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Outdoor fibres air pollution monitoring in the Crotone area (Southern Italy)

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ABSTRACT - This paper shows the results of a monitoring activity of airborne fibres in the central eastern edge of Calabria, including the cities of Crotone and Strongoli (South Italy). The investigated area is actually occupied by largely disused industrial settlements and areas adjacent to them, where activities related to biomass power plant and agricultural production are carried out. The aim is to ascertain whether such an use of the territory caused changes in the natural levels of fibres in the air and subsequently a risk of exposure for the population. Moreover, establish the average level of exposure to asbestos and consequently the background environmental concentration of asbestos in air. The sampling was conducted on the air particulate of Strongoli and Crotone cities, industrial settlement of Strongoli Marina and disused industrial area of Crotone (ex Pertusola). Fibres had been collected for all the sites, from May to September 2012. For each investigated site (no. 8), areas potentially poor of pollutants had been selected as blank spots in order to appreciate the background environmental concentration of fibrous minerals in the air. The collection asbestos fibers were performed by using low-medium flow sampling systems, with cellulose filters of 25 mm in diameter over a short time period (6 or 8 h). The surprising outcome of this work is that, despite the high presence of asbestos roofings in the sampled areas, the level of dispersed asbestos fibers (chrysotile and tremolite) is very low.

Keywords: Asbestos fibers; ambient living environments monitoring; Crotone and Strongoli cities.

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1. INTRODUCTION

Asbestos exhibit outstanding properties that have been exploited to create asbestos containing materials (ACMs) used in a huge number (more than 3000) of practical and industrial applications and for manufacturing various types of products (asbestos cement, eternit, disc brake pads, pipes, reinforcing agents, fire retardants and many more) (Gualtieri, 2017). The major chemical-physical and technological properties of commercial asbestos are resistance to abrasion, resistance to heat (non-flammable even at very high temperatures) and to chemicals, flexibility, resiliency, low sound transmission coefficient, high surface area, extremely high tensile strength, low thermal conductivity (Bloise et al., 2016a; Gualtieri, 2017). Chrysotile is by far the predominant asbestos fibre ever used. The asbestos-cement industry is the largest user of chrysotile fibres (about 85% of all applications).

It is estimated that more than 95% of the commercially developed asbestos ore deposits were chrysotile asbestos (Ross et al., 2008).

The urban area of Crotone is considered one of the locality with the highest concentration of asbestos in Italy (European Environmental Agency's: https://www.eea.europa.eu). Moreover, the territory is actually still occupied by largely disused industries. Recently, the World Health Organization recorded in Crotone an average increase in the incidence of carcinoma (Cannata, 2016). An epidemiological study carried out by the Strategic Program of Health and Environmental Ministry of Italy numbered the city of Crotone among the list of 57 sites of national interest (SIN: http://www.regione.calabria.it/ambiente) as settlements exposed to risk of pollution and to be decontaminated (Comba and Pitimada, 2016).

The study analyzed 63 groups of causes of mortality in 1995-2002 with excess of mortality for malignant tumors of the pleura, both for men and women. The increase in mortality reflects an actual increase in the risk of incurring pleural mesothelioma because of pollutant exposition in the city area of Crotone. Human health can be damaged by inhaling fibres, when they become airborne due to weathering or human activities that produce dust (Harper 2008; Bloise et al., 2012, 2016b, 2017; Gaggero et al., 2013, 2017; Baumann et al., 2015; Punturo et al., 2015).

On the basis of the effects of asbestos on biological systems, several authors ascribe the fibre toxicity to the synergetic effect of fibre size, crystal habit, surface reactivity, ability to generate reactive oxygen species (ROS), biopersistence and chemical composition (e.g., Dodson et al., 2003; Pugnaloni et al., 2013; Bloise at al., 2016c).

For this reason, they are classified as carcinogenic substances group1 "substance carcinogenic to humans" by the Agency for Research on Cancer (IARC, 1987). In most studies, several patients with mesothelioma do not report any occupational or other exposure to asbestos and thus, there seems to be a small spontaneous background incidence of the cancer (Enterline and Henderson, 1987; Yates et al., 1997; Hillerdal et al., 1999; Gualtieri et al., 2009).

In fact, the global scientific community also acknowledged the fact that there is no evidence of a threshold level of exposure to asbestos fibres below which there is no risk of mesothelioma (Ramazzini, 2010). Hence, a discussion of the risks from low exposure must include the determination of the background concentration.

