Article

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Occurrence of asbestos in soils: state of the art

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In the last decades, it has been widely demonstrated the risk to human health related to asbestos fibres exposure. Many studies have mainly focused on the mineralogical and geochemical characterization of ophiolites (i.e., serpentinite and metabasite rocks) since they are the main lithotypes associated with Naturally Occurring Asbestos (NOA). Nevertheless, derivative soil from these rocks inherits the mineralogical and geochemical composition of bed rock and may contain hazardous fibres, thus making its examination necessary as well. This paper provides a summary of asbestos-containing soils investigation worldwide with the purpose of providing an overview of the data obtained so far. To this aim, the most relevant available literature, testifying the presence of fibrous minerals in soils have been considered. This allowed the global territory mapping in order to depict the distribution of natural asbestos in soils worldwide.

Introduction

Asbestos is a term used to indicate six fibrous silicate minerals belonging to serpentine (i.e., chrysotile) and amphibole (i.e., tremolite, actinolite, anthophyllite, amosite that is the fibrous-asbestiform variety of grunerite also known as brown asbestos, and crocidolite that is the fibrous-asbestiform variety of riebeckite, commercially known as blue asbestos; Gualtieri et al., 2017) super-group (WHO, 1986; NIOSH 2008; Ballirano et al., 2017). These minerals have been widely exploited to create Asbestos-Containing Materials (ACMs) due to their physical properties (Bloise et al., 2017a, 2018a, b; Bloise 2019a).

The term NOA means the asbestos fibres present in rocks (i.e., serpentinite or altered ultramafic rocks) and soils, referring to those that have not been extracted for commercial purposes (Bloise et al., 2008; Harper, 2008; Pugnaloni et al., 2013; Belluso et al., 2020; Bailey 2020a,b; Cahill 2020; Cagnard et al., 2020; Erskine 2020; Gualtieri 2020; Léocat 2020; Pierdzig 2020; Wroble et al., 2020). The natural asbestos occurrences is widespread in the environment, some examples include chrysotile deposits in Ural Mountains in the Russian Federation (Ross and Nolan, 2003), Appalachian Mountains (USA), Canada (Virta, 2006) and also in India, China, Italy, South Africa, Australia, Greece, Cyprus (Ross and Nolan, 2003) and other countries. Figure 1a, shows a global map with the main asbestos mines. The most frequent asbestos occurring form is chrysotile, whose fibres are normally found as veins in serpentine rocks, followed by anthophyllite, crocidolite, tremolite, actinolite and amosite (Virta, 2002; Bloise et al., 2019b).

Human activity and weathering processes may disturb NOA and provoke the dispersion of fibres, potentially inhalable, in the environment. Many studies confirmed that death from lung diseases can be associated with environmental exposure to asbestos (IARC, 2009). In fact, the risk to human health is represented by the inhalation of asbestos fibres that penetrate in the lungs and may cause cancer pathologies. It is worth noting that, fibrous minerals such as, erionite, ferrite fluoro-edenite, antigorite (Gianfagna et al., 2003; Cardile et al., 2007; Ballirano et al., 2018a,b; Gualtieri et al., 2018; Petriglieri et al., 2020) and others, may have toxic effects as asbestos fibres and, if inhaled, can be dangerous.

To date, many studies are based on the knowledge of natural asbestos in rocks whereas much less literature refers to asbestos-bearing soils. Asbestos in soils may be found for: i) improperly removal of Asbestos-Containing Materials; ii) proximity to asbestos factory/ mine; iii) inheritance from mother rocks (natural occurrences).

In this scenario, this work aims to provide an overview about the presence of asbestos fibres in soils worldwide pointing to improve knowledge of asbestos global issue. Therefore, the most relevant study from various disciplines (i.e., geology, mineralogy, medicine, etc.) that testified the presence of asbestos in soil were hereby considered, thus making it available an overview of the data obtained so far and providing a contribution to the mapping of the territory (Fig. 1b).

