater discharge and public sewage discharge (these two last aspects are also subject to national rules), which impact heavily on groundwater pollution.

In the second case the Districts, delegated by their Regions, are required to find the pollution sources, to define the extent of the phenomenon, to stop the discharge of pollution and to reclaim the aquifers downflow.

In this programme about 40 investigations and intervention projects were approved, most of them looking at pollution by organo-halogenated compounds.

The technique used in Lombardy to protect the aquifers and for the reduction or removal of organo-halogenated compounds are the following:

Hydraulic systems. - In the saturated zone, where there is flow in the aquifers, continuous pumping from contaminated wells creates hydraulic discontinuity and prevents the pollution flow reaching other wells.

This is the the most frequently used approach until now. When pollutants are fixed in the unsaturated zone, there are two ways of intervention:

- to accelerate the seepage and collect polluted waters in a decontamination well;

- to strip organo-halogenated compounds by means of air suction from unsaturated zone, based on the high volatility of these compounds. The air extracted from soil must be treated, for example, by means of activated carbons filters.

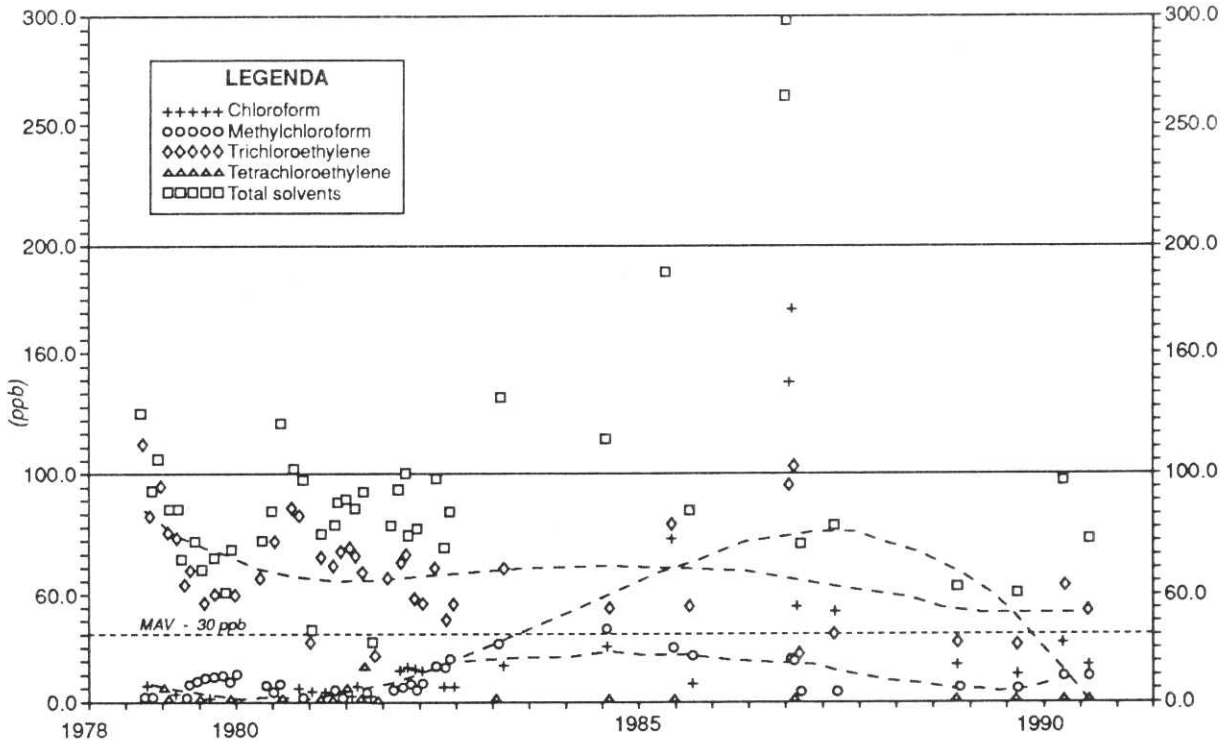


Fig. 1. - Chlorinated solvents: distribution and trends.

