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Abstract

The natural occurrence of asbestos (NOA) in rural areas is a serious concern for human health and the dispersion route of asbestos in the proximity of natural asbestos-rich settings has been marginally evaluated so far. NOA may affect air, but also water and soil quality. In rural areas population may be exposed to asbestos with a largely unknown impact on human health. This work investigates the potential exposure of a farmer cultivating a field nearby the largest former asbestos mine of Western Europe (Balangero, Italy). The concentration of waterborne asbestos in the stream used to water the field was measured (ca. $2 \cdot 10^5$ fibers per liter, ff/L) and the cultivated ultramafic topsoil characterized, evidencing a remarkable occurrence of chrysotile. The worker's personal exposure and the environmental fiber dispersion during a simulated agricultural activity (tillage) were quantified in two independent trials. During the trials, the worker was exposed to average concentrations of 16 and 26 ff/L, with a peak of 40 ff/L. These data inform about the possible exposure of an agricultural worker to asbestos concentration higher than the accepted threshold of 2 ff/L. The release of asbestos fibers into the environment was negligible (0-2 ff/L).

Keywords: natural occurrence of asbestos (NOA); waterborne asbestos; chrysotile; ultramafic soil; personal exposure

1. Introduction

Asbestos is one of the most harmful occupational carcinogens causing more than 100’000 deaths per year and exposure to airborne asbestos fibers is held responsible for half of the deaths from occupational cancer [1]. Cases of mesothelioma, the fatal malignancy most commonly caused by exposure to asbestos, have also been described in individuals exposed to the natural occurrence of asbestos (NOA) or asbestos-like minerals in Turkey, Greece, Cyprus, Corsica, Sicily, New Caledonia, Yunnan province (China) and California (USA) [2, 3]. International organizations of health and safety agree on the absence of a “safe” level of asbestos exposure for any type of asbestos fiber, including chrysotile [1, 2]. An environmental background level of 2 fibers per liter (ff/L) is widely accepted and is enforced in many national and international regulations (e.g., Italian law on the remediation of indoor asbestos-contaminated environments, DM 6/9/94).

The asbestos route from natural and anthropic sources to human lungs has to be increasingly understood to maximally reduce both occupational and environmental exposures [4]. Environmental asbestos contamination has been primary studied in the air [5], but both water and soil have increasingly deserved the attention of public health agencies and researchers [6-9]. High levels of asbestos fibers have been widely documented in the raw waters of rivers and wells in the surroundings of active and inactive asbestos mines [10-12]. The presence of asbestos fibers in drinking waters has mainly attracted the research interest because of potential direct effects on human health [13-16].The hydrographic network may also contribute to the transport of fibers in the environment, where asbestos may eventually disperse in the air [17]. In particular, exceptional floods of asbestos-polluted waters were shown to deposit centimeter-thick layers of asbestos-rich sediments on soils [18]. Furthermore, contaminated waters from asbestos-rich natural areas are used for irrigation and livestock watering [19].

In this scenario, soil plays a role as both sink and source along the route of asbestos dispersion [20]. Dry, undisturbed, asbestos-rich soils may generate respirable airborne asbestos fibers [21], but low levels of moisture (5-10%) reduce or completely suppress the fiber dispersion in the air [22]. However, specific studies that assess the risk associated to the exploitation of asbestos-contaminated soils in agriculture or construction are scarce. Only one report on the agricultural practices on an asbestos-polluted industrial ground is available. Authors show a significant increase of airborne asbestos concentration (up to 16 ff/L, fiber length > 5 μm) in the site after soil cultivation [23]. At the best of our knowledge, the exposure of farmers managing waters and soils in the agricultural surroundings of dismissed asbestos mines is still completely unexplored.

To assess if a farmer may be exposed to asbestos fibers, this study: i) characterizes the occurrence of asbestos in surface waters used for irrigation and in an agricultural topsoil in the proximity of the largest former asbestos mine in Western Europe (Balangero, Italy); and ii) investigates the exposure to airborne fibers during a simulated agricultural activity. A corn-farmed field irrigated and periodically flooded by waters draining the former asbestos mine was selected. Waterborne fiber concentration was measured and fibrous minerals in the topsoil characterized. The environmental fiber dispersion and the human exposure during tillage were simulated and quantified in two independent trials with different meteorological conditions. For the first time, this work assesses the potential risk of a common agricultural activity in an asbestos-polluted environment, addressing the role of both soil and water as source of natural fibers.

