



## Research Paper

# Chrysotile asbestos migration in air from contaminated water: An experimental simulation

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## ABSTRACT

In Naturally Occurring Asbestos (NOA) rich areas, water flows through asbestos bearing rocks and soils and generates waterborne fibres that may migrate in air and become a risk for humans. Research on the migration and dispersion after water vaporisation has been so far only marginally evaluated. This study investigates the migration in air of asbestos from a set of suspensions contaminated by chrysotile from Balangero (Italy), under controlled laboratory conditions. We evaluated i) the morphological modifications that might occur to chrysotile during migration from water to air, and ii) the amount of airborne chrysotile mobilised from standardised suspensions. Morphological alteration of asbestos fibres occurred during water-air migration and impacted on the analytical response of electron microscopy. Waterborne asbestos concentration higher than  $40 \cdot 10^6$  f/L generates in air concentration higher than 1 fibre per litre [f/L], the alarm threshold limit set by World Health Organization for airborne asbestos. A possible correlation between the waterborne fibre concentration as mass or number of fibres per volume unit [ $\mu\text{g/L}$  or f/L] was observed.

## 1. Introduction

Asbestos is a commercial term that applies to a group of six silicate minerals (i.e., chrysotile, tremolite asbestos, actinolite asbestos, anthophyllite asbestos, grunerite asbestos also known as amosite and riebeckite asbestos also known as crocidolite) which occur with fibrous morphology. It is widely known that these minerals, when respired, may induce fatal diseases, such as malignant mesothelioma and lung cancer, in humans.

Despite restrictive regulations, asbestos is still the main occupational risk factor for mesothelioma, a not curable cancer of the respiratory apparatus. The International Agency for Research on Cancer (IARC) has classified all six asbestos minerals as carcinogenic to humans (Group 1) (IARC, 2012). Several studies show the possible outbreak of mesothelioma in humans who live in areas that are rich in asbestos bearing rocks and therefore exposed to naturally occurring asbestos (NOA) released in

air (e.g., Noonan, 2017). The relation between environmental natural exposure and the outbreak of this malignancy is still debated. Therefore, for asbestos, a no-observed-effect level (NOEL) below which mesothelioma risk is negligible has not been set so far.

To reduce the risk for humans, World Health Organization (WHO) Air quality guidelines for Europe (WHO, 2000) indicates a threshold limit of 1 fibre of asbestos per litre of air [f/L] in outdoor urban environment. Furthermore, Italian regulation on asbestos has not yet enforced specific guidelines for outdoor ambient and the Italian Institute for Insurance against Accidents at Work (INAIL, 2010) applies the same limit indicated by WHO (2000).

While a lot is known about the diseases caused by respiration of airborne asbestos and similar asbestiform minerals (e.g., asbestiform fluor-edenite) (Baumann et al., 2013), not enough has been yet comprehended about the potential health risk posed by waterborne fibres and their migration pathways in the environment.

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It is known that two main exposure routes to waterborne asbestos exist: i) ingestion, especially when asbestos is present in large amounts in drinking water or food, and ii) migrated airborne asbestos after water vaporisation. This latter pathway may occur also when asbestos-polluted waters deposit on soil that might be subjected to erosion/ weathering and dust mobilisation.

The hypothesis that ingested asbestos (both chrysotile and amphiboles) could have carcinogenic effects at the level of the digestive system developed at the beginning of the '70s. Many researchers studied this possibility but uncertainties are still present (Di Ciaula, 2017; Di Ciaula and Gennaro, 2016; IARC, 2012; Cantor, 1997; Marsh, 1983; Levy et al., 1976). WHO has not defined a safe concentration level for asbestos in water yet (WHO, 2020). However, United States Environmental Protection Agency (U.S.-EPA) defined a maximum contaminant level of asbestos in drinking water of  $7 \cdot 10^6$  f/L (U.S.-EPA, 2004), referred to fibres longer than  $10 \mu\text{m}$  (U.S.-EPA, 1994).

Few studies have published data on the potential risk due to airborne asbestos and asbestiform minerals released following water mobilisation/vaporisation. This is mainly due to the difficulty to correlate waterborne asbestos concentration with airborne fibres concentration in open, natural environment. One limited indoor study (Webber et al., 1988) identified a correlation between airborne and waterborne asbestos in houses where asbestos (chrysotile and amphiboles) was found in tap water, reporting that  $2.4 \cdot 10^7$  f/L waterborne fibres might generate up to 120 f/L airborne fibres. However, the strong variability between different environments prompted the authors to conclude that each exposure scenario is unique and requires some site-specific evaluations. More recently, Roccaro and Vagliasindi (2018) examined the indoor occurrence of airborne asbestiform fibres when contaminated water was used in humidifiers and showers. They found that the percentage of fibres transferred from contaminated water to air ranged between 0.04–0.07% and 4.3–10.8%, respectively. These data are consistent with those reported by Hardy et al. (1992) which revealed a percentage of transferred asbestos fibres of 0.03% up to 4.7% from water to air, using an ultrasonic humidifier fed by contaminated water.

Water pollution by asbestos is linked to anthropogenic and natural causes. In the countries where the industrial use of asbestos is banned (Europe: EU) or strictly regulated, the anthropogenic causes include the dumping of asbestos-contaminated water from mines and quarries where asbestos and NOA in general might occur. Also, deteriorated asbestos cement pipes may release fibres in water and are an important source of anthropogenic pollution. In addition, the flow of water through asbestos-contaminated waste improperly disposed in non-waterproof sites might also become a source of pollution (Mohanty et al., 2021). Waterborne asbestos is however mainly generated by natural processes. When surface and deep waters flow into rock formations containing asbestos minerals, such as metaophiolites and serpentinite rocks, fibres are weathered and might be mobilised from rock matrix to water.

This work is focused on natural environmental generation of waterborne fibres and three matrices are considered to be in reciprocal relation when NOA dispersion occurs: soil/rocks, air and water. On this basis, asbestos environmental occurrence and potential human exposure should be evaluated considering the possible interactions among these matrices.

Weathering of asbestos-bearing rocks is the principal natural cause of asbestos water dispersion in NOA-settings. Many studies were carried out in areas where NOA in rocks and soils are widespread (Wei et al., 2013; Schreier, 1987; Bales et al., 1984; Hayward, 1984; McGuire et al., 1982; Millette et al., 1983, 1980; McMillan et al., 1977) or in the vicinity of active and inactive asbestos mines (Koumantakis et al., 2009; Anastasiadou and Gidarakos, 2007; Kashansky and Slyshkina, 2002). In these latter cases, up to  $10^{12}$  f/L were detected in surface waters (Schreier, 1987). Fibres were mainly short with length  $< 5 \mu\text{m}$ , but the possible asbestos dispersion in air from stream water was not considered (Schreier, 1987).