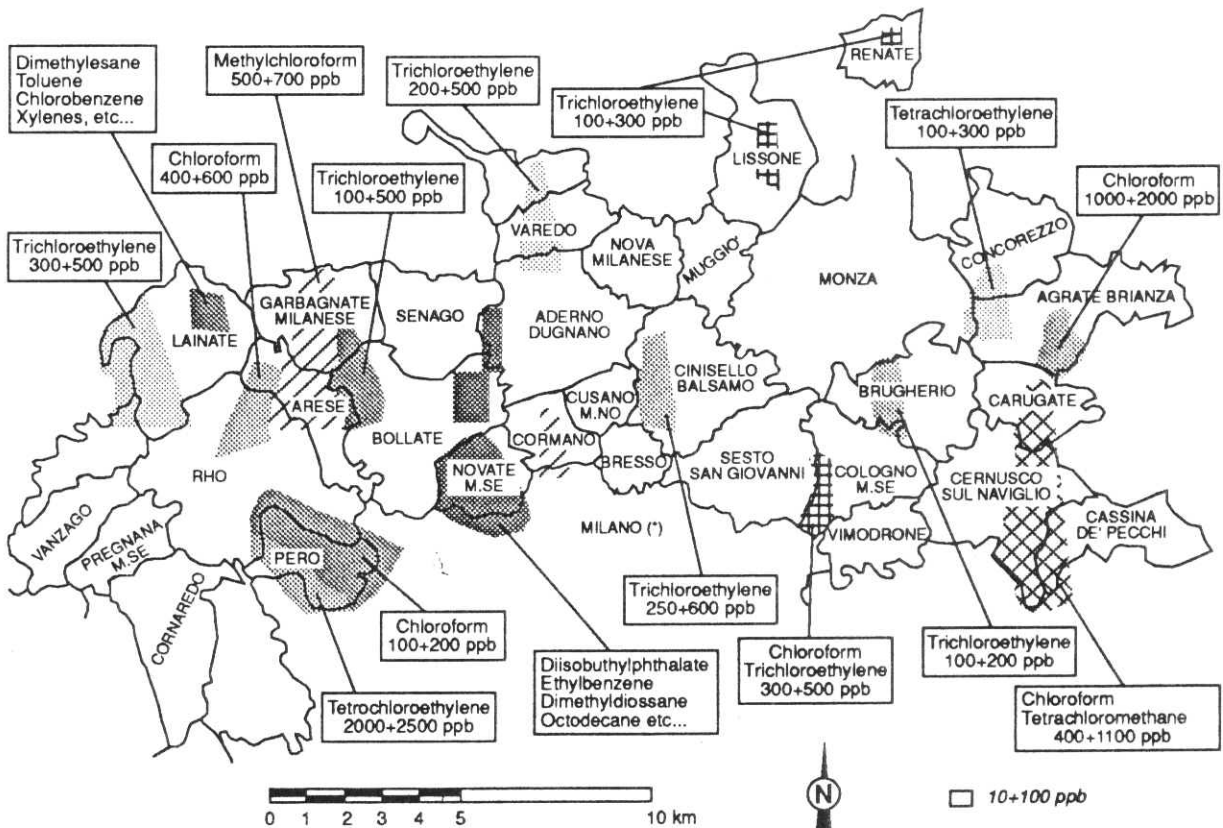


Fig. 2. - Organic compounds in industrialized areas in North Milan.

Table 8. - Removal efficiency of some organo-chlorinated compounds by means of aeration

Compound	Nebulization (%)	Aeration (%)
Chloroform	35-55	-
Methylchloroform	30-45	-
Carbon tetrachloride	23-30	50
Trichloroethylene	25-40	65
Tetrachloroethylene	23-37	60

This approach is being used in a highly industrialized area in North Milan (more than 1,000 industries included in the census). This area is contaminated by chlorinated solvents, the distribution of which is shown in Fig. 1.

In this diagram it is possible to see the trends of the concentrations of different compounds in one of the four wells from upflow to downflow along the ground water direction. The above mentioned area is reclaimed by means of:

1) insulating wells which avoids diffusion of the pollutants in the aquifer;

2) soil reclamation, stripping pollutants from the unsaturated zone.

