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Results of a survey on asbestos fibre contamination of drinking water in Tuscany, Italy

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ABSTRACT: The extent of asbestos contamination in drinking water and its relationship with water aggressiveness in the distribution network in Tuscany was investigated. Fifty-nine samples of drinking water were collected and analysed for asbestos content by scanning electron microscopy (SEM) and X-ray dispersive microprobe. Every sample was also analysed to determine water aggressiveness. Asbestos fibres were detected in 24% of samples, at concentrations which were always lower than 0.04 million fibres/L. In 79% of these samples, asbestos fibres could have been released into the water from asbestos-cement pipes; in the remaining 21% they may have originated from asbestos-containing aquifers. 43% of the waters were non-aggressive, 54% moderately aggressive and 3% highly aggressive. Samples of aggressive water taken from asbestos-cement pipes were too few to determine any significant correlation between aggressiveness of drinking water and release of asbestos fibres from asbestos-cement pipes.

INTRODUCTION

'Asbestos' is a common word which is used to identify a class of naturally occurring fibrous silicates. There are two main groups of asbestos: the serpentines (chrysotile) and the amphiboles (crocidolite, amosite, tremolite, anthophyllite and actinolite). It has been one of the most widely used materials in industry this century, used in ship and car manufacturing, construction and electrical, chemical, textile and food industries. This is due to its particular properties: it is fire resistant, can provide thermal and electrical insulation, and can be woven. Asbestos is also a material highly resistant to dissolution and biological removal.

Asbestosis, lung cancer and malignant mesothelioma are serious diseases associated with asbestos exposure, and in particular with the inhalation of very thin asbestos fibres. The International Agency for Research on Cancer (IARC) has classified asbestos among the carcinogenic agents for humans (group 1) [1].

In the early 1970s the theory that human ingestion of asbestos may also have a carcinogenic effect was postulated. Consequently, the relevance of asbestos fibres in food, drinks, drugs and drinking water was evaluated [2]. The asbestos fibres, ingested directly or indirectly as a result of inhalation, would penetrate into or through the gastrointestinal mucosa where they might have a carcinogenic effect remaining *in loco* [3]. As yet there is not sufficient evidence for carcinogenicity resulting from ingestion of asbestos contaminated drinking water [4,5]. Positive results have been obtained in few studies; possibly because most of the studies lacked sufficient statistical power to evaluate the risk related to asbestos fibre ingestion and were characterised by a weak definition of exposure [6,7]. The World Health Organisation (WHO) has not defined any threshold limit value for asbestos in drinking water [8], and in Italy, no asbestos quality parameter for drinking water has been defined [9]. Whatever the case, it must be considered that the diffusion of asbestos fibres through drinking water might represent an important source of environmental pollution.

There are several possible routes by which drinking water becomes contaminated with asbestos: mineral deposits released from asbestos-cement pipes of aqueducts, and untreated industrial drains [10–14]. Natural asbestos pollution can be found in spring water or in rivers that run in basins containing asbestos minerals, as was observed in Canada [4,5,15,16]. A large number of aqueducts have been built using asbestos-cement material: it was estimated that there are 2.5 million km of asbestos-cement pipes in the whole world. Five hundred and sixty thousand kilometres of asbestos-cement pipes are in the USA, 257 000 in UK and 125 000 in Italy [17].

Asbestos-cement is a very good material from a mechanical point of view; it contains almost 85% silicates, hydrate calcium aluminate and calcium hydroxide, and 15% asbestos fibres, particularly chrysotile, and/or crocidolite and amosite. The release of asbestos fibres from asbestos-cement pipes into the water they carry seems unlikely unless the pipes are old or worn out. This is particularly true when a protective layer of calcium carbonate is used to cover the internal surface of the pipes. Problems can arise in cases of pipe leakage or of solubility of the cement that bind the asbestos fibres. The microcristalline structure of the binding is characterised by 10–20% porosity; the pores contain water in equilibrium with calcium hydroxide.