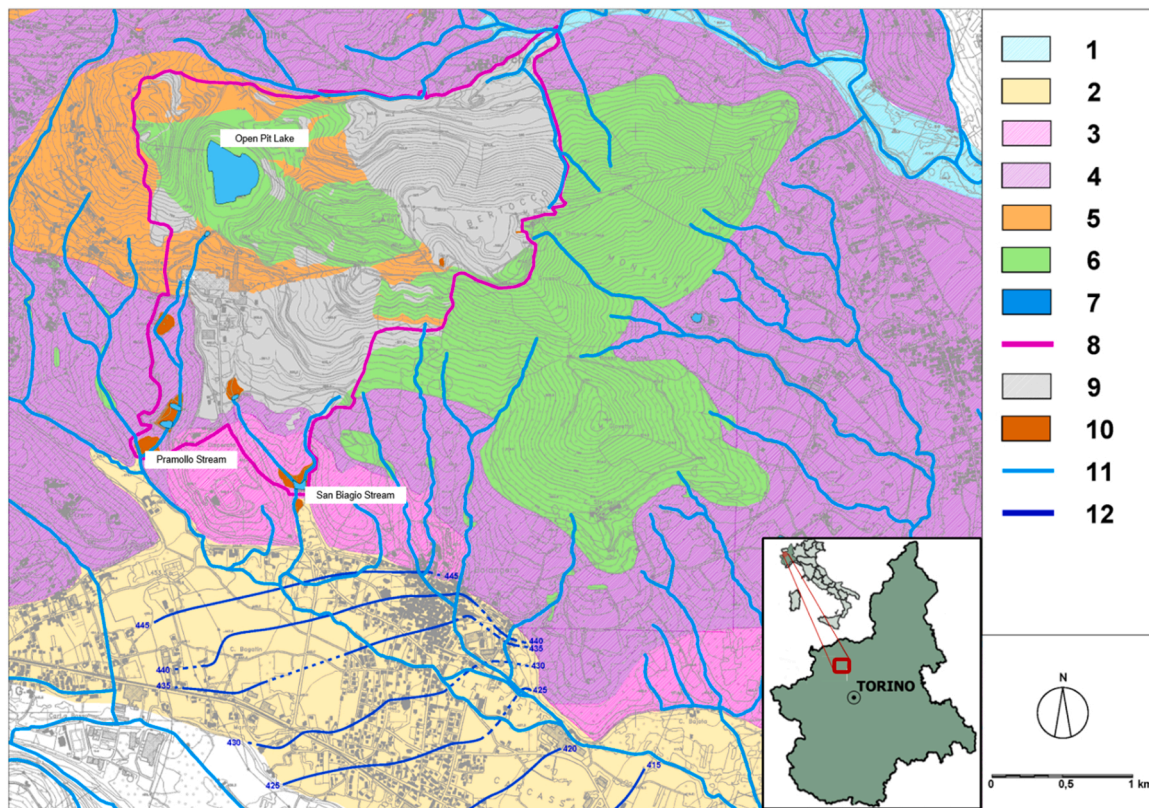
Turci et al. (2016) reported a non-negligible airborne chrysotile and tremolite asbestos occurrence during agricultural activities in a rural area in the surroundings of the former asbestos mine of Balangero and Corio ( $45^{\circ}17'40''\text{N}$   $7^{\circ}30'23''\text{E}$ , North-West Italy, about 30 km north from Turin), where water containing asbestos was used to irrigate a field characterised by ultramafic topsoil. Approximately  $2 \cdot 10^5$  f/L waterborne chrysotile fibres were detected by Scanning Electron Microscopy coupled with Energy Dispersive Spectroscopy (SEM-EDS) in a stream water draining the southern slope of the former mine. When agricultural activities (soil tillage) were performed on a crop watered with that stream, airborne asbestos concentrations up to 40 f/L and 2 f/L were monitored by a personal sampler on the field worker and by an environmental sampler on the border of the field, respectively. Asbestos dispersion and migration from the Balangero mining area have been the object of pioneer studies (Buzio et al., 2000; Caramuscio et al., 1992) and the mine itself still represents a remarkable source of data for research on environmental asbestos occurrence. The former mine is located in a geographical area characterised by partially serpentinized peridotites (e.g., Aoki et al., 2020). The San Vittore Mount ( $45^{\circ}17'32''\text{N}$   $7^{\circ}31'01''\text{E}$ ), an asbestos-rich serpentinitic outcrop, has been exploited over a long period of time for chrysotile asbestos extraction and constituted the largest and most productive chrysotile asbestos mine in western Europe (e.g. Virta, 2005). The mining area is nowadays characterised by an open pit which is partially filled by a lake and whose terraces are subjected to constant weathering and occasional landslides. A network of streams conveys the drainage waters to the plain below, the so called Balangero Plain. In constant interaction with surface waters, groundwater circulation in the mountainous area is guaranteed by surface fracture system, while the Plain is characterised by aquifers flowing in NW-SE direction, hosted by alluvial deposits (De Luca et al., 2020).

The area is monitored by RSA Srl (from now on indicated as RSA), the public company in charge of the remediation and environmental development of the former asbestos mine site of Balangero and Corio municipalities. In the framework of works for safety assessment and improvement, RSA created decantation tanks at the basis of the mine to allow water purification of the principal streams (Rio Pramollo and Rio San Biagio) draining the mining area (see Fig. 1) and collected a great amount of data on asbestos occurrence in the superficial hydrographic network.

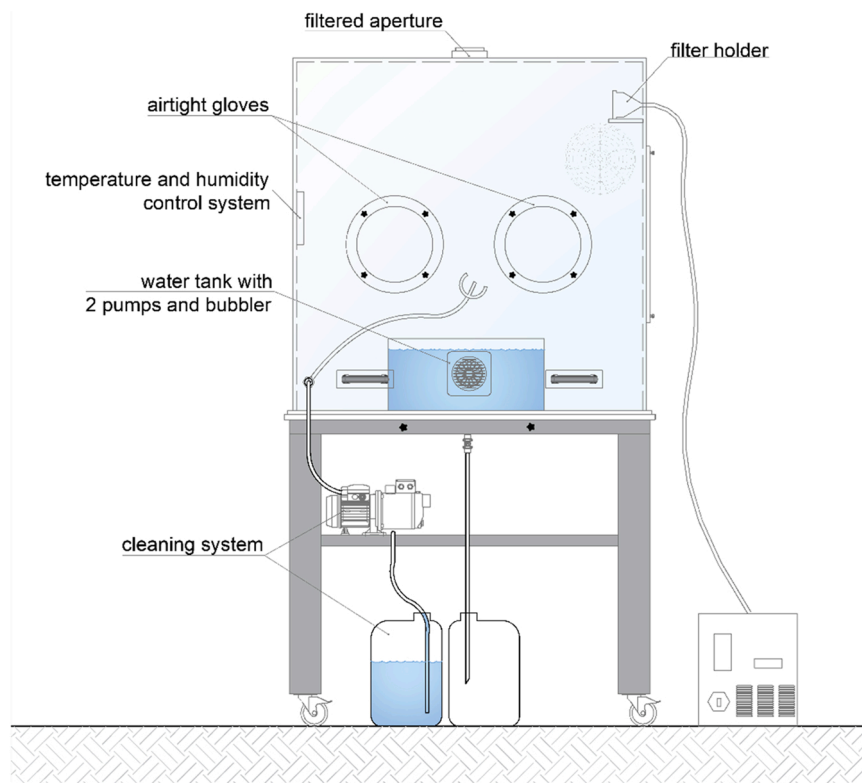
A sampling and analysis campaign was carried out during 2018 and 2019 and mainly chrysotile occurrence was detected in streams (RSA Srl, 2019; see Table S1, Supplementary Information). Average waterborne asbestos concentration was  $10^6$  f/L and peaked up to  $20 \cdot 10^6$  f/L after intense precipitation events. Waterborne asbestos concentration was measured according to the procedure set up by the Regional Agency for the Protection of the Environment (ARPA) (ARPA Piemonte, 2016) and these values are not directly comparable with U.S.-EPA maximum contaminant level (U.S.-EPA, 2004) due to different analytical approaches. However, the data signal a constant and relevant presence of waterborne asbestos throughout all months of the year. Clearly, when the superficial network is considered, stream flow results in bubbling and foaming of water and in water evaporation, particularly in the summer. These two processes could be implied in waterborne fibres passage in air, with possible implications in terms of human exposure to airborne fibres.