2. Experimental

2.1 Sampling sites and simulated agricultural activities

The study area (Fig. 1) is the Balangero plain (440 m above sea level, average rainfall 1160 mm/yr., average annual temperature 10 °C) downstream the southern boundary of the former chrysotile mine, which is located in the Ultramafic Lanzo Massif. A main water stream (Rio Banna) collects superficial waters that drain the southern tailings of the Balangero former asbestos mine. Surface waters were sampled along the Banna stream after the confluence with the brooks draining the southern tailings of the mine (water sampling site "W1" in Fig. 1) and ca. 2 km southeast nearby a cultivated corn field (soil sampling site "CF"; 3500 m^2) irrigated or periodically flooded by the same untreated water ("W2"). A stream flowing through the serpentine area outside the mine asbestos-rich tailings ("W3") and two wells (deep water sampling sites "Wd1" close to the mine southern tailings, and "Wd2" upstream the mine basin, as negative control) were also sampled and compared.

In the cultivated corn field ("CF"), agricultural activity was simulated during two independent trials ("Day1", "Day2") under different meteorological conditions. Temperature, rainfall events, humidity and wind velocity were monitored during the weeks before the trials (Fig. 2). Day1 and Day2, following two and seven days without rain, respectively, mostly differed in relative humidity (67% and 52%, respectively). During each trial, a "farmer" carried out tillage operations in four independent sectors (12×70 m each) of the field with a small agricultural tractor operated for 45 minutes per sector.