Table 1 Characteristics of aqueducts studied

Location of the aqueduct	No. of samples	Estimated length of network (km)	Length of asbestos-cement pipes (km) (% total length)	Type of water	Treatments applied to water	Characteristics of aqueducts *				
						A	B	C	D	E
Arezzo	2	260	n.a.	surface water	coagulation, filtration, chlorination	×			×	
Montevarchi	1	60	n.a.	ground water	chlorination	×				
Florence	7	1.000	10 (1%)	surface water	sedimentation, filtration, chlorination, ozonation	×			×	
San Godenzo	1	n.a.	n.a.	ground water	chlorination					×
Scandicci	2	140	35 (25%)	surface and ground water	chlorination	×			×	
Empoli	2	260	n.a.	ground water	chlorination, air-stripping	×				×
Castelfiorentino	1	n.a.	n.a.	ground water	chlorination, air-stripping					×
Montaione	1	160	n.a.	surface and ground water	sedimentation, filtration, chlorination			×		
Grosseto	1	120	n.a.	surface and ground water	filtration, ozonation, chlorination				×	×
Acquedotto Fiora	2	2000	n.a.	spring water	chlorination, ozonation		×			×
Livorno	2	560	164 (29%)	ground water	filtration, chlorination	×			×	
Piombino	2	132	75 (57%)	ground water	chlorination	×				×
Elba Island	5	60	n.a.	surface and ground water	chlorination	×		×		×
Lucca	1	630	n.a.	ground water	none				×	
Massa	2	453	4 (0.8%)	ground and spring water	filtration, chlorination	×	×		×	×
Carrara	4	118	n.a.	ground and spring water	UV, chlorination	×	×		×	×
Aulla	2	100	n.a.	ground, spring, surface w.	chlorination		×			×
Pisa	2	422	22 (5%)	ground and spring water	chlorination	×			×	×
Montopoli V.no	3	32	32 (100%)	ground water	chlorination	×				×
Pistoia	4	850	n.a.	ground, spring, surface w.	chlorination	×			×	×
Montale	2	80	n.a.	surface and spring water	filtration, chlorination	×				×
Prato	3	450	11 (2,5%)	surface and ground water	filtration, chlorination	×		×	×	×
Acquedotto Vivo	4	45	n.a.	surface water	chlorination	×	×			×
Colle Val d'Elsa	2	105	10 (9.5%)	ground and spring water	chlorination	×				×
Poggibonsi	2	n.a.	n.a.	surface and ground water	sedimentation, filtration, chlorination, ozonation	×				×

* Characteristics of aqueducts relevant for choosing sampling points (A: presence of asbestos-cement pipes; B: possibility of aggressive water conveyed; C: basin with serpentine minerals; D: highy inhabited area served; E: reference sample).
n.a.: information not available.