Starting from this background, we focussed on the necessity to clarify whether the passage of NOA from water to air can be simulated under controlled laboratory conditions and qualitatively and quantitatively investigated.

Specifically, we develop here a method to quantify the amount of fibres that can be released in air from a standardised chrysotile-polluted water, under simulated conditions. Our simulation uses an improved version of the experimental laboratory test that was envisaged by Jones et al. (2009) and Addison et al. (1988). This work allows to evaluate i) morphological modification that might occur to chrysotile during



**Fig. 1.** Location of the investigated area (modified from Sacchi et al., 2021). 1 = recent alluvial deposits. 2 = alluvial deposits of the Balangero Plain (Middle–Upper Pleistocene). 3 = fluvioglacial deposits (Middle Pleistocene). 4 = fluvial deposits (Lower Pleistocene). 5 = Sesia-Lanzo zone. 6 = Lanzo Massif serpentinite. 7 = lakes. 8 = remediation site. 9 = tailing piles. 10 = waste sludge. 11 = rivers. 12 = piezometric levels of the phreatic aquifer measured on Nov. 10–11, 2008 (m a. s.l.).



**Fig. 2.** Schematic representation of the cube device.

migration from water to air, and ii) the amount of airborne chrysotile mobilised from standardised waters that are polluted by a known chrysotile asbestos concentration. Further, an attempt has been made to define a waterborne chrysotile concentration threshold that could be related to the 1 f/L threshold for airborne asbestos in outdoor ambient, given that beyond this value an alarm situation for asbestos pollution is indicated (INAIL, 2010; WHO, 2000).

## 2. Materials and methods

### 2.1. Experimental system

To study chrysotile passage from water to air an experimental setup was modified after Jones et al. (2009) and Addison et al. (1988), the so called “Cube”: a 1 m<sup>3</sup> close system, to avoid ambient interferences, in which a water tank subjected to bubbling is placed and air circulation is guaranteed (Fig. 2). In this system, four experimental tests were run with different chrysotile content (see Section 2.4) in water to evaluate consequent airborne concentration.

As the Cube walls are transparent and sealed, experimental activities could be visually monitored and operators can safely operate the experiment through airtight gloves.

Inside the structure, one or two filter holders (47 mm diameter) for airborne asbestos measures can be placed and connected to an external air sampler. A filtered aperture on the top of the Cube compensates the depression caused by aspiration during air sampling. Air circulation inside the box is guaranteed by fans. Internal pressure, temperature and humidity are constantly monitored by a calibrated thermo-hygrometer.

A tank containing 38.28 L of water with 2 pumps to move the liquid and a bubbler to help aerosol formation is placed inside the box.

The device is equipped by an aspiration and washing system which allows a complete cleaning of the device before and after each experimental test. Disposal and tested waters are stocked in special containers for asbestos-polluted material and discarded accordingly.

### 2.2. Water movement system

The bubbler is an electronically adjustable air pump SERA air 275R plus, suitable for oxygen enrichment by means of an airstone, with two connectors reaching the water tank. It introduces a 4.5 L/min air flux in water, with a pressure  $\geq 0.014$  mPa. The water movement system is completed by a stream pump which guarantees a wide and gentle water flow thanks to a volume flow rate of 33.3 L/min and a submersible pond pump with a 9.17 L/min flow rate. The two pumps and the bubbler connectors are fixed on the bottom of the tank, generating a stronger movement in the lower part of the tank which soften up towards the water surface, aimed at avoiding the suspended solids deposition.

### 2.3. Experimental design

The test developed as follows.

1. An air “background” is sampled inside the Cube before placing water in the system.
2. Tap water clear or with added chrysotile (blank and experiments, respectively) is put in the tank, 1 g of sodium hypochlorite is added to avoid mould proliferation and the movement (stirring and bubbling) system is activated.
3. The water suspension is homogenised for 1 h and a water sample is collected from the tank and prepared for the analysis following the ARPA Piemonte U.RP.M842 rev.03 operating method (ARPA Piemonte, 2016).
4. A 47 mm diameter polycarbonate (PC) air sampling membrane with 0.8  $\mu\text{m}$  pores is put in the filter holder, connected to an external pump.

5. After 16 h of water movement the air sampler is activated: about 3000 L of air are aspirated through the membrane with a 10 L/min flow rate.
6. The air sample membrane is removed from the holder and prepared for the analysis following the Italian regulation on airborne asbestos analysis (DM, 1994, All. 2B).
7. The system is cleaned and an air sample is collected inside the box.

Four tests with nil, low, mid and high concentration (see Table 1) of waterborne chrysotile are carried out. Three air samples are collected for each water concentration.

An air sampling is done outside the Cube during each experimental test to verify that there is no chrysotile dispersion from the Cube. Temperature and humidity inside and outside the Cube are measured before and after each test. As the Cube was placed outdoor, temperature was dependent on the weather and ranged from 13.5 to 27.8 °C. Internal relative humidity was clearly higher than external, due to the presence of water in the close system. Internal relative humidity was checked to be always over 90% when water movement was activated: this has to be considered as a control limit. In fact, if airborne asbestos occurrence is verified despite the high relative humidity, it is likely to be higher in a natural system, where the environment is gradually drier, moving away from the water surface.

### 2.4. Preparation of the waterborne chrysotile suspensions

A 1:1 mixture of mid to long (“Class 5mx”, after RSA classification) and short fibres (“Filler”, after RSA classification) of processed chrysotile from the Balangero former mine (Fig. 3a, b) was gently crushed in an agate mortar with acetone (hereafter named chrysotile-powder) and used to create the suspensions for the Cube. The gentle manual wet grinding promoted the disaggregation of fibres bundles, and avoided the amorphization of fibres which can occur when more energetic grinding methods are operated in dry conditions (Scognamiglio et al., 2021). The four suspensions are produced by adding chrysotile powder to tap water in order to obtain concentrations reported in Table 1. To completely disperse the fibres in water, the suspensions were stirred for 1 h before use.