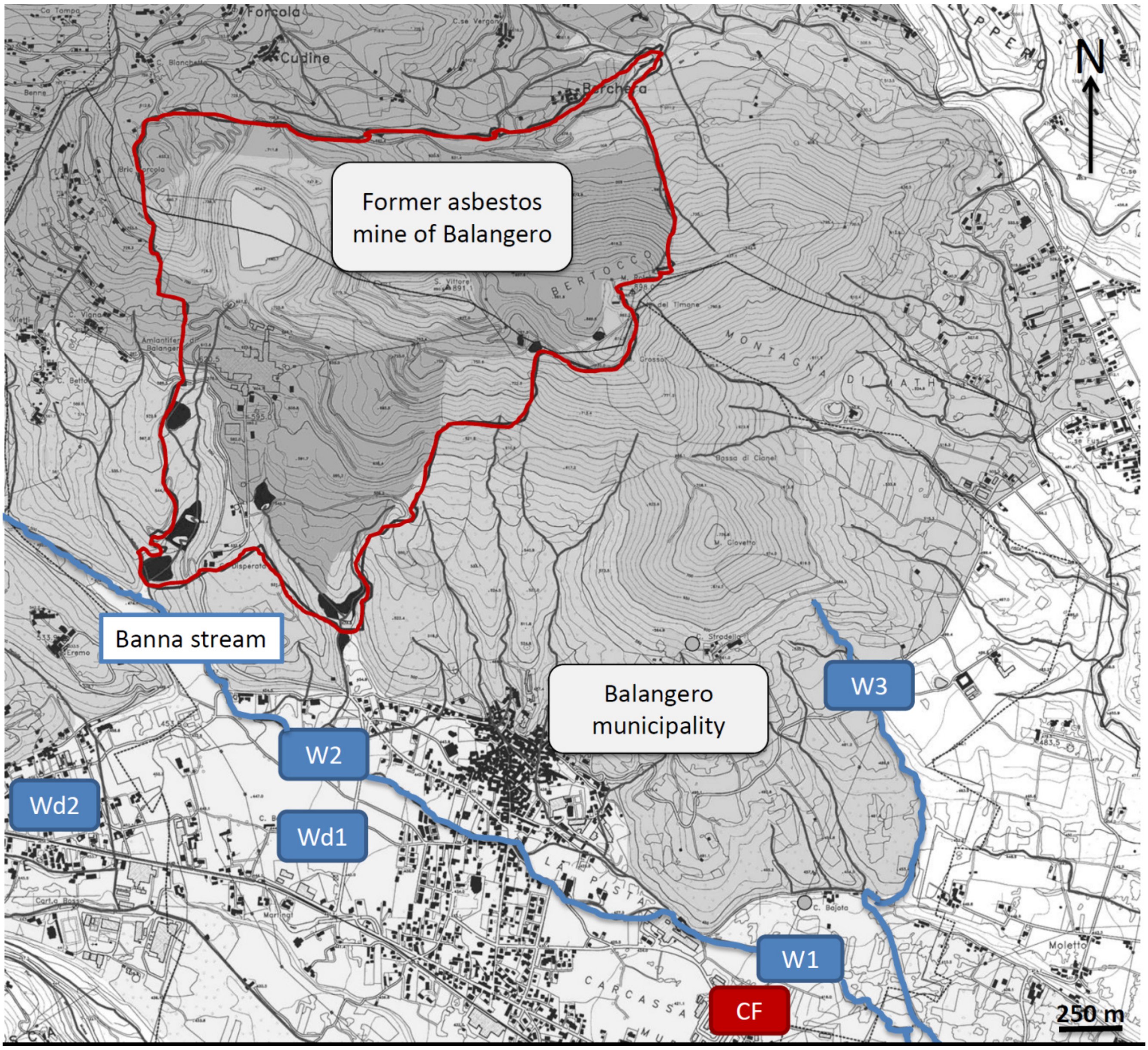

Fig. 1. Study area of Balangero plain. The boundaries of the former asbestos mine are highlighted in red. Superficial water sampling sites: W1, W2, and W3 (negative control); deep water sampling sites: Wd1, and Wd2 (negative control); cultivated corn field (CF), where simulated agricultural operations and soil and air sampling were carried out.

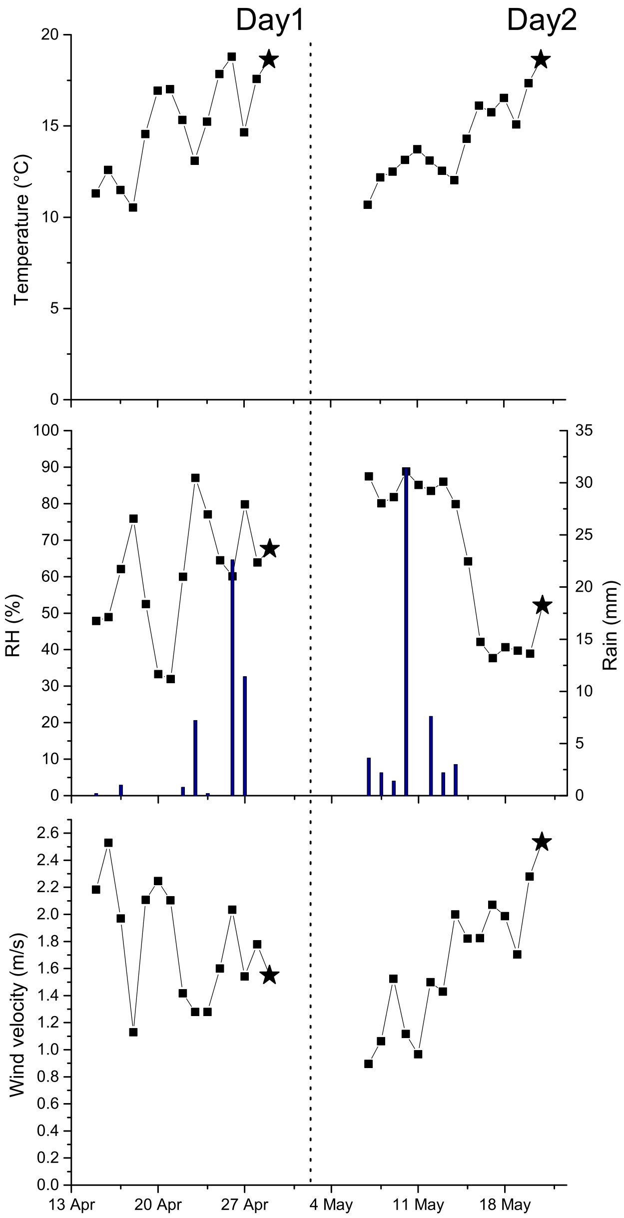

Fig. 2. Meteorological conditions during the weeks before the trials (Day1 and Day2, stars): temperature, rainfall events, humidity, and wind velocity were monitored by the meteorological station of Balangero.

2.2 Waterborne fiber sampling and analysis

One liter of water was collected for each sampling site and stored in pre-rinsed bottles at 4 °C to minimize biological contamination. Samples were processed and analyzed according to the technical procedure U.RP.M842-Rev2 prepared by the Italian Environmental Protection Agency (ARPA) for waterborne fiber quantification. Aliquots of the collected waters (200 or 100 mL according to the sample turbidity) were filtered on polycarbonate porous membrane (pore size 0.8

μm) and dried at 100 °C for 3 hr. A conductive gold coating was sputtered on the membranes to allow for morphological and chemical analysis by scanning electron microscopy coupled with energy dispersive spectrometer (SEM-EDS: Carl Zeiss EVO50-XPV equipped with an INCA Oxford apparatus). A representative portion of 1 mm^2 was observed at 4000 × and each particulate with fibrous or acicular habit (length/width ratio > 3) was dimensionally and chemically characterized. The detected chemical elements, the relative peak intensities, and the morphology of the particles were used for the fiber identification (see [24]).