Because of considerable asbestos-related diseases (Skinner et al., 1988) all of the six asbestos minerals are considered toxic to human health and therefore regulated by law. However, currently only in 67 over 195 countries (34%) in the world the use of regulated asbestos minerals is restricted (Table 1). Since many studies provide the epidemiological evidence that asbestos crocidolite and tremolite are apparently more dangerous than chrysotile (Hodgson et al., 2000), many countries employ it since this is considered a "safe use" for industrial purposes. For example, Russia is the biggest producer in the world (tons/year) followed by China, Brazil, Kazakhstan and India as shown by the 2014 (chrysotile) asbestos trade data (Gualtieri, 2017).

The different global use of asbestos is due to the various political and economic situation of countries and it is constantly changing. For instance, some countries like Canada and Colombia have recently changed their regulations relatively to asbestos exploitation and use. Indeed, in



Colombia the new law is taking effect on January 1st, 2021 with five years transition period for companies currently using asbestos minerals. The ban prohibited the mining, commercialization and distribution of all asbestos types including its export. In the case of Canada, despite asbestos fibres have been recognized as hazardous to human health and well-being (World Health Organization's International Agency) for more than 30 years, Canada remained one of the major exporter of this material until 2011. The federal Prohibition of Asbestos and Products Containing Asbestos Regulations (Regulations: SOR/ 2018-196) came into force in Canada on January 2019, prohibiting the import, sale and use of asbestos as well as of products containing asbestos. However, there are certain exceptions such as asbestos contained in household product intended for personal use or in a military equipment (IBAS, 2019).

Even in the scientific community a division is noted: some scien-

tists promote the "safe use" of chrysotile and assume that it has little potential for causing mesothelioma (e.g., Liddell et al., 1997; McDonald et al., 1997; Camus, 2001) whereas others are totally opposed to this and claim that all six asbestos types may induce lung diseases if inhaled (Skinner et al., 1988; Yarborough, 2007). Therefore, the International Agency for Research on Cancer define them belonging to Group 1 "substance carcinogenic to humans" (IARC, 2012).

It is difficult to univocally define the relationship (cause-effect) between disease and exposure to different fibres types because of the variability in the chemistry, size, molecular arrangement, surface activity (Pollastri et al., 2014; Bloise et al., 2016b) of mineral fibres. For instance, one hypothesis of higher toxicity of amphiboles compared to chryso-tile is based on the behavior of fibres in the lungs. In particular, unlike the amphiboles that are more durable and remain in the lungs for a long time, chrysotile dissolves reasonably quickly due to its low biodura-

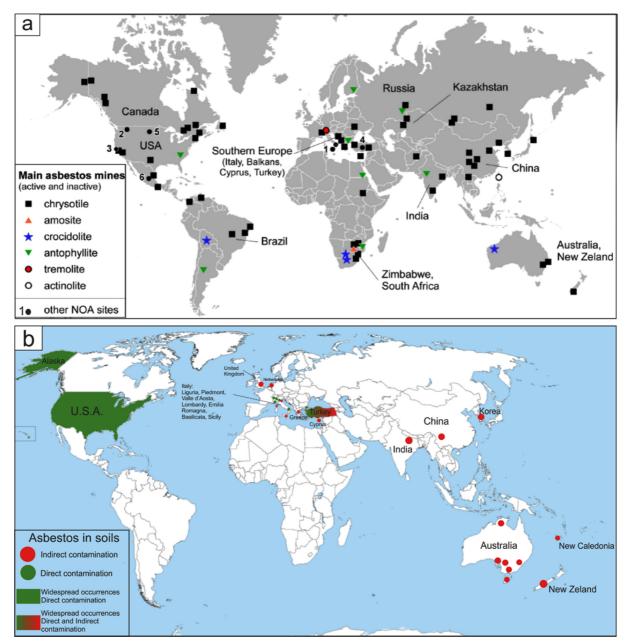


Figure 1. a) global map with the main asbestos mines (active and inactive), modified after Virta 2002; b) global map with the asbestos-containing soil at the state of the knowledge.