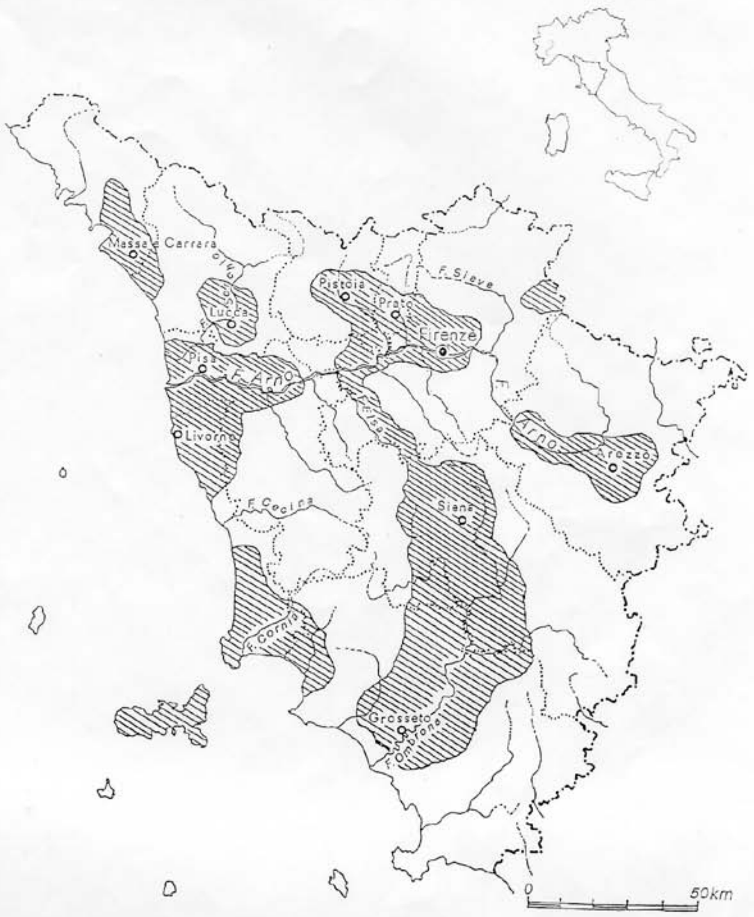

Fig. 1 Map of Tuscany with sampling areas indicated.

If this equilibrium changes due to the solubilisation of the calcium hydroxide through the pores, the asbestos-cement of the pipes is damaged by the gradual dissolution of the calcium hydroxide, increasing porosity, and the progressive dissolution of silicates and hydrate calcium aluminate. This process can produce a reduction in the structural characteristics of the pipe and the release of asbestos fibres [18]. This can happen in particular when the water is aggressive, because it can be corrosive and cause the deterioration of the metallic and cement material of the pipes. In addition to the water type conveyed, fibre release may also be due to the age of the pipe, and its state of decay (this decreases if water stagnation is present) [12,19,20].

In Italy the use of crocidolite in pipes has been prohibited by a regulation in force since 26 June 1986 [21].

While natural and industrial water contamination problems can be solved with adequate treatments (flocculation and filtration reduce 99% of the fibre content of water) [22,23] it is more difficult to identify and solve those problems found downstream from the aqueduct of where large diffusion asbestos-cement pipes are mainly used.

The vast majority of the studies on asbestos water contamination are carried out in the USA and Canada. More than 80% of the towns examined in the United States are served by drinking water with less than 2 million fibres/L (MFL). Eight per cent of towns are served by water with 10 MFL. However, contamination is a possibility and can mean that the water content can reach 1000 MFL [12,24]. In Canada, it has been estimated that 5% of the population receives drinking water with more than 1 MFL, 0.6% with more than 100 MFL [25]. In Europe, the situation has not been studied in depth due to the paucity of geological areas containing asbestos minerals; data on asbestos contamination in drinking water are not as readily available as in North America. The maximum values have been found in Sweden (3.6 MFL), in the UK (2.2 MFL), in the Netherlands (0.24 MFL), and in Germany (0.4 MFL) [12,25]. In Italy, papers on the evaluation of the asbestos content of drinking water are scarce; we therefore thought it important to