2.3 Top-soil sampling and analysis

During the tillage operations, the tractor was equipped with a home-made device (Fig. 3) to sample the topsoil. Specifically, an industrial vacuum cleaner (HZQ570 Single Motor Vacuum, Numatic International, Chard, Somerset, UK) equipped with a ULPA output filter, a HEPA-FLO vacuum bag, and a collector nose placed at ca. 40 cm from the ground was used. This method allowed for a homogeneous collection of composite, average soil samples from each tilled sector of the field. One sample per trial per sector was collected and homogenized by powdering in an agate mortar. The chemical composition was determined with an EDAX Eagle III energy dispersive micro-XRF (μXRF) spectrometer equipped with a Rh X-ray tube, an EDS Si(Li) detector and an Edax Vision 32 microanalytical system software. The mineralogical composition was determined with X-Ray powder diffraction analyses performed using a PANalytical PW3040/60 X' Pert Pro MPD with Debye-Scherrer geometry and Cu Kα radiation. NaCl was added at 5% weight concentration as internal standard. Soil samples were coated with gold and observed under scanning electron microscopy (SEM: Carl Zeiss EVO50-XPV) to qualitatively assess the occurrence of fibrous minerals.

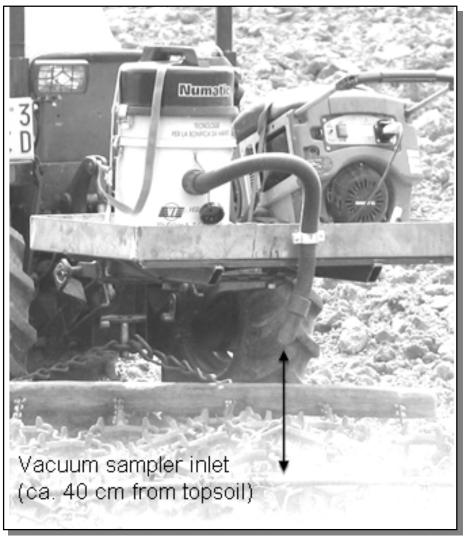

Fig. 3. Tractor performing the simulated tillage was equipped with an industrial vacuum cleaner to sample the stirred and overturned topsoil.

2.4 Airborne fiber sampling and analysis

During the tillage operations, airborne asbestos fibers were sampled to evaluate both the exposure of the "farmer" and the dispersion of fibers in the environment.

The "farmer" operating the tractor was equipped with a personal air sampler fasten to the worker's lapel (NIOSH Method 7400) at approx. 150 cm from the ground, mounting a cellulose acetate filter (diameter = 25 mm, pore size = 0.8 μm) and operated at 3 L min^{-1} during the 45 minute tillage operations. Four samples per trial were thus collected, i.e. one sample for each sector for each trial. During the work the "farmer" wore personal protective equipment for asbestos exposure which did not interfere with fiber sample.

Airborne fibers in the environment were collected during each trial using a conventional environmental sampling apparatus placed leeward at the border of the field, equipped with a cellulose acetate filter (diameter = 36 mm, pore size = 0.8 μm) and operated at 10 L min^{-1} for 3 hours. The sampling nose was placed at approx. 150 cm from the ground.

The filters were divided in four parts, one of which was coated with gold and observed with SEM (Carl Zeiss EVO50-XVP) operated at 2000× magnification and equipped with an Oxford EDS apparatus. The presence of inhalable (length/width ratio > 3; L > 5 μm; w < 3 μm; WHO 1986) and non-inhalable (L < 5; w > 3) mineral fibers on each filter was quantified in an area of 1 mm^2.

The following equation was used to quantify asbestos concentration:

(Eq. 1) $$\text{Asbestos concentration} = \frac{n}{a} \times \frac{A}{V},$$

where *n*, is the number of fibers counted on each filter, *a* is the area analyzed, *A* is the total area of the filter, and *V* is the volume of sampled air.

The chemical analysis of each fiber was performed using a Microanalysis Suite Issue 12 (INCA Suite, Oxford Instruments) system. The fibers were identified on the basis of their chemical composition, the relative peak intensities, and the morphology (see [24]).