estosi Zist compilen sy Zi	National Asbestos Bans						
Algeria	Czech Republic	Iraq	Mauritius	Seychelles			
Argentina	Denmark	Ireland	Monaco	Slovakia			
Australia	Djibouti	Israel	Mozambique	Slovenia			
Austria	Egypt	Italy	Netherlands	South Africa			
Barain	Estonia	Japan	New Caledonia	Spain			
Belgium	Finland	Jordan	New Zeland	Sweden			
Brazil	France	Korea (South)	Norway	Switzerland			
Brunei	Gabon	Kuwait	Oman	Taiwan5			
Bulgaria	Germany	Latvia	Poland	Turkey			
Canada	Gibraltar	Liechtenstein	Portugal	United Kingdom			
Chile	Greece	Lithuania	Qatar	Uruguay			
Colombia	Honduras	Luxembourg	Romania				
Croatia	Hungary	Macedonia	Saudi Arabia				

Malta

Table 1. Exemptions for minor uses are permitted in some countries listed; however, all countries listed must have banned the use of all types of asbestos. List compiled by Laurie Kazan-Allen and modified and revised in July 15, 2019 (http://www.ibasecretariat.org/alpha_ban_list.php)

bility (Hume and Rimstidt, 1992; Bernstein et al., 2008; Oze and Solt 2010). In this work, we summarize and discuss data of the most relevant studies available in literature which testify the presence of asbestos fibres in soils worldwide.

Iceland

Methods and Materials

Cyprus

In this work, we carried out a literature search on articles of various disciplines that appeared over the past thirty years and subdivided them according to the source of contamination: i) indirect contamination; and ii) direct contamination (Table 2).

Results and Discussion

The articles that reveal the presence of asbestos fibres in soils due to indirect contamination are mainly of three types: i) case-report; ii) case-control study; iii) environmental study.

The first two are mainly epidemiological studies based on the investigation of documented cases of pathologies consequently to the occupational and environmental exposition to asbestos. In particular, case-report consists in a detailed description of an individual case whereas case-control study involves the comparison of two existing groups differing in outcome (e.g., effected by issues: case; non-effected: control).

Case- reports and case-control studies involved in the present work, revealed that in most cases the contamination is due to the presence of fibres in materials used for aims like dirt roads (Baris et al., 1987; Viallat et al., 1991; Luce et al., 2000; Comba et al., 2003; Luo et al., 2003), and whitewash (Constantopoulos et al., 1991; Sichletidis et al., 1992; Sakellariou 1996; Luce et al., 2000; Metintas et al., 2002).

For instance, Luce et al. 2000, carried out a study on respiratory cancers in New Caledonia, where a high incidence of malignant pleural mesothelioma had been observed. In particular, a case-control study has been conducted in regard to the association between tremo-

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lite exposure and the risk for respiratory cancer from different sites. Results revealed that the risk of pleural mesothelioma was associated with exposure to whitewash (Table 3) that have widely been used for indoor and outdoor walls of houses.

Serbia

Differently, the other articles taken into consideration are based on environmental and monitoring study where the source of contamination is mainly represented by asbestos removal operations (Davies et al., 1996; Gualtieri et al., 2009), asbestos in material used for construction (Famoso et al., 2012) or asbestos waste in soil (Driece et al., 2010; Bint et al., 2017), proximity to asbestos mine (Gualtieri et al., 2014; Lee et al., 2015; Turci et al., 2016) as well as asbestos cement factory, widely documented in India (Musthapa et al., 2003; Subramanian et al., 2005; Trivedi et al., 2011, 2013) where the use and production of asbestos is not banned yet.

Gualtieri et al. 2009, presented the results of a monitoring activity conducted on particulate, fall-out and soil samples of selected inhabited areas in Italy (i.e., Emilia Romagna region). The aim of the work was to detect asbestos content in air and the risk of exposure for the population in addition to the assessment of the nature of other mineral phases composing the particulate matrix. To this purpose, various analytical techniques have been used such as XRPD, PLOM, SEM, TEM. In the specific case of the analyzed soils, asbestos fibres were found in samples taken from a residential zone of Sassuolo and near the Bologna Central Railway Station. According to authors, in the first case the contamination is likely due to asbestos removal operations of ACMs whereas in the second case, natural dispersion from ophiolite rock used as track ballast represent the source of contamination. For the results interpretation they elaborate a general model of environmental asbestos pollution (Fig. 2a) referring to the pollution mechanism proposed by Chiappino et al., 1993. The latter consists of a primary and secondary pollution stages. In the primary, the dispersing materials release coarse fibres which settle near to the source because of their higher mass and at the same time release ultra-fine materials. In the secondary, the settled fibres break up into ultra-fine and ultra-short fibrils that are able to remain suspended in the atmosphere for long periods thanks to their minimal mass.