Our study is focused on ambient monitoring of fibres air particulate (no. 8 sites), in particular asbestos, in strategic planning sites, like the industrial area of Crotone, the biomasses power plant of Strongoli village, the schools of Crotone and Strongoli, during a narrow time period. The aim of the paper is to investigate the mineralogical composition, amount and sources of airborne fibres on Crotone and surrounding area (Southern Italy) where approximately 150,000 people live and could be potentially at risk of toxic fibers exposure.

2. AREA DESCRIPTION

The sampling campaign had been planned on strategic sites in the city of Crotone and Strongoli (Cannata, 2016) (Figs. 1, 2). The monitoring involved industrial agglomerations and areas of public use, like infant or high schools that are potentially manufactured with cement-asbestos (Figs. 3, 4). The sampling was carried out during spring-summer. In detail, four sampling campaigns were performed on the month of May, June, July and September 2012 (Tab. 1).

Anemologic information of wind direction, frequency and average speed were collected (Appendix I).

Moreover, in order to detect the background environmental concentration of asbestos in the air, air samples from asbestos-free areas (background level) have been also collected.

Since the prevailing winds come from NE, we refer ad background level "Villa Ermenegilda" agritourism (CP1, Tab. 1) for the Crotone area, which is located at about 6.8 km north of the center of Crotone and the football field (SP1, Tab. 1) for Strongoli respectively. The latter is located on the NE edge of Strongoli in a vegetation free place and far from asbestos roofings (Tab. 1).

The campaign Crotone lasted 4 days: from 25 to 29 June, and interested the urban center of Crotone. The monitoring was made on three sites. "Alfieri" station (CP5, Tab. 1) has been located near schools such as areas of public domain in a crowded area close to a road junction and a disused area with asbestos roofings.

The station CP7 had been placed in the garden of Consortium Industrial Development located in the industrial area of Crotone 80 m far from the road E90 and less than 1 km NW from the industrial area comprising warehouses and industrial plants interspersed with various green areas or land lacking of vegetation. The station CP8 was placed in the garden at the Regional Services and Development in Agriculture, at approximately 700 m NW from the CP7 station and about 20 m far from the road E90.

These sites were the headquarters of the metallurgic industrial plant of electrolytic zinc "Pertusola", born in 1924 on the initiative of the French "Sociétè Minière et Mètallurigique de Pennaroya" and closed down the production on February 1999. The campaign Strongoli was conducted in the yard of the public school of Crotone "A. Lucifero" (SP6 station) "Pianette school" (SP3 station), at about 25 m from the Provincial Road 51 and 1.4 km far from the sea.

3. METHODS AND MATERIAL

3.1. Dardos Control System

An efficient, fast and cheap method for detection of eternit roofings in urban environment is represented by Dardos Control (http://www.datageo.it) from DataGeo s.r.l. Dardos Control system is useful to reproduce maps according to the textural features of remote sensing images in order to identify illegal landfill, quarry and pits, eternit roofings, river channels, etc. (Fig. 4). The system employs algorithms that allow textural analysis of the images by processing their visible radiation spectrum only. As opposite to traditional remote sensing-based investigative method Dardos control exploits the typical textures of eternit roofings, pits, deposits or relics, as detected from aircraft or satellite remote sensing images within the visible radiation spectra. High resolution aerial photo or free satellite imagery are filtered highlighting areas with the typical textural features of the examined object (http://www.datageo.it).

Textural property of Eternit roofings are easily recognized but further investigation on the field are anyway necessary. Dardos control system supplies a cartographic support equipped with coordinates and address of edifice having eternit roofs.

The identification of eternit roofs based on color, shape and texture of the artifacts is followed by a visual and systematic control of points detected for the elimination of fake positive.

The percentage of false positives was estimated to be in an order of 10 percent for Crotone images, whose quality (res 50 cm/px) is higher than Strongoli ones for which the

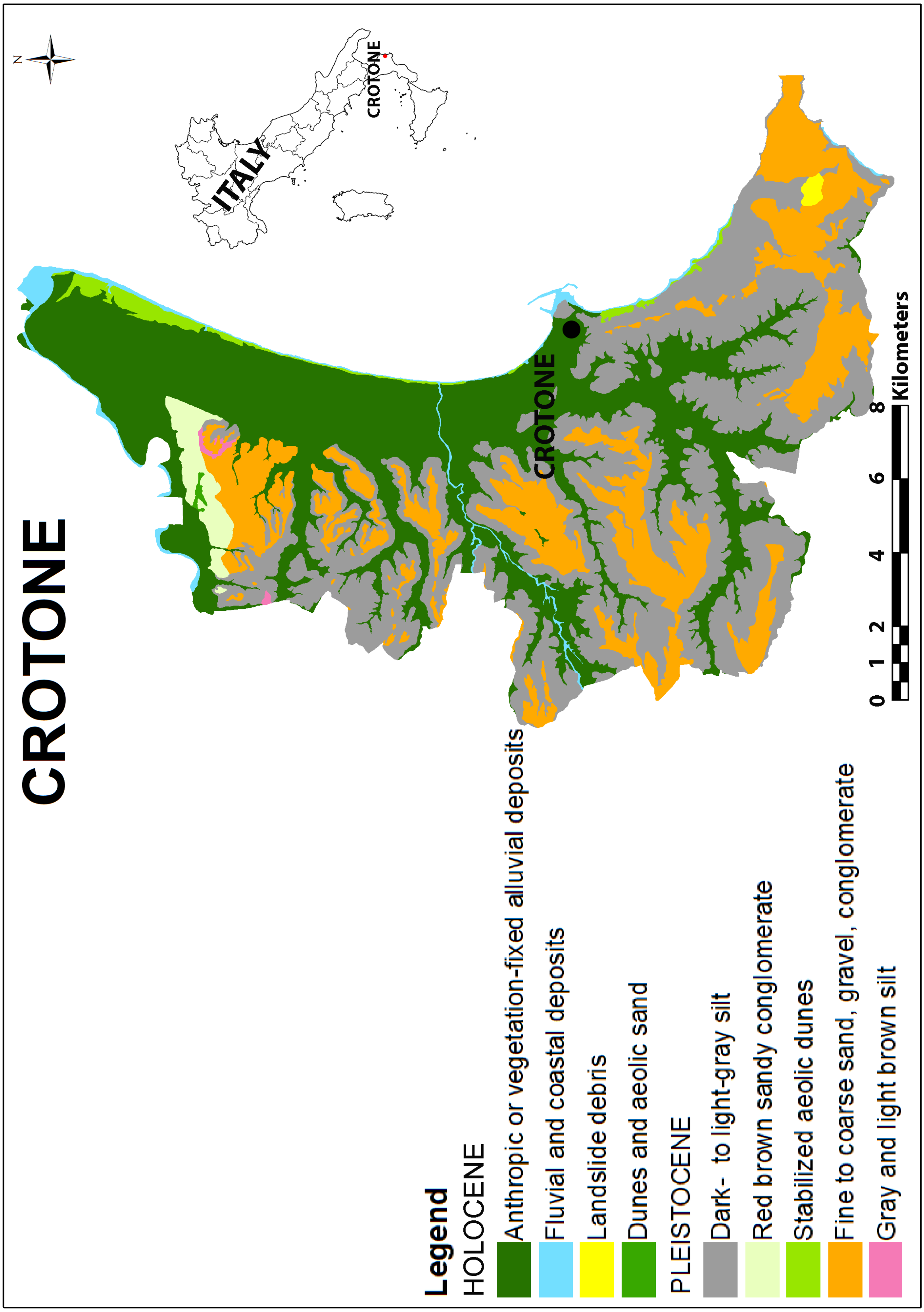

Fig. 1 - Geological maps of Crotone (UTM: 684088.402, 4328119.858).

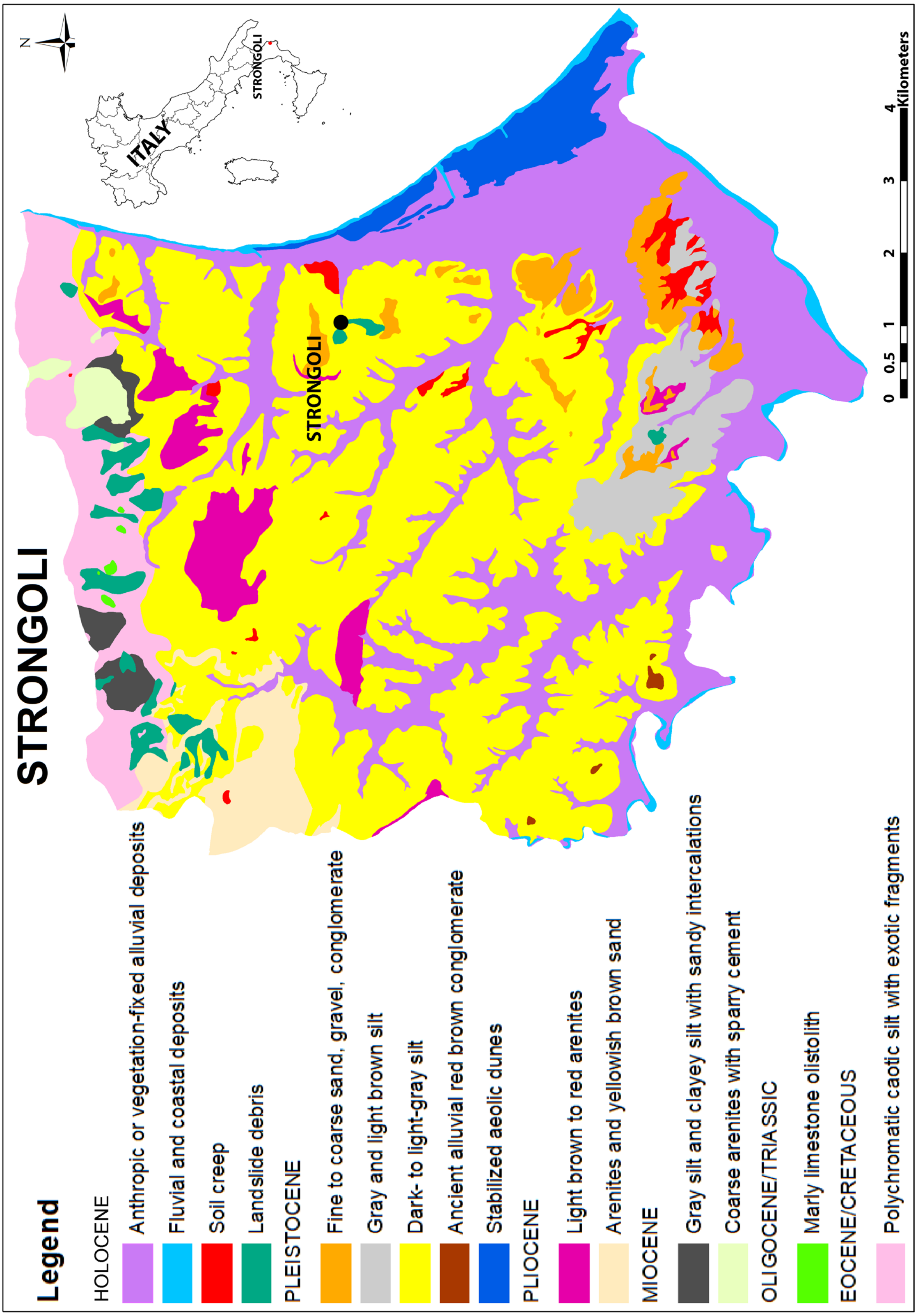

Fig. 2 - Geological maps of Strongoli (UTM: 676947.629, 4348633.024).

Fig. 3 - Sites of sampling and stations of measurement (SP1-UTM: 677331, 4348692; SP3-UTM: 677753, 4348421; CP1-UTM: 681329, 4334247; CP5-UTM: 683656, 4327931; CP7-UTM: 681526, 4330594; CP8-UTM: 681080, 4331119; SP6-UTM: 683224, 4328344).

error is probably greater as the images are characterized by a lower resolution.

The figure 4 shows the contouring of areal density distribution of cement-asbestos roofs in Crotone city. In appendix II is reported the numerical out-put with geographic coordinates and address of buildings with eternit roofs.

3.1.1. Sampling and Analytical protocol

Table 1 shows the UTM position, name and date, for the all site of sampling. The sampling of asbestos fibers was carried out by employing a low-flow sampling system (air flux: 8 L/min, Fig. 5) with 25 mm diameter mixed-cellulose ester filter papers (European and Italian Guideline: 83/477/CEE, LD 277/1991; DM 6/9/1994). The analytical protocol for asbestos fibers in air particulate considers electron microscopy (ESEM FEG QUANTA 200, FEI/Philips coupled to Energy Dispersive X-ray Spectroscopy micro-analysis system with Si/Li crystal detector EDAX GENESIS-4000), both for mineral identification and fibers quantification.

Regarding to instrumental conditions we adopted the protocol provided by DM 6/9/1994, in order to identify fibers having dimensions of at least 0.2 μm in diameter.

DM 6/91994 recommends to use an accelerating voltage (VA) of at least 20 keV (the law provides that it is a value between 20 and 30 keV), a tilt angle of 0, a working distance (WD) equal to 13.

In agreement with Gualtieri et al. (2009) we have counted:

- all the visible fibers regardless of their length and chemical composition, with an aspect ratio (length to diameter) of >3:1 (fibers);

-the fibers with a length greater than 5 μm, diameter smaller than 3 μm, and length to diameter ratio equal to or greater than 3:1, regardless of their chemical composition (regulated fibers);

- all the visible asbestos fibers regardless of their length identified on the basis of their chemical composition, with an aspect