3. Results & Discussion

3.1 Fiber content in water

The superficial waters of the river (Table 1) draining the southern slope of the former asbestos mine in Balangero (W1, W2) contained approx. $2 \cdot 10^5$ fibers per liter (ff/L). The vast majority of the fibers were identified as asbestos. Specifically only chrysotile among the six minerals defined as asbestos was detected. The fiber content for the two sampling sites was virtually the same, indicating that the distance from the mine tailings does not affect the waterborne fiber concentration. A direct comparison with literature data on similar environments is prevented by the heterogeneity of adopted analytical protocols and instrumentations or by the metric choice (e.g. fiber number or mass per volume). For instance, the fiber concentration from this work is lower than those detected by TEM analysis for asbestos-rich basins in Europe (e.g. from 4 to $156 \cdot 10^6$ ff/L in [11]) and North America (e.g. $5 \cdot 10^7$- $3 \cdot 10^{11}$ in [25-27]), but higher than those detected by light microscopy in Russia (e.g. $1 \cdot 10^4$ ff/L in a river near Asbest city [13]). The fact that the more potent is the microscopy used, the higher is the level of asbestos, suggests the need for standardized sampling and analysis procedures to consistently compare measurements of waterborne asbestos . Notwithstanding the lack of an absolute reference for waterborne fiber concentration, our analysis shows that fibers in the stream draining the serpentine area outside the mine (W3) were approx. a half of W1 and W2, suggesting that the waters draining the mine tailings may be enriched in asbestos with respect to the natural background of the Ultramafic Lanzo Massif. Similarly, the deep-water sample (Wd1) from a well close to the southern mine tailings contained > $1 \cdot 10^6$ff/L, while the asbestos content related to the environmental background (Wd2) was ca. $1 \cdot 10^4$ ff/L. These preliminary data suggest that the streams collecting superficial waters draining the asbestos mine tailings, enriched in waterborne asbestos, may become a source of soil pollution.

Table 1. Waterborne asbestos concentration in superficial and deep water sampling sites. Lower and Upper Fiducial Limit (LFL and UFL) of fiber concentration calculated assuming a Poisson distribution for fiber count on each filter. Total fibers (asbestos + non asbestos mineral fibers) are also reported.

Sample	Location	GPS position (UTM coordinates)	Asbestos fibers (ff/L)	LFL (ff/L)	UFL (ff/L)	Total fibers (ff/L)
W1	Mine stream -, upper tract	X:383244 Y:5014545 Z:32N	215'373	129'668	336'331	294'720
W2	Mine stream -, corn field	X:384863 Y:5013811 Z:32N	192'702	133'452	269'281	192'702
W3	External stream (control)	X:385427 Y:5014871 Z:32N	102'018	46'469	193'663	113'354
Wd1	Well	X:383271 Y:5014090 Z:32N	1'076'863	929'180	1'241'349	1'102'368
Wd2	External well (control)	X:381915 Y:5014499 Z:32N	11'335	1'373	40'947	17'003

3.2 Chemical and mineralogical composition of the topsoil

The chemical (μXRF) and mineralogical (XRPD) composition of the topsoil collected in the crop filed reflects the proximity of the site to the ultramafic outcrops of Lanzo Massif. High Mg/Ca ratio (> 2) and presence of heavy transition metals (Fe >> Cr, and Ni; Table 2) are typical of ultramafic soils [28]. Serpentine [$Mg_3Si_2O_5(OH)_4$] is indeed detected (Fig. 4) as a major constituent of the soil together with iron oxides (Fe_xO_y), commonly found in ultramafic soils. Abundant quartz (SiO_2), which is among dominant components of every soil, and albite ($NaAlSi_3O_8$), possibly deriving from abundant gabbric and/or gneiss outcrops in the surroundings [29], are also detected. Clay-minerals also occurred (low 2 theta XRPD peaks), which on the basis of their chemical composition can be mainly identified as smectites, which are the main pedogenetic derivatives of serpentine [18, 28]. As expected, SEM observations highlighted that serpentine mainly occurred in the lamellar habit (antigorite), with a minor presence of fibrous individuals (chrysotile; Fig. 5A and B), often organized in fibrous bundles up to 20 μm in diameter. Some fibrous occurrence of smectite (Fig. 5C), the main weathering product of chrysotile [18], were also detected.

Table 2. Micro-XRF analysis of the topsoil averaged from the eight samples (4 sectors per 2 sampling days). Results are reported as atomic %.