Table 2 Results of drinking water sample analysis

Aqueduct	Sample* ID no.	up-stream	down-stream	*AI*	Asbestos concentration in MFL (95% confidence limits for Poisson distribution)	Asbestos type reported
Arezzo	1	×		11.2	BDL	–
	2		×	11.0	BDL	–
Montevarchi	3		×	12.3	BDL	–
Florence	4	×		11.7	BDL	–
	5		×	11.8	0.005 (0.001–0.014)	chrysotile
	6		×	11.8	BDL	–
	7		×	11.8	BDL	–
	8		×	12.0	BDL	–
	9		×	12.0	BDL	–
	10		×	11.9	BDL	–
San Godenzo	11		×	12.1	BDL	–
Scandicci	12		×	12.6	BDL	–
	13		×	12.5	0.002 (0–0.009)	crocidolite
Empoli	14	×		12.6	0.002 (0–0.009)	chrysotile
	15		×	12.2	BDL	–
Castelfiorentino	16	×		12.6	BDL	–
Montaione	17	×		12.6	BDL	–
Grosseto	18		×	11.3	BDL	–
Acquedotto Fiora	19	×		10.6	BDL	–
	20		×	9.7	BDL	–
Livorno	21		×	11.8	0.040 (0.020–0.060)	chrysotile, amosite, crocidolite
	22		×	11.8	0.002 (0–0.009)	chrysotile
Piombino	23	×		12.5	BDL	–
	24		×	12.5	BDL	–
Elba Island	25		×	11.9	BDL	–
	26	×		11.9	BDL	–
	27	×		10.9	BDL	–
	28		×	10.0	0.005 (0.001–0.014)	chrysotile, amosite
	29		×	9.9	0.002 (0–0.009)	amosite
Lucca	30		×	12.2	BDL	–
Massa	31	×		11.4	0.002 (0–0.009)	tremolite
	32		×	11.3	BDL	–
Carrara	33	×		11.6	BDL	–
	34	×		11.7	BDL	–
	35		×	11.8	BDL	–
	36		×	11.5	BDL	–
Aulla	37	×		12.0	BDL	–
	38		×	11.7	0.002 (0–0.009)	tremolite
Pisa	39	×		11.9	BDL	–
	40		×	11.8	BDL	–
Montopoli V.no	41	×		12.4	BDL	–
	42		×	12.5	BDL	–
Pistoia	43	×		12.0	BDL	–
	44		×	12.1	BDL	–
	45	×		12.0	BDL	–
	46		×	11.9	BDL	–
Montale	47	×		11.5	BDL	–
	48		×	11.5	0.002 (0–0.009)	chrysotile
Prato	49	×		11.3	BDL	–
	50		×	12.2	0.002 (0–0.009)	chrysotile
	51	×		12.2	0.02 (0.01–0.03)	chrysotile

Continued

Table 2 (*continued*)

Aqueduct	Sample* ID no.	up-stream	down-stream	*AI*	Asbestos concentration in MFL (95% confidence limits for Poisson distribution)	Asbestos type reported
Acquedotto Vivo	52	×		10.2	BDL	–
	53		×	10.5	0.003 (0.0003–0.011)	chrysotile
	54		×	10.6	BDL	–
	55		×	10.8	BDL	–
Colle Val d'Elsa	56	×		12.0	BDL	–
	57		×	12.1	0.002 (0–0.009)	crocidolite
Poggibonsi	58	×		12.2	BDL	–
	59		×	12.3	BDL	–

*Localisation of sample along distribution system is specified (up-stream sample or down-stream sample);
Aggressiveness Index.
MFL: million fibres/L.
BDL: below detectable limit of 0.002 MFL.

study this problem. The paper describes the results of a 1995–96 survey on the asbestos contamination of drinking water, carried out in Tuscany, Central Italy.

MATERIALS AND METHODS

In Tuscany, drinking water is distributed by many aqueducts, both small and large, that take water from wells, springs, rivers. Different procedures are used to treat the water. The first step of the study has been to collate of information about networks in order to choose the sampling points. Choices were made according to one or more of the following characteristics:

(A) presence of asbestos-cement pipes in the distribution system;

(B) estimated aggressiveness of water conveyed;

(C) basin with serpentine minerals;

(D) aqueduct conveying water to densely inhabited areas.

The Environmental Office of Tuscany Region Administration, according to the Ministerial Decree of 13 December 1991 [26], gathered data on Tuscan aqueducts using a self-administered questionnaire. The available data concerned the presence of asbestos-cement pipes, their year of installation, the pipe length, and the type of water spring. The data are available for 48% of the municipalities, corresponding to 61% of the entire region. Basins with serpentine minerals have been identified using geological maps of the National Geographical Army Institute located in Firenze.

Table 1 summarises the basic information collected about the aqueducts in the study: i.e. number of sampling points, estimated length of network and asbestos-cement pipes, types of water and treatments applied, characteristics relevant for choosing sampling points.

Fifty-nine samples of drinking water, taken in different areas from Tuscany, Italy (Fig. 1), were taken. When available, the samples were taken up-stream and down-stream from the possible source of pollution in order to see if the water characteristics were modified by the pollutants.