Table 2. Literature data of asbestos-containing soil

Place	Fibres type	Source of contamination	Methods	Reference
Australia	Amosite, Crocidolite, Chrysotile	Indirect (ACMs in the soil)	Case - report	Genever et al., 2017
China (Da-yao)	Crocidolite	Indirect (Dirt roads, stucco, dishes)	Review of clinical/epidemiological studies	Luo et al., 2003
Corsica	Tremolite	Indirect (use in the flooring)	Case - report	Viallat et al., 1991
Cyprus	Tremolite Crisotile	Indirect (Stucco, gutters)	Case - report Radiological studies on population	McConnochie et al., 1989
Greece	Tremolite, Chrysotile	Indirect (Whitewash)	Case – report XRPD	Constantopoulos et al., 1991
Greece (Macedonia)	Tremolite, Chrysotile	Indirect (Whitewash)	Case – report X-ray study on population	Sichletidis et al., 1992
Greece (Metsovo)	Tremolite	Indirect (Whitewash)	Case – report X-ray study on population	Sakellariou et al., 1996
India (Mohanlalganj, Lucknow)	Chrysotile	Indirect (Vicinity to Asbestos cement factory)	РСОМ	Subramanian et al., 2005; Trivedi et al., 2011, 2013; Musthapa et al., 2003
Italy (Basilicata)	Tremolite	Direct (Natural occurrences)	Case – report; OM, SEM-EDS, TEM-EDS XRPD, XRF, DTG, DSC	Bernardini et al., 2003; Pasetto et al., 2004; Bloise et al., 2016, 2018; Punturo et al., 2018, 2019
Italy (Emilia-Romagna)	Serpentine asbestos	Indirect (Asbestos removal operations); Direct (Natural occurrences)	PLOM, XRPD, SEM, TEM	Gualtieri et al., 2009
Italy (Liguria)	Tremolite, Actinolite, Chrysotile	Direct (Natural occurrences)	SEM-EDS	Barale et al., 2020; Militello et al., 2019; Turci et al., 2020
Italy (Lombardy)	Chrysotile, Tremolite	Direct (Natural occurrences)	SEM-EDS	Cavallo et al., 2020
Italy (Piedmont)	Chrysotile	Indirect (Vicinity to asbesos mine)	μXRF, XRPD, SEM	Turci et al., 2016
Italy (Sicily)	Fluoro-edenite	Indirect (Quarries, dirt roads, use in mortar and plasters)	Case – report; PCOM, SEM-EDS	Comba et al., 2003; Famoso et al., 2012
Italy (Valle d'Aosta)	Tremolite, Chrysotile	Indirect (Vicinity to asbestos mine)	OM, XRPD, FTIR, SEM, DTA	Gualtieri et al., 2014
Korea (Hongseong; Janghang)	Chrysotile, Tremolite, Actinolite	Indirect (Vicinity to asbestos mine), Direct (Natural occurrences)	PLM, XRD, PCM, FE-EDS, SEM-EDS, TEM-EDS	Lee et al., 2015; Yoon et al., 2020
Netherlands (Hof van Twente)	Crocidolite, Chrysotile	Indirect (Asbestos waste in soil)	TEM on air samples	Driece et al., 2010
New Caledonia	Tremolite	Indirect (Whitewash, dirt roads)	Case – control study	Luce et al., 2000; Petriglieri et al., 2020b
New Zeland	Asbestos fibres	Indirect (Construction waste)	Guidelines	Bint et al., 2017
Turkey (Anatolia)	Tremolite, Actinolite, Chrysotile	Indirect (Whitewash, stucco, terra- cotta); Direct (Natural occurrences)	· Cohort study; XRPD	Metintas et al., 2002, 2017
Turkey (Cappadocia)	Erionite	Indirect (Dirt roads, brick)	Case - control study	Baris et al., 1987
United Kingdom	Amosite, Crocidolite	Indirect (Asbestos removal operations)	PCOM quantitative study	Davies et al., 1996
USA	Amphibole asbestos	Direct (Natural occurrences)	XRD, SEM	Thompson et al., 2011
USA (California)	Chrysotile, fibrous amphiboles	Direct (Natural occurrences)	TEM-EDX Electron diffraction analysisi	Bailey 2020a
USA (Nevada)	Actinolite, Fibrous Erionite	Direct (Natural occurrences)	SEM-EDS, FE-SEM, XRD	Buck et al., 2013; Ray 2020
USA (Whashington)	Chrysotile, Actinolite	Direct (Natural occurrences)	PLM	EPA, 2009