	Mg	Al	Si	K	Ca	Ti	Cr	Mn	Fe	Ni
Topsoil of CF (at. %)	10.8	14.2	48.7	3.3	5.8	1.2	0.3	0.3	14.1	0.2

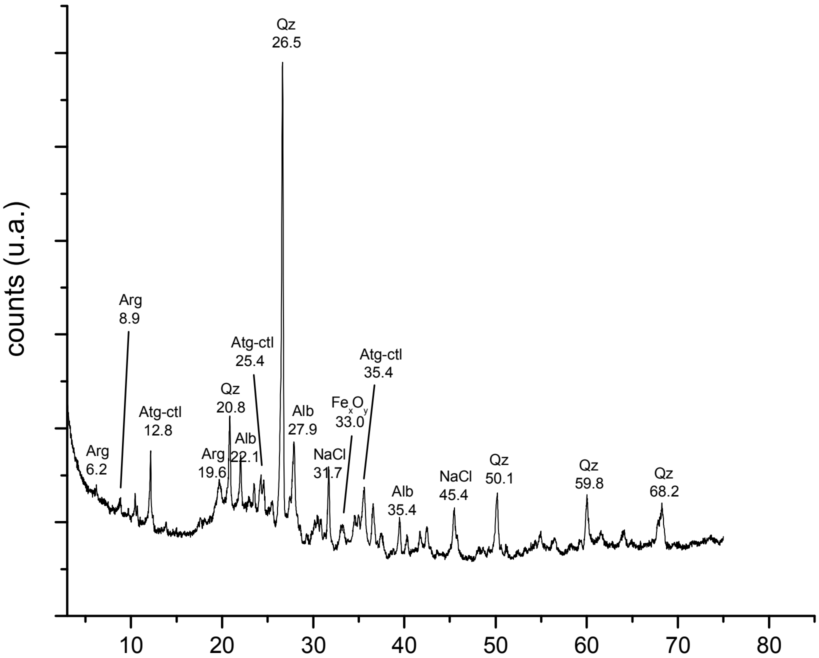

Fig. 4. Powder X-ray diffraction pattern (XRPD) of a representative topsoil sample from CF. Alb: albite, Atg-Ctl: antigorite-chrysotile (serpentine), Arg: clay minerals (e.g. smectite), Fe_xO_y: iron oxides (e.g. magnetite, hematite), and Qz: quartz. Sodium chloride (NaCl) was added as internal standard.

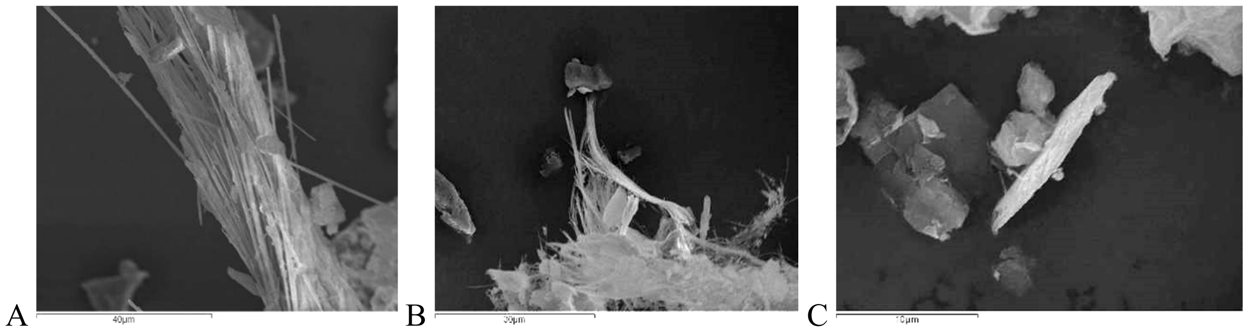

Fig. 5. Representative SEM images of fibrous mineral detected in the topsoil of CF. Morphological and chemical analyses (EDS) indicate: chrysotile (A, B), and fibrous smectite (C).

3.3 Exposure to airborne mineral fibers during the simulated agricultural activity

Tillage operations on the four sectors of the crop field were replicated in two independent trials (Day1 and Day2). Data, summarized in Table 3, indicate that in all cases the "farmer" was exposed to an asbestos concentration higher than 2 ff/L, the accepted environmental background level for asbestos and the threshold value for indoor environment after asbestos-removal work (e.g., Italian law on the remediation of indoor asbestos-contaminated environments, DM 6/9/94). In the two trials, the personal exposure to total inorganic inhalable fibers (according to the WHO definition, [30]) ranged around 42 and 68 ff/L and peaked up to 60 and 103 ff/L on specific field sectors (Table 3 - row A). Inhalable asbestos fibers (mostly chrysotile) ranged around 16 and 26 ff/L and peaked up to 25 and 40 ff/L, in the two trials respectively. Far from being representative of a time-weighted occupational exposure, our data suggest that the "farmer" may be exposed to a not negligible concentration of airborne asbestos, exceeding by a factor of 10 the 2 ff/L threshold. Interestingly, the only previous work dealing with agricultural activity performed on an asbestos-polluted soil reports about a similar airborne asbestos concentration (up to 16 ff/L, by SEM-EDS, fiber length > 5 μm) [23]. Lower concentrations of not inhalable fibers (length < 5 μm and/or diameter > 3 μm) were also detected (Table 3 - row B). The fiber concentrations did not significantly differ in the two trials ($p > 0.05$), but average values were higher (+60% and +70% for total fibers and asbestos respectively) on the trial performed after a drier period (Day2). Notably, environmental release of inhalable asbestos was only detected in the second trial (2.1 ff/L), with a value close to the 2 ff/L threshold. This possibly indicates that agricultural activity on asbestos-contaminated soils could have a limited impact on the contamination of the surrounding areas and may not pose a risk for general population. Additional field experiments, covering a wider range of agricultural practices and meteorological conditions, are needed to further support this evidence.