SEM images of the chrysotile powder suspension (Fig. 3c, d) showed predominant isolated dispersed fibres coexisting with several aggregates and bundles. The size distribution of dispersed fibres extended towards the shortest fibres, which showed a length ranging mainly from 1 to 12  $\mu\text{m}$ , with the main fraction  $\leq 5$   $\mu\text{m}$ . Aggregates and bundles of fibres had the typical fibrous, curvilinear morphology of chrysotile with frayed and splayed ends. They showed length of up to 200  $\mu\text{m}$  and width ranging from 0.3 to 4.5  $\mu\text{m}$ .

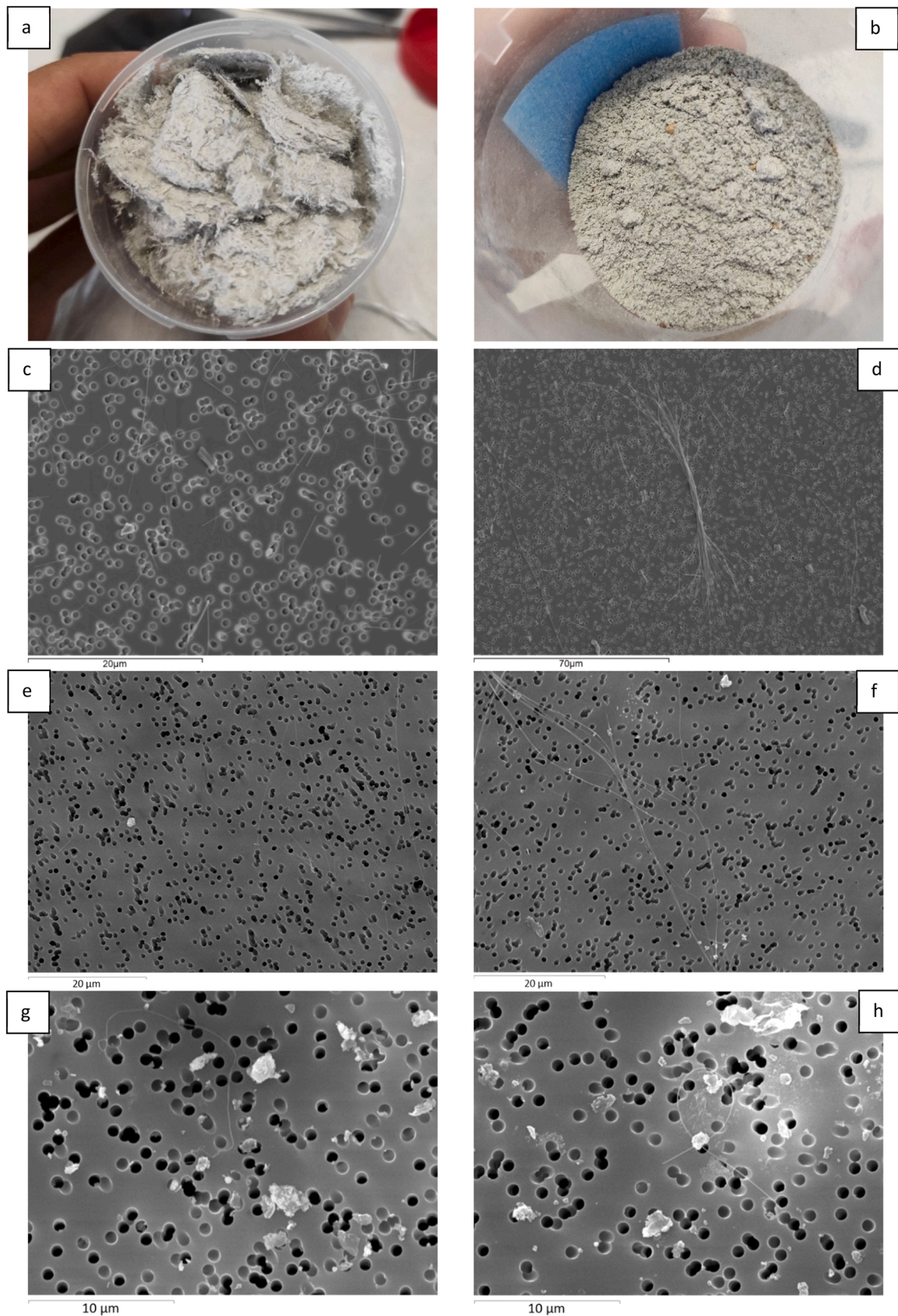
The decision to use a chrysotile sample from Balangero mine was intended to best represent the real situation of streams of the reference geographical area. The selected chrysotile powder quantities added to each water sample were chosen to have a sufficiently wide concentration range in order to make valuable any variation in the consequent airborne concentration.

### 2.5. Samples preparation and analysis

All air and water membranes were analysed by means of a TESCAN VEGA 3 SBH Vega TC ver. 4.2.25.1 SEM, with W filament operating at

**Table 1**  
Waterborne chrysotile concentration for each water sample used in the test.

Sample name	Waterborne chrysotile in prepared suspension [ $\mu\text{g/L}$ ]
W_0	Drinking water, no chrysotile added
W_1	13.74
W_2	27.48
W_3	137.40



**Fig. 3.** Macroscopic image of material used to prepare the chrysotile-powder: 5 mx (a) and “dusty” filler sample (b). Secondary electrons images acquired by SEM on chrysotile-powder suspension, showing dispersed fibres (c) and a fibre bundle (d). Examples of dispersed fibres (e) and a fibre bundle (f) found in W\_3 sample. Secondary electron images of airborne fibres longer than 5 μm (g, h). Black dots are the 0.8 μm pores of the membrane surface.

20 kV, coupled with an INCA microanalysis suite EDS, Oxford Instruments.

Air membranes were prepared and analysed by SEM-EDS following the Italian regulation (DM 06/09/1994, All. 2B) with a few modifications: 47 mm diameter polycarbonate (PC) membranes were used instead of 25 mm and these were scanned at a magnification of 4000× instead of 2000×, producing images with a 15 pixel/μm resolution. Water membranes were prepared filtering a water aliquot on 0.8 μm pores PC membranes. Membranes were analysed by SEM-EDS following the ARPA Piemonte U.RP.M842 rev.03 operating method (ARPA Piemonte, 2016), acquiring images at 4000× magnification (15 pixel/μm resolution). For all samples, membranes were covered by a thin gold layer and an area of about 1 mm<sup>2</sup> was scanned following a boustrophedon path with evenly spaced acquisition positions. Fibres are counted after verifying their chemical composition and thus chrysotile concentration is calculated in f/L, depending on the water or air volume filtered through the porous membrane.

Following the Italian regulations, concentration data are provided with lower fiducial limit (LFL) and upper fiducial limit (UFL) which represent the 95% confidence limit, based on the hypothesis of a Poisson distribution of fibres on the membrane.

The laboratory is accredited for airborne asbestos analysis by SEM-EDS, in compliance with UNI CEI EN ISO/IEC 17025:2018. It participated to inter-laboratories circuits with a positive score for air samples analyses.