Concerning direct contamination, according to the most relevant studies it has been recognized into three countries: Italy, Turkey and USA. In Italy, many studies have been conducted in the Basilicata and Calabria region (Campopiano et al., 2018; Bloise et al., 2019c; Colombino et al., 2019; Dichicco et al., 2019; Laurita and Rizzo, 2019) where tremolite is the main asbestos mineral found in soils (Pasetto et al., 2004; Bloise et al., 2016a; 2018a,b; Punturo et al., 2018, 2019). Tremolite, in actinolite and chrysotile has also been observed in soil samples investigated by Militello et al., 2019 (Liguria region), whereas samples analysed by Gualtieri et al., 2009, testify the presence of serpentine asbestos in soils occurring in Valle d'Aosta region.

For example, the purpose of the study carried out by Bloise et al., 2016a was to assess the occurrence of asbestiform minerals in serpentinite and serpentinite-derived soils cropping out in the area of Sila Piccola. To this aim, they characterized both serpentinite and agricultural soil samples by means of various analytical techniques such as

Table 3. Pleural mesothelioma risk associated with exposure to whitewash, New Caledonia, 1993-1995. *Odds ratio adjusted for age and gender; F Numbers in parentheses, 95% confidence interval (Modified after Luce et al., 2000)

Exposure	No. of cases	No. of controls	Odds ratio*
Never exposed	1	223	1
Ever exposed	14	82	40,9 (5.15, 325) I
Exposure duration			
< 20 years	4	38	22.2 (2.33, 211)
\geq 20 years	10	41	65.1 (7.69, 551)
Age at first exposure			
Birth	13	61	52.8 (6.53, 427)
≤ 16	1	11	20.0 (1.09, 368)
>16	0	10	0

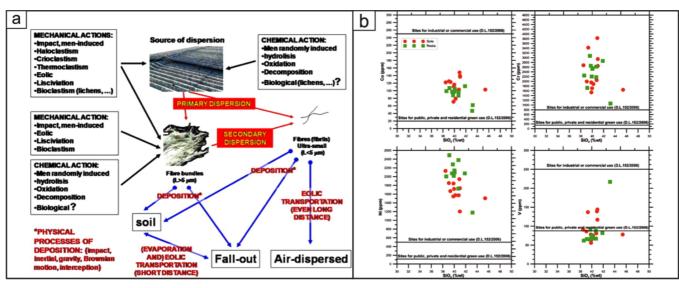


Figure 2. a) Model of asbestos fibre dispersion from cement-asbestos (modified after Gualtieri et al., 2009); b) Correlation diagrams of SiO₂ versus Co, Cr, Ni and V for soils and rocks of the studied area (after Punturo et al., 2018). Thresholds values regulated by Italian law (D.L.152/2006) are also indicated for each heavy metal.

Table 4. Mineralogical assemblage detected by PLM, XRPD, SEM/EDS, DSC/TG, µ-R reported in order of decreasing abundance. PS=polygonal serpentine, Atg= antigorite, Lz= lizardite, Ctl=chrysotile, Tr-Act=tremoliteactinolite, Mag=magnetite, Chl=chlorite, Ms=muscovite, Ab=albite, Qtz=quartz, Cal=calcite, Rt=rutile, Sp=spinel (Bloise et al., 2016)

Sample	Phase detected	
CNF-S1	Clay>Qtz>Tr-Act>Chl>Ms>Ctl>Liz>Ab>Atg>Rt	
CNF-S2	Clay>Qtz>Chl>Ms>Tr-Act>Ctl>Ab>Rt>Liz	
GML-S1	Clay>Qtz>Chl>Ms>Tr-Act>Ctl>Ab>Liz>Rt	

PLOM, XRPD and SEM/EDS. Moreover, for a better discrimination of serpentine polytypes, Differential Scanning Calorimetry, Thermogravimetric and μ -Raman spectroscopy were used. Results show high amount of chrysotile and asbestos tremolite-actinolite in agricultural soils (Table 4).