Table 3. Fibers detected in personal and environmental air samples. A, Inhalable fibers (L > 5 μm, d < 3 μm, L/d > 3). B, not-inhalable fibers (L < 5 μm o d > 3 μm, L/d > 3). Data are expressed as means ± standard deviation. The maximum value registered in the four sectors of the field is marked with *.

	Sampling day	Personal sampling		Environmental sampling	
		Asbestos fibers (ff/l)	Total fibers (ff/l)	Asbestos fibers (ff/l)	Total fibers (ff/l)
A	1	15.6 ± 7.4 (*24.9)	42.4 ± 12.7 (*59.8)	0.0	2.3
	2	26.4 ± 13.9 (*39.9)	68.1 ± 34.4(*103.0)	2.1	7.3
B	1	6.3 ± 2.5 (*7.5)	16.8 ± 11.2 (*32.4)	0.0	1.1
	2	5.0 ± 5.4 (*12.5)	13.1 ± 8.9(*24.9)	1.1	2.3

Asbestos represented ca. 40% of inhalable and non-inhalable fibrous minerals detected in both personal and environmental filters. Fig. 6 highlights the average occurrence of different fiber types within the different samples, as discriminated on the basis of morphology and EDS analysis. A Mg/Si ratio between 1.3 and 1.7 was used to discriminate fibrous serpentine, including both chrysotile and rare occurrence of fibrous antigorite (Ctl-Atg), from other asbestos, i.e. tremolite-actinolite (Trm-Act). Fibers showing a (Mg+Ca+Fe)/(Si+Al) between 1.3 and 1.7 were also identified as chrysotile-antigorite (Ctl-Atg*) to include those fiber which already underwent a partial pedogenetic transformation [18]. The vast majority of asbestos fibers could be ascribed to the fibrous serpentine. Representative SEM images and EDS elemental analyses of fibrous serpentine are reported in Fig. 7. Bundles of long, flexuous fibers , having the typical habit of chrysotile (Fig. 7A), were observed. Both micro-bundles and fibrils with nanometric diameters were imaged in such cases. Short, thin fibers, showing an intermediate habit between fibrous and laminar and an aspect ratio between 3 and 10 (Fig. 7B) were also observed. The only other asbestos type detected was fibrous tremolite-actinolite (Trm-Act), identified on the basis of the relative amount of Si, Ca, Mg and Fe. The lack of any other asbestos (e.g. crocidolite and amosite) indicates the absence of disturbance factors from anthropic sources (e.g. asbestos cement materials). Most of the non-asbestos fibers were clay minerals, which are known to occasionally display fibrous habit. Chemical compositions indicated smectite as the dominant clay mineral observed, accordingly to its abundance revealed in topsoil. The occurrence of fibrous smectite is consistent with previous report of airborne fibrous clay phyllosilicates found in similar environment [31].

These data show a strict mineralogical identity or a close pedogenetic relationship between the airborne fibers (par. 3.3), the fibrous component in waters (3.1) and top-soil (3.2), and the fibers formerly exploited in the Balangero mine [29]. Chrysotile (Ctl-Atg) and its weathered phases (Ctl-Atg*) are the predominant asbestiform minerals detected in the three investigated media (air, water, and soil). This mineralogical identity highlights the existence of a dispersion pathway where fibers move from the mining area to the Balangero plane via water and air transportation. Accordingly, human exposure to asbestos in the proximity of an asbestos-rich mine may not only derive from the exposed asbestos outcrops and the uncovered tailings in the mining area, but also from its agricultural surroundings, characterized by polluted soil and water.