For water samples a result in mass per litre [μg/L] as well as number of fibres per litre [f/L] is provided, based on the conversion method reported in the Italian regulation for massive samples (DM 06/09/1994, All. 1B): the length and width of each fibre is measured, and the volume is calculated approximating the fibre to a cylinder. Mass is then calculated by multiplying volume by density (2.6 g/cm<sup>3</sup> for chrysotile).

The analytical method, given the instrumental characteristics and the experimental conditions used, has a limit of detection (LOD) of 0.2 f/L considering airborne asbestos and 1888 f/L for waterborne asbestos.

### 3. Results

#### 3.1. Water samples

Waterborne chrysotile concentration revealed by SEM-EDS analysis of the four water suspensions loaded in the Cube is reported in Table 2. For each suspension the chrysotile concentration was evaluated as f/L and μg/L.

Waterborne fibres were counted according to WHO criteria for respirable fibres (WHO, 1986) and only those with length ≥ 5 μm, width < 3 μm and Aspect Ratio (length to width) > 3 were considered.

Large bundles of fibres or aggregates (see Fig. 3f) were considered even if their width and Aspect Ratio laid outside the WHO dimensional criteria, because they are a potential source of a great quantity of thinner fibres, upon disaggregation. In case of bundles with ends that open up in fibrils, they were counted as a number of fibres equal to those whose both ends were visible.

For the drinking water sample (W<sub>0</sub>) no fibres were found as

**Table 2**

Waterborne chrysotile concentrations in the W<sub>0</sub>, W<sub>1</sub>, W<sub>2</sub>, and W<sub>3</sub> suspensions used in the experiments. The results are expressed as μg/L and f/L (with lower fiducial limit – LFL- and upper fiducial limit – UFL, calculated following the Italian regulation) for each water sample. No fibres were detected for the W<sub>0</sub> sample and chrysotile concentration is thus below the limit of detection (LOD) of the method (LOD = 1888 f/L).

Sample name	Description	Waterborne chrysotile [μg/L]	Waterborne chrysotile [f/L]	LFL	UFL
W <sub>0</sub>	Drinking water	0.00	< LOD	0	6954
W <sub>1</sub>	Low chrysotile concentration	4.10	3.70·10 <sup>6</sup>	3.41·10 <sup>6</sup>	4.02·10 <sup>6</sup>
W <sub>2</sub>	Mid chrysotile concentration	4.57	11.14·10 <sup>6</sup>	10.35·10 <sup>6</sup>	11.98·10 <sup>6</sup>
W <sub>3</sub>	High chrysotile concentration	39.54	44.36·10 <sup>6</sup>	41.34·10 <sup>6</sup>	47.59·10 <sup>6</sup>

expected, while samples W<sub>1</sub>, W<sub>2</sub> and W<sub>3</sub> showed an increasing concentration value in number of fibres, partially reflecting the waterborne chrysotile concentration of the prepared suspensions.

Regarding results in μg/L, concentrations in water based on revealed fibres were always lower than the nominal waterborne chrysotile concentration. The W<sub>1</sub> suspension was prepared to have a 13.74 μg/L chrysotile concentration and the analysis revealed a 4.10 μg/L waterborne chrysotile content (less than 1/3 of the calculated value for the prepared suspension). For W<sub>2</sub> the analysis revealed a value of 4.57 μg/L instead of 27.48 μg/L (about 1/6 of the calculated value) and for W<sub>3</sub> the actual concentration was 39.54 μg/L instead of 137.40 μg/L (less than 1/3).

Waterborne fibres showed the typical chrysotile morphology with winding thin fibres (see Fig. 3e) that occasionally split in thinner fibrils. Regarding width of detected fibres, the three chrysotile water suspensions were mainly constituted by fibres thinner than 0.1 μm (68.4% in W<sub>1</sub>, 63% in W<sub>2</sub> and 91.2% in W<sub>3</sub>). Concerning length, about 40% of fibres in W<sub>1</sub> was shorter than 10 μm and just over 50% in both W<sub>2</sub> and W<sub>3</sub> (Fig. 4).

The average mass of a fibre [μg/f] in W<sub>1</sub>, W<sub>2</sub> and W<sub>3</sub> was obtained dividing the concentration expressed as μg/L by the concentration in f/L for each water sample and is reported in Table 3 ([μg/L] / [f/L] = [μg/f], i.e., the mass in μg of one fibre).

The average masses found for the three kinds of water suspensions, considering fibres ≥ 5 μm, indicate that fibres with masses ranging between 10<sup>-6</sup> and 10<sup>-7</sup> μg were detected in general, but a considerable variability is evident among the three water suspensions.

#### 3.2. Air samples

Table 4 shows results obtained on air membranes sampled inside the Cube.

Only airborne fibres falling under the WHO criteria for respirable fibres were counted (Fig. 3g, h). Any fibre shorter than 5 μm or characterised by a chemical composition essentially different (i.e. asbestiform balangeroite: Compagnoni et al., 1983) from that of chrysotile has been recorded but not included in the concentration calculation.

The presence of chrysotile fibres in air samples is below or very close to the LOD (limit of detection) for background and blank (drinking water) samples and for all the repetition of cycle 1 (using W<sub>1</sub> water) and 2 (using W<sub>2</sub>). For cycle 3, where water with the highest chrysotile concentration was used (W<sub>3</sub>), the consequent fibres concentration in air is relevant. In particular, airborne chrysotile concentration was 1.7 f/L, 4.9 f/L and 2.4 f/L for the three replicate experiments. All the repetitions produced with W<sub>3</sub> resulted in airborne dispersion greater than the alarm limit of 1 f/L (INAIL, 2010) and 2 out of 3 results are over the pollution limit of 2 f/L (DM 06/09/1994, paragraph 6b).

Fibres shorter than 5 μm were detected in air in all cycles, even for the experimental test conducted with low chrysotile concentration water (W<sub>1</sub>).

A final control air sample was collected after cleaning the Cube and no airborne fibres were detected, confirming the reliability of the cleaning system.