Treatment of contaminated water. - When raw waters, polluted by organo-halogenated compounds, are used for drinking, they must be treated.

At present, two kinds of processes are applied:

- air stripping of volatile compounds;
- activated carbon adsorption.

The technological aspects of these two systems are well known. They are applied widely and were also tested in Lombardy for removing industrial organic compounds from groundwater.

In the first instance removal was based on air stripping by means of either water nebulization or water aeration [15, 16]. This technique was applied to waters contaminated by tetrachloroethylene (1,200-1,400 µg/l), trichloroethylene (100-750 µg/l), methylchloroform (250-750 µg/l), chloroform, carbon tetrachloride and tetrachloroethylene in equal proportions (250-600 µg/l), trichloroethylene and tetrachloroethylene (100-400 µg/l). These waters were pumped from wells situated in the North Milan area (Fig. 2).

The removal efficiency for each compound is summarized in Table 8. It depends on various parameters.

They are:

- the ratio: treated flow (m³/h) versus volume (m³) of nebulization chamber;
- the ratio between pollutant concentrations in treated water and pollutant concentrations in raw water;
- the speed of air exchange (in nebulization chamber, vol/h);

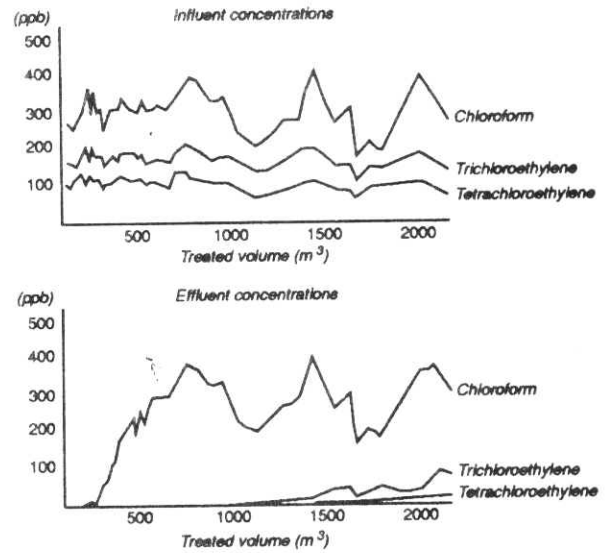


Fig. 3. - Treatment tests of a mixture of organo-halogenated pollutants.

Carbon volume: 122 l.

Filtration speed: 10.2 m/h.

Apparent contact time: 15 min.

- the specific efficiency of treatment, defined as the ratio of concentration of pollutants downflow versus concentration upflow for that specific treatment;

- the ratio between recycled flow and effluent flow.

Activated carbons are usually employed for the removal of organic micropollutants and they are specifically used for treatment of organo-halogenated solvents [17-21].

Fig. 3 shows the behaviour of a water from a well located in north Milan with a concentration of 500-730 µg/l of chlorinated solvents with the following average constitution: chloroform 50%, trichloroethylene 30%, tetrachloroethylene 18%, methylchloroform 1%, Freon 113 1%.

This water was treated with activated carbon, however there was a decrease in activated carbon adsorption capacity for each compound compared to what it would have been if present alone demonstrating competition for adsorption sites.

Conclusions

The presence of organo-halogenated compounds in drinking waters is of concern to drinking water suppliers, because there are no clear regulations and the MAC vary between different countries.

Investigations in Lombardy demonstrated that groundwater pollution by organo-halogenated compounds was widespread. The compounds usually

present are: dichloromethane, tetrachloroethylene, trichlorotrifluoroethane (Freon 113), dichloropropane, chloroform and carbon tetrachloride. They are especially found in very highly industrialized and urbanized areas, and derive from chemicals disposed of either on the soil or in pits. The Mantua District is an exception: here the organo-halogenated compounds derive from treatment of raw waters with sodium hypochlorite.

The steps taken to protect the groundwater and to reclaim aquifers have been effective. With the aid of a very precise regulations, these actions will certainly help to improve the quality of drinking groundwaters.

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