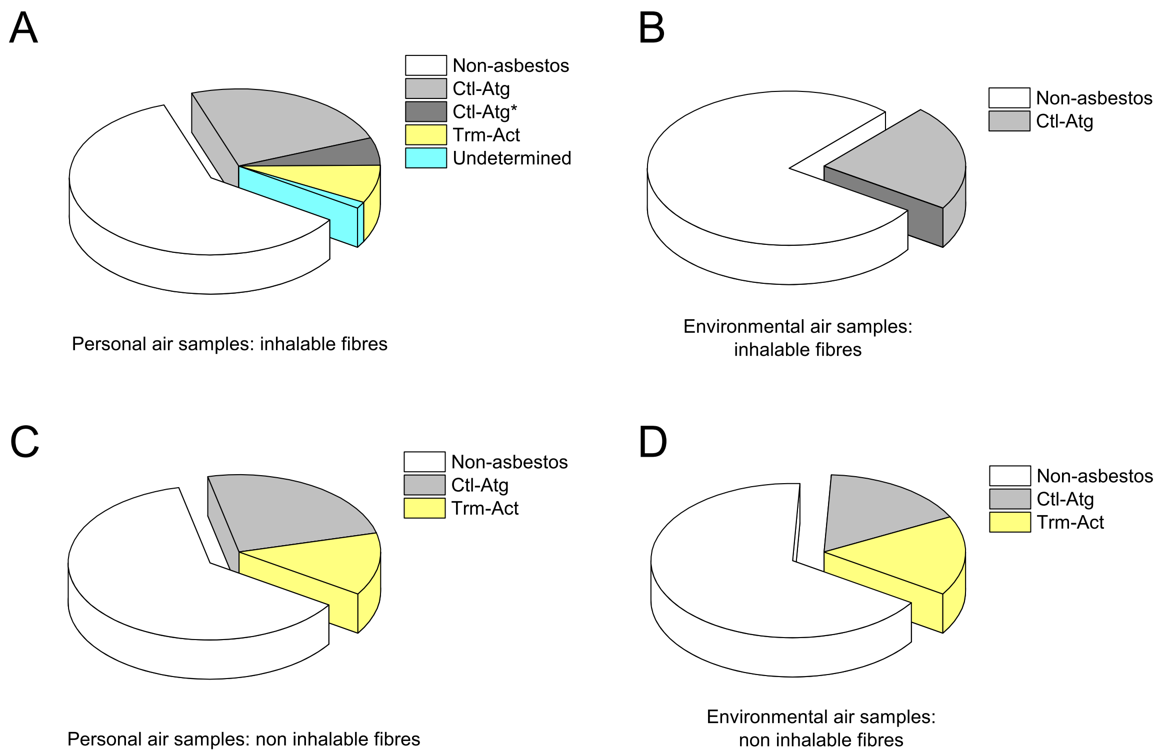

Fig. 6. Occurrence of different fiber types within the personal (A, C) and environmental (B, D) samples, differentiating between inhalable (A, B) and non-inhalable (C, D) fibers. Serpentine fibers: Ctl-Atg (1.3 < Mg/Si < 1.7); and Ctl-Atg* (1.3 < (Mg+Ca+Fe)/(Si+Al) < 1.7); fibrous tremolite-actinolite (Trm-Act).

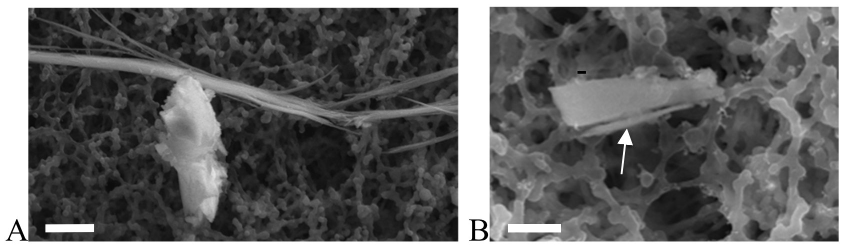

Fig. 7. Representative SEM images of serpentine fibers detected on the “farmer” personal samples. Scale bars, 5 µm (A) and 2.5 µm (B).

4. Conclusion

This study informs about the possible exposure of an agricultural worker to an inhalable concentration of asbestos fibers up to 40 ff/L, when a tillage operation is conducted in an asbestos-contaminated crop field. During two trials representing two different meteorological conditions, the worker was always exposed to an asbestos concentration much higher than 2 ff/L, the accepted environmental threshold. This work also confirms that asbestos fibers are widespread in the proximity of asbestos-mining sites and affect air, soil, and water quality. The relationship between the environmental proximity to an asbestos-rich setting and the risk of exposure during rural activities should be increasingly considered to enforce proactive policies of environmental safety.

Acknowledgments

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