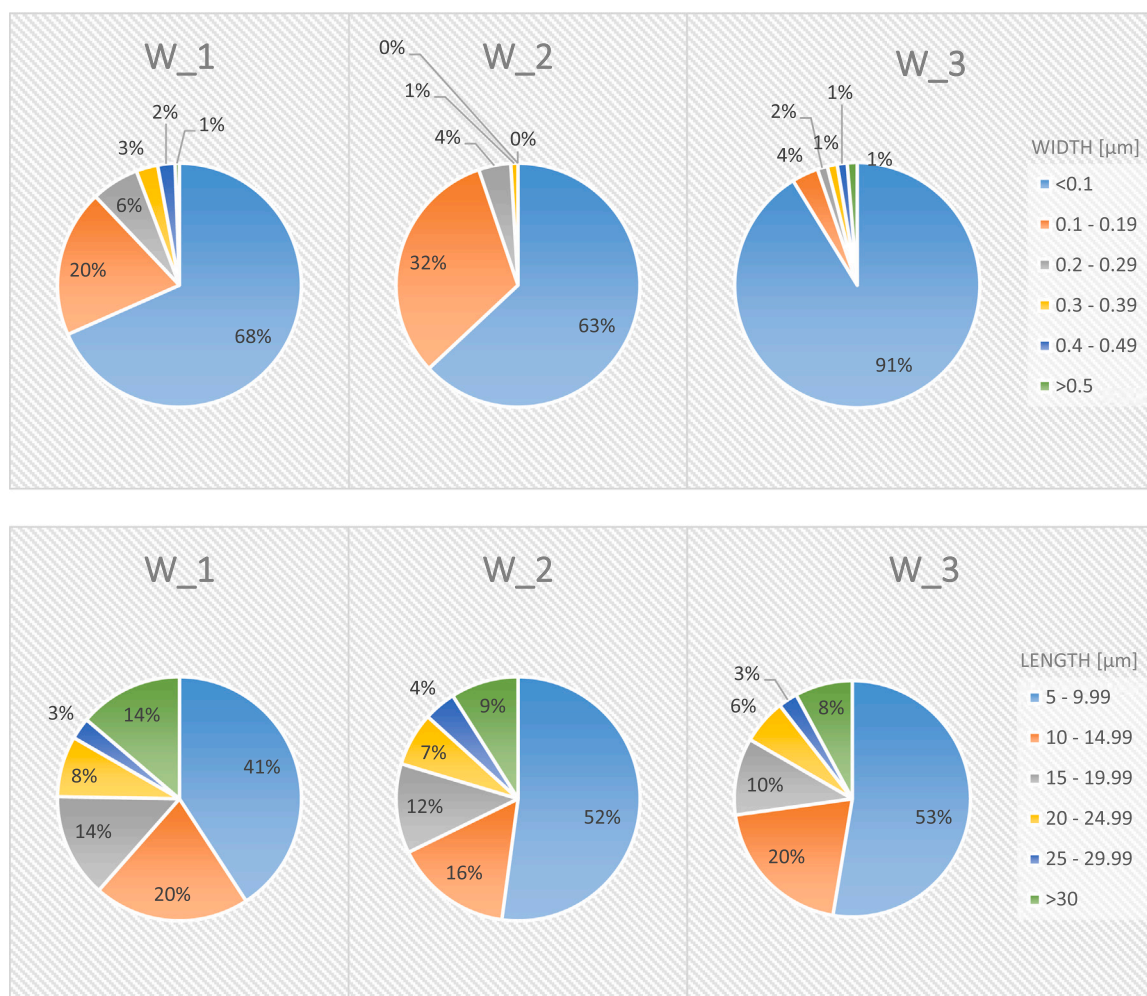


Fig. 4. Width and length distribution of respirable fibres detected in W\_1, W\_2 and W\_3 water samples. Dimension ranges are reported in micrometres.

**Table 3**

Average fibre mass in each water sample, based on chrysotile concentration calculated after analyses.

Sample name	Average fibre mass [ $\mu\text{g}/\text{l}$ ]
W_1	$1.11 \cdot 10^{-6}$
W_2	$4.10 \cdot 10^{-7}$
W_3	$8.91 \cdot 10^{-7}$

#### 4. Discussion

It is of paramount importance to evaluate the migration of pollutants in the environment among different matrices (air, water and soil/rock) because of the creation of novel potential diffusion pathways. Migration may result in contaminants being present in matrices that are not commonly monitored and there possibly posing a new risk to human health and environment. Under these circumstances, asbestos fibres might be considered an Emerging Pollutant (EP) (e.g., Geissen et al., 2015).

Asbestos has been identified for decades among the most dangerous air pollutants and analytical techniques and methods have been set up to efficiently measure the concentration of respirable airborne fibres. Nowadays, the potential hazard related to NOA has drawn attention on environmental natural exposure to airborne mineral fibres but also to other matrices, such as water. Water is here considered as a natural carrier for fibres released from NOA-rich rocks and soils. Fibre accumulation and possible long-range transport in water may result in

asbestos diffusion even very far from the pollution source. Water is also a potential source for fibres migration to air, when motion-generated bubbles collapse and/or evaporation occur.

For the first time, this study considers asbestos as an EP for the water matrix and a method to measure waterborne/airborne fibres concentrations and their possible relationship are proposed.

To verify whether water to air migration occurs, this study was designed to measure the amount of airborne asbestos that can migrate in air, starting from standardised chrysotile-polluted water suspensions. Our data underline the necessity to devote specific research efforts to the water matrix in order to define a safe limit of asbestos content in water.

Although asbestos might be considered an EP in the water matrix, a safe limit has not been proposed for asbestos contamination in water in EU, at present. U.S.-EPA indicates a safe limit of  $7 \cdot 10^6$  f/L for asbestos in drinking water (U.S.-EPA, 2004), which is relevant for possible indoor water/air migration, but it is not completely applicable to non-drinking water. Further, U.S.-EPA regulation does not set any correspondence between waterborne safe limit and airborne concentration of fibres that can be achieved upon water motion/evaporation of asbestos-contaminated water, especially outdoor. To produce a science-based safe limit for waterborne fibres it is of paramount importance that waterborne concentration is put in relation with the pivotal limit of 1 f/L of airborne asbestos.

Our data showed that only in the worst-case scenario, i.e., the migration test carried out with W\_3 water suspension, which has the highest chrysotile content, significant water-to-air migration of fibres occurred. In this case, all the three airborne asbestos measurements

**Table 4**

Chrysotile asbestos occurrence in air samples in f/L, with lower fiducial limit – LFL- and upper fiducial limit – UFL (calculated according to the Italian regulation). For W\_1, W\_2 and W\_3 suspensions, 3 repetitions of air samples were collected. L = length.