The presence of asbestos fibres and the hydrochemical characteristics were evaluated for each sample.

Evaluation of asbestos fibres

The sampling procedures and the analysis of the water sample was made according to EPA (Environmental Protection Agency of USA) procedure [27], modified by the use of scanning electron microscope (SEM).

The sampling procedure and analysis can be summarised as follows:

- drinking water samples were collected in unused, screw-capped, one-litre glass bottles which had been rinsed three times with double distilled water. Samples were stored in the dark and refrigerated at about 4 °C. No preservatives were added.
- Prior to filtering the water, samples were manually and ultrasonically shaken for up to 15 min to ensure a uniform deposition of particulate on the filter. A volume of 200 mL was then filtered through a 0.8 μm pore size Nucleopore® polycarbonate filter of 25 mm diameter. No special treatment to remove organics was required.
- One blank determination was made along with every group of samples prepared at any one time, by filtration of 200 mL of double distilled water.
- The filter was mounted on a stub using double-sided conductive adhesive tape and then coated with a carbon layer on a vacuum evaporator.
- The counting of fibres and the determination of their dimensions were made by a scanning electron microscope. The estimated resolution limit was about 0.05 μm for diameter and 1 μm for length.
- The identification of asbestos type was made primarily by

morphology and then determining the elemental composition of each fibre by energy dispersive X-ray analysis (EDXA) and comparing it with asbestos NIOSH standards (National Institute of Safety and Health of USA).

- A 95% confidence interval on fibre concentrations was calculated assuming a Poisson distribution of fibres on filter.

Evaluation of hydrochemical characteristics and of aggressiveness

For each water sample an analysis was conducted to determine its hydrochemical characteristics: temperature, colour, odour, turbidity, pH, conductivity at 25 °C, calculated fixed residue, total hardness, cationic concentrations (Ca^{++}, Mg^{++}, $NH4^{+}$, Na^{+}, K^{+}), anionic concentrations (Cl^{-}, NO_3^{-}, SO_4^{2-}, NO_2^{-}, HCO_3^{-}), and total organic carbon (TOC).

Some of these parameters were used to evaluate the aggressiveness of each type of water collected. Aggressiveness Index (*AI*) was calculated, from the formula:

$$AI = \text{pH} + \text{Log}(A \times H)$$

where A = total alkalinity in mg/L $CaCO_3$ and H = calcium carbonate hardness in mg/L $CaCO_3$.

Estimates of *AI* values was made by the following criteria:

$AI \leqslant 10 \rightarrow$ aggressive water

$10 < AI < 12 \rightarrow$ moderately aggressive water

$AI \geqslant 12 \rightarrow$ non-aggressive water

The Agressiveness Index is derived from Langelier Index, modified to compensate for the temperature dependency of the solubility product constant of calcite and for ionic strength of the solution. This formulation, elaborated by American Water Works Association (AWWA), and then utilised by Environmental Protection Agency (EPA) and World Health Organisation (WHO), has been adopted by Italian legislation with an *ad hoc* ordinance of the Ministry of Health [28].

RESULTS

The results of the analysis are summarised in Table 2. Samples are identified by a specific number and described as up-stream or down-stream in relation to their location along the aqueduct. For each sample the following parameters are reported: *AI* value, concentration of asbestos in MFL (and relative 95% confidence limits assuming a Poisson distribution) and the type of asbestos detected. The threshold detecting limit is 0.002 MFL, calculated as 1 fibre per 200 mL filtered water in 1 mm^3 filter diameter.

In some water samples gathered from asbestos-cement pipes, concentrations generally between 0.002 and 0.005 MFL were observed; only one sample from the town of Livorno showed a high concentration of fibres (0.04 MFL). In other situations characterised by asbestos-cement pipes, no asbestos fibres were detected.

Regarding spring water, asbestos fibres were detected in only two samples (0.002 MFL) from the Alpi Apuane area. The type of fibre found was tremolite, an amphibolic fibre not often used commercially.