Instead, Punturo et al. 2018 conducted a detailed mineralogical and geochemical investigation of both rocks and soil collected in the Basilicata region, with the aim to understand their potential contribution to human health caused by asbestos exposure. Therefore, the presence of asbestos

fibres (chrysotile and asbestos tremolite) and the concentration levels of toxic elements (Cr, Co, Ni, V) have been determined. In the specific case, in almost all samples, detected values exceed the regulatory thresholds for public, private and residential green use (D.L. 152/2006; Fig. 2b).

In the other two countries, fibrous amphiboles and chrysotile are the main fibres detected. In particular, a total of 1251 soil samples in Anatolia region (Turkey) were collected and analyzed by Metintas et al., 2017 (Fig. 3a). XRPD analysis results revealed that chrysotile, tremolite or mixed asbestos fibres were contained in 514 soil samples.

In USA, high occurrence of actinolite and chrysotile has been monitored (EPA, 2009; Thompson et al., 2011; Buck et al., 2013). Thompson et al., 2011 discussed the geographic distribution of amphiboles in the USA, using the mineralogical data from selected sand and/or silt fraction of soils from the USDA-NRCS National Cooperative Soil Survey database, which shows the presence of amphiboles in all states except for Rhode Island. A total of 212,839 horizons (layer within soil with unique morphological characteristics) within 34,326 pedons (body of soil that consists of all the horizons at that location) were sampled (Fig. 3b).

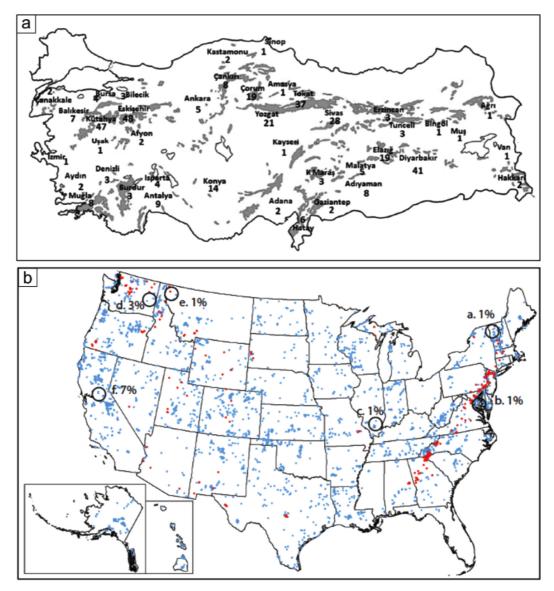


Figure 3. a) Map of Turkey showing the distribution of villages where asbestos exposure was definite (after Metintas et al., 2017; b) Map of the U.S.A. showing asbestos-containing soils (more detail from Thompson et al., 2001))

On the basis of data available in literature of the past thirty years, it is possible to draw up various considerations. First of all, asbestos-related diseases are certainly of significant concern in terms of occupational and public health. Indeed, many studies confirmed a significant malignant mesothelioma risk due to asbestos environmental exposure (Baumann et al., 2016; Liu et al., 2017). Compared to NOA in rocks (Bloise et al., 2019a), asbestos content in soils is today poorly investigated. The soil contamination plays an important role relatively to the environmental exposure especially if soils are used for agricultural purposes, becoming a risk for both workers (Bellomo et al., 2018) and people living near NOA (non-occupational exposure; Bloise et al., 2012; Pugnaloni et al., 2013; Bloise et al., 2017b; Bloise et al., 2018c; Pinizzotto et al., 2018; Punturo et al., 2018). According to Januch et al., 2013, people exposure derived from disturbance of asbestos-contaminated soil, is mainly investigated by measuring asbestos concentration in breathing zone air during soil-disturbance activities. Nevertheless, in our opinion the determination of asbestos content in top-soil or sub-soil is essential as it constitutes the primary threat to health. Indeed, the tendency of fibres is to settle out of air and water and deposit in soil (EPA, 1979). Moreover, some fibres are sufficiently small to remain in suspension and can be transported for long distance thus increasing the contamination area. It is worth specifying that, asbestos itself is very stable and, if not disturbed can remain in the soil indefinitely (ATSDR, 2001). Indeed, concerning ACMs asbestos is bonded in a matrix which does not favour the release of free respirable fibres unless it is extremely weathered, or exposed to acid material (NEPC, 2011). However, many common forms of asbestos containing materials may slowly degrade if left in soil leading to more asbestos fibres being released over time (Bint et al., 2017). Since the extensive use in the past, ACMs represent a persistent source of environmental pollution despite the cessation of asbestos mining and legal prohibitions adopted by many countries. It is estimated that about 150 million m² of asbestos-based products and more than two thousand million m² of cement-asbestos roofings are still present today (Gualtieri et al., 2009).