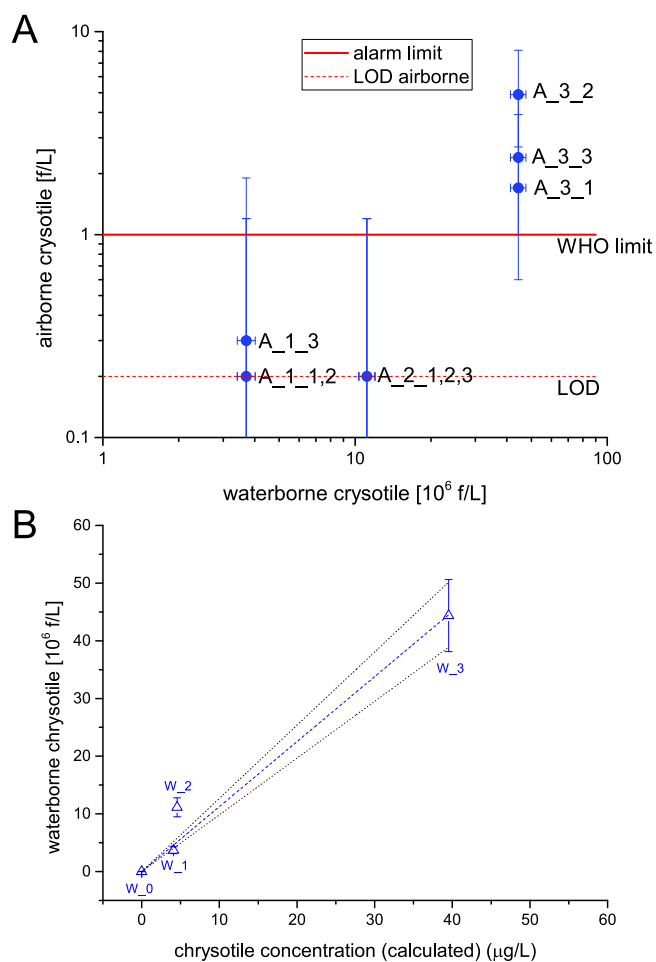
Sample name	Description	Airborne asbestos [f/L]	LFL [f/L]	UFL [f/L]	Notes
Background	No water	< LOD	0.0	1.3	
Blank	W_0 water	< LOD	0.0	1.3	
A_1_1	W_1 water, repetition 1	< LOD	0.0	1.2	Short chrysotile fibres (L < 5 µm) detected
A_1_2	W_1 water, repetition 2	< LOD	0.0	1.2	
A_1_3	W_1 water, repetition 3	0.3	0.0	1.9	
A_2_1	W_2 water, repetition 1	< LOD	0.0	1.2	Short chrysotile fibres (L < 5 µm) detected
A_2_2	W_2 water, repetition 2	< LOD	0.0	1.2	
A_2_3	W_2 water, repetition 3	< LOD	0.0	1.3	
A_3_1	W_3 water, repetition 1	1.7	0.6	3.9	Short chrysotile fibres (L < 5 µm) detected
A_3_2	W_3 water, repetition 2	4.9	2.7	8.1	
A_3_3	W_3 water, repetition 3	2.4	1.0	5.0	
Control	No water, sampled at the end of all the runs	< LOD	0.0	1.2	

signalled concentration of fibres higher than the alarm limit of 1 f/L. W\_3 revealed waterborne chrysotile asbestos concentration that exceeds  $40 \cdot 10^6$  f/L and that corresponds to an airborne concentration of 1.7 f/L or more. Instead, a waterborne fibres contamination level as high as  $11 \cdot 10^6$  f/L (W\_2) produced no airborne respirable fibres to be detected. Therefore, the waterborne fibres concentration limit which can release at least 1 f/L in air has to be set in the range  $11\text{--}40 \cdot 10^6$  f/L (Fig. 5a).

More data have to be collected in this concentration range to define the relation that quantitatively describes the fibres migration from water to air, in this experimental setup, and those measures are being currently designed.

Because only results expressed as number of fibres per litre are considered relevant to human exposure, with regard to possible health implications (WHO, 2000), and factors to convert asbestos concentration from number to mass per volume were elusive, mass concentration had been historically considered of secondary importance in the definition of a safe limit. Despite this, we consider that an attempt to define a correlation between asbestos concentration in number and mass per volume would enhance analytical procedures introducing improvements in health protection strategies. Based on our data, the threshold limit of waterborne chrysotile asbestos in mass which could release airborne fibres to a concentration of 1 f/L could be between  $4.57 \mu\text{g/L}$  and  $39.54 \mu\text{g/L}$ , i.e., the W\_2 and W\_3 chrysotile asbestos concentrations, respectively.

As asbestos is an inorganic solid pollutant, the migration from water to air is not as relevant as for volatile compounds migration.



**Fig. 5.** Chrysotile airborne concentration in f/L in relation to waterborne concentration in  $10^6$  f/L generating that amount of fibres in air (a). Lower and upper fiducial limits (LFL and UFL) are reported as error bars. The red solid and the dashed lines represent the “alarm limit” set for asbestos in air and the limit of detection (LOD) for airborne fibres, given the conditions used for this study. Graphical representation of the relation between waterborne concentrations of chrysotile reported as  $\mu\text{g/L}$  and  $10^6$  f/L (b). Linear fit and upper and lower confidence limits (95% confidence band) are reported. ANOVA was performed on the linear regression data and a significance p-value  $< 0.05$  was obtained. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Nonetheless, migration might become relevant when water nebulisation and/or evaporation is triggered by bursting bubbles from whitecaps in foaming streams and rivers as seen for oceans (e.g., Huntlin et al., 2010). Under these conditions a large number of water droplets is generated, and the extent of the water/air interface dramatically increases. Small inorganic particles (micrometric to nanometric), such as NOA fibres, may thus migrate in air during the rapid evaporation of the droplets.

To estimate the transfer rate of fibres from contaminated water to air in our experimental setup, the percentage of fibres mobilised was calculated as the ratio between the number of airborne fibres per litre of air and the number of waterborne fibres per litre of water. Analysing the data from W\_3 experiment, the mobilisation ratio ranged from  $4 \cdot 10^{-6}\%$  to  $1.1 \cdot 10^{-5}\%$ .

These values significantly differ from data presented in the studies by Hardy et al. (1992) and Roccaro and Vagliasindi (2018). In the former, the percentage of fibres transferred from contaminated water to air that ranged between 0.03% and 4.7% when mobilisation was promoted by an ultrasonic humidifier. In the latter, the percentage of mobilisation ranges from 0.04% to 0.07% and from 4.3% to 10.8%, when



contaminated water was used in humidifiers and showers, respectively. The greater migration of fibres to air that was observed in these studies might be due to a more effective system to induce fibres migration and to the smaller size (both length and width) of the waterborne fibres used, particularly in the study by Hardy et al. (1992). Indeed, submicrometric particles are expected to be easily aerosolised by humidifiers, depending on the size of droplets and particles (Webber et al., 1988). In addition, the airborne and waterborne concentration calculated by Hardy et al. (1992) was based on all fibres, even those that did not fall in the WHO criteria for respirable fibres. Lastly, it is noteworthy that in the experimental setup by Roccaro and Vagliasindi (2018) the water consumption changed in accordance with the experiment and the remarkable passage of fibres in air in some tests could be linked to low relative humidity.

In the light of the above considerations, the migration percentages obtained in our study are consistent with a low migration rate that can occur in natural systems. In fact, it is considered that the production of aerosol from stream waters may be compared to that of oceans (Huntlin et al., 2010) and, therefore, is less efficient than a shower or a humidifier (Sain et al., 2018). Our study was indeed designed to give indications about the potential migration of chrysotile from water to air in conditions that simulate, for the first time, water motion in streams. Despite the limited migration ratio, the absolute number of airborne fibres that were detected in the Cube environment during the experiment that simulates the worst-case scenario (W\_3), is greater than 1 f/L. Obviously, in a real-life scenario, wind likely modifies local fibre concentration and the transportation of the fibres in air makes the real system virtually impossible to be investigated quantitatively.

Our study was also aimed to evaluate the possible transformation of the morphology and the aggregation state of the chrysotile-powder used to prepare asbestos polluted water suspensions that were subjected to bubbling and foaming. In fact, moving water action may favour disaggregation/defibrillation of large bundles in a high number of thinner and shorter fibres that might easily migrate in air. The chrysotile defibrillation prompted by water motion resulted in a difference in airborne fibres morphology compared to waterborne fibres (see Fig. 4), as already reported in Hardy et al. (1992).