The three samples from the basins located in the provinces of Prato, Livorno and Firenze, characterised by the presence of serpentine minerals, in two samples no fibres were found, and in the sample from Prato the fibre concentration was 0.02 MFL. It must be considered that the sample is of spring water which does not serve any aqueduct.

The samples characterised by aggressive water (with moderate or high aggressiveness) are those from the island of Elba (AI = 9.9–10.0), from Acquedotto del Fiora (AI = 10.2–10.8) and from the province of Grosseto (AI = 9.7–10.6). In three of these samples asbestos fibres were detected (0.005 MFL, 0.003 MFL and 0.002 MFL, respectively).

The water samples from aqueducts serving highly inhabited areas show a concentration of asbestos fibre which is lower than the threshold detection limit, except those from the town of Livorno. The most prevalent type of asbestos was chrysotile (64.3%).

DISCUSSION AND CONCLUSIONS

The instrument that must be used for detecting asbestos fibres in water, according to EPA recommendations, is the transmission electron microscope (TEM). Using this detector it is possible to detect very thin fibres which have a diameter of < 0.1 μm and a length of < 1 μm. The SEM, although less powerful than TEM, still enables us to detect the fibres which are considered to be dangerous to human health (diameter < 1.5 μm and length > 8 μm) [3,29–32]. Moreover, the preparation of the samples is less complex and expensive than for TEM. Therefore, we decided to use SEM for this investigation. However, some water samples were also analysed by TEM in order to obtain a control of the quality of the procedure. The results of this analysis are shown in Table 3: there is substantial agreement between the results from the two different microscopes for all the samples analysed, except no. 39, which was negative using SEM and positive (0.02 MFL) using TEM.

With regard to the aggressiveness determination, several authors have suggested different indexes (Langelier index, Ryznar index, etc.) instead of the Aggressiveness Index we used. The *AI* does not always seem to correctly predict fibre release and internal pipe surface degradation (in particular in waters which are undersaturated with respect to calcite) and for other criticisms [17,33]. In any case it has generally proved more likely to falsely predict pipe deterioration than to predict falsely pipe stability. The *AI* was in fact considered to be the best index by the American Water Works Association and by the EPA, particularly for waters conveyed by asbestos-cement pipes. Moreover, according to present regulations for the determination of aggressiveness in Italy it is the index that has to be used [28], that also prohibits the use of asbestos-cement pipes with aggressive water.

Table 3 Asbestos type and concentrations in water samples analysed by SEM and TEM

Sample	Asbestos concentrations in MFL*		Asbestos type reported	
	SEM	TEM	SEM	TEM
19	BDL	BDL	–	–
20	BDL	BDL	–	–
21	0.04 (0.02–0.06)	0.02 (0.01–0.04)	chrysotile, amosite, crocidolite	chrysotile
34	BDL	BDL	–	–
36	BDL	BDL	–	–
39	BDL	0.02 (0.01–0.04)	–	amosite
52	BDL	BDL	–	–
55	BDL	BDL	–	–

* In parenthesis 95% confidence limits assuming Poisson distribution.
BDL: below detectable limit of 0.002 MFL.

Asbestos fibres were detected in 24% of drinking water samples, with concentrations which were always less than 0.04 MFL. In 79% of positive samples, the causes of contamination were very likely to be due to the release of fibres from asbestos-cement pipes: this hypothesis seems very convincing to us, due to the detection of fibres only in down-stream samples. Asbestos-cement pipes which are in good condition do not pose a risk. However, problems could arise with ageing of the pipes, thus causing dis-aggregation of the linking material. Therefore the asbestos fibre concentration in down-stream water of asbestos-cement pipes might increase in time.

Natural contamination was observed in 21% positive water samples. Only 3% of the drinking water samples analysed were aggressive, too few to be able to draw any conclusions regarding the association between asbestos fibre release from asbestos-cement pipes and water aggressiveness.

In conclusion, the percentage of positive water samples for the presence of asbestos fibres was low and the concentrations of asbestos fibres never exceeded 0.04 MFL.

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