In summary, the source of asbestos contamination in soils can be indirect and direct. In the first case, it is originated by asbestos fac-

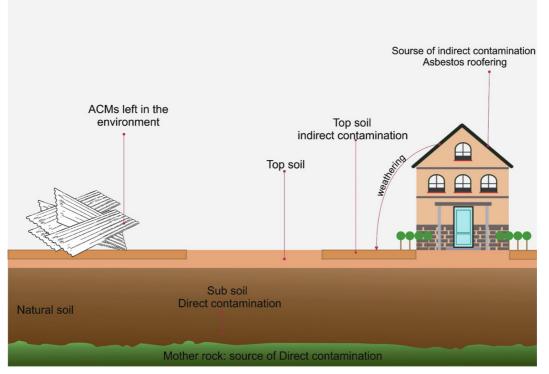


Figure 4. The image shows where asbestos fibres can be found depending on the source of contamination (direct/indirect).

tory, mines and ACMs subject to weathering or improperly removed. Direct contamination instead, derive by the disturbance of natural deposits by weathering or anthropic activity which cause the dispersion of fibres into the environment.

Depending on the source of contamination, asbestos fibres can be found in different portion of soil. Indeed, to verify the presence of fibres in the surrounding of asbestos factory/mine it is important to sample the top-soil in the proximity of the emission point. Differently, in the case of direct contamination, sub-soil samples are more representative since the presence of fibres is not due to deposition but rather to the characteristics of the mother rock. Figure 4, summarize where fibres can be found depending on the contamination source.

Observing the collected data, it is possible to notice that the indirect contamination has been widely detected in the investigated soils and their distribution cover a big range of the territory. This is likely due to the high use and commercialization of asbestos in the past, which resulted in his exportation all over the world.

Instead, since natural asbestos occurrences depends on the geology of the area, the presence of contaminated soils is limited to the distribution of the mother rock outcrops.

In recent years a new line of research asserts that the ingestion of asbestos by drinking water containing fibres, represents another factor that may cause an increase in cancer incidence in expose populations (DHHS Committee, 1987) although this issue is still debated (Rodelsperger et al., 1991; Di, 2017; Li et al., 2019). The possible sources of contamination include erosion of rock, industrial effluents, disintegration of asbestos-containing products (WHO, 2000, 2004). In this context, it is essential to identify the source dispersion and act on it. Therefore, knowledge on contaminated soil as well as territory mapping need to be improved.

Conclusions

In this study, a detailed examination of the most relevant literature studies revealing the presence of asbestos in soils worldwide, has been conducted. The aim of this work is to provide an overview about the distribution of asbestos-containing soils, thus improving the global mapping and the knowledge obtained so far.

Main results from our investigation showed that the presence of asbestos content in soils is still poorly studied. At this time, many studies are mainly based on the measuring of asbestos concentration in breathing zone air during disturbance activities. Moreover, several studies provide epidemiological and experimental evidence that trace metals may provoke lung cancer thus making necessary their investigation. In this scenario, the identification of the source of contamination and actions on it are essential.

For instance, one way to locate the potential "asbestos-containing soil" is to: i) identify the natural outcrops of rocks that may contain asbestos and investigate on derivative soil (natural contamination); and ii) identify old factory and asbestos mine (even the abandoned ones) and examine the surrounding soil (indirect contamination). As far as ACMs is concerned, it may be more difficult because of their high diffusion all over the world due to the extensive use in the past, but removal or renewal action are essential.

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