From the health and safety point of view, this defibrillation process poses further concern on the possible role of asbestos larger bundles, that are usually disregarded by the numeric evaluation as fibre per litre, but that would be included in the overall level of contamination if mass per volume unit are used. In our study, the waterborne chrysotile concentration, reported as f/L for W\_1, W\_2 and W\_3 suspensions, generally reflects the relative content dispersed in water, as shown in Table 2. On the contrary, the calculated waterborne concentration, expressed as  $\mu\text{g/L}$ , is always lower than the nominal concentration of the water suspension prepared to feed the Cube (see Tables 1 and 2).

The discrepancies among the two data (nominal and calculated) are likely due to the analytical protocols conventionally used to measure asbestos. The protocol indeed calculates the concentration summing the masses of all fibres  $\geq 5 \mu\text{m}$ . However, preliminary SEM-EDS analyses indicate that the standard chrysotile dispersed in water is mainly made of shorter fibres ( $< 5 \mu\text{m}$ ) that are not counted under the adopted analytical method. In addition, during water motion, longer and thicker bundles may undergo defibrillation achieving undetectable length ( $< 5 \mu\text{m}$ ). These phenomena may generate a bias that explains the difference between the nominal and calculated mass concentration of chrysotile in water.

Interestingly, the calculated chrysotile mass in water suspensions still reflects the nominal ratios among the original masses dispersed in water. Only the W\_2 concentration of  $4.57 \mu\text{g/L}$  deviates from the trend showed by other water samples and has to be considered as an outlier. The relation between the number of fibres and the mass is strongly dependent on fibres dimension. In W\_2 the great amount of chrysotile in f/L does not generate an equally large amount in mass because of the smaller dimensions of fibres detected in this sample (average mass  $4.10 \cdot 10^{-7} \mu\text{g/f}$ , as reported in Table 3).

Despite the limited number of experimental simulations carried out ( $n = 4$ ), our data may indicate a possible linear relationship ( $R^2 = 0.99$ ) between the waterborne fibre concentration that is calculated as the mass of fibres per volume unit [ $\mu\text{g/L}$ ] and the number of fibres detected per litre of water [f/L], as reported in Fig. 5b. A larger number of experimental simulations has been already designed to support this hypothesis.

Different bubbling time and power could influence fibres dimension distribution because they can affect the degree of fibres disaggregation and separation from original bundles. Consequently, the more the fibre bundles disaggregate, the higher value in fibres per litre is found, the mass remaining unchanged. Further studies should be designed to assess how bundles mechanical defibrillation in mobilised water occurs and how it might affect the possible mobilisation of fibres in air, depending on bubbling time and fan power.

The abundant presence of fibres thinner than  $0.2 \mu\text{m}$  in water constituted an analytical problem because it was difficult to have a clear chemical characterisation due to the electron beam resolution, using SEM-EDS. Furthermore, in real samples from streams and rivers, organic fibres which are flexible and winding and morphologically very similar to chrysotile fibres are often detected: asbestos can only be discriminated from those fibres by analysing chemical composition with EDS. For this reason, the US regulation requires the use of Transmission Electron Microscopy (TEM) to analyse water samples (U.S.-EPA, 1994, 1983), which allows the detection of width down to few tens of nanometres (Millette et al., 1980; Schreier, 1987). On the other hand, the Italian regulation requires the use of SEM because it guarantees easier samples preparation and a greater sample representativeness (Buzio et al., 2000).

In addition, the evaluation of waterborne fibre number and mass was non-trivial, because of the presence of bundles of fibres. Thick bundles (width  $> 3 \mu\text{m}$ ) indeed do not represent a risk for human health if they become airborne in this form because of their non-respirable dimensions. However, they could be disaggregated by water action and become a relevant source of health risk. For this reason, we chose to include bundles in the quantification of fibres, even if they are normally not considered as the lay outside the WHO dimensional criteria.

Several difficulties had to be overcome to achieve sufficiently sound data about waterborne chrysotile concentrations. The dependence of concentration, particularly when data are expressed as f/L, from the microscope resolving power and, more dramatically, from operator's skills in discriminating asbestos from non-asbestos fibres suggests that a standardisation of analysis procedures is needed to improve the reliability of waterborne measurements (Turci et al., 2016), in particular if applied to non-drinking water.

On the other hand, the quantitative data regarding airborne fibres were easily collected for the availability of standardised analytical methods, shared since the '90s (in Italy, DM 06/09/1994, All. 2B). Analytical procedures for airborne fibres are well established and ambiguities are very rare. Airborne chrysotile samples analysed for this study generally consisted of fibres slightly thicker than the waterborne ones, therefore easier to be chemically characterised and more precisely measured.

At present, this experimental approach has been tested on chrysotile only, which could be considered the worst case scenario given the above-mentioned difficulties linked to fibres morphology. It is therefore conceivable that it could be used to assess other fibrous minerals (e.g., amphibole asbestos) easier to be detected due to their stiffer shapes and generally greater diameters (Schreier, 1989).

## 5. Conclusions

This study simulated the generation of airborne fibres from chrysotile asbestos contaminated water to investigate the migration that can occur under collapse of bubbles and foams from polluted waters in nature. An experimental device of  $1 \text{ m}^3$  was designed to simulate the

water/air migration and tested under several conditions.

Our data indicate that at a concentration of ca.  $40 \cdot 10^6$  f/L waterborne fibres in a polluted water might transfer in air a sufficient amount of fibre to produce an airborne concentration greater than 1 f/L, a safety threshold for several health and safety organizations. Further, as no airborne fibres were detected at lower waterborne concentrations, our study suggests that a safe limit concentration for waters might exist and that this value should reflect airborne migration factor.

Our study evidenced the necessity of standardised methods for non-drinking water analysis to provide reliable analytical procedure for waterborne asbestos quantification with SEM-EDS.

### CRedit authorship contribution statement

**Chiara Avataneo:** Conceptualization, Methodology, Investigation, Visualization, Writing – original draft, Writing – review and editing. **Jasmine R. Petriglieri:** Investigation. **Silvana Capella:** Investigation, Formal analysis. **Maura Tomatis:** Investigation, Formal analysis. **Mariagrazia Luiso:** Investigation. **Giuliana Marangoni:** Investigation. **Elisa Lazzari:** Visualization, Resources, Supervision. **Silvio Tinazzi:** Conceptualization, Methodology, Writing – review and editing, Supervision. **Manuela Lasagna:** Visualization. **Domenico A. De Luca:** Supervision. **Massimo Bergamini:** Conceptualization, Methodology, Resources, Writing – review and editing, Supervision. **Elena Belluso:** Conceptualization, Methodology, Resources, Writing – review and editing, Supervision, Funding acquisition. **Francesco Turci:** Conceptualization, Methodology, Resources, Writing – review and editing, Supervision, Funding acquisition.

### Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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### Appendix A. Supplementary information

Supplementary data associated with this article can be found in the online version at [doi:10.1016/j.jhazmat.2021.127528](https://doi.org/10.1016/j.jhazmat.2021.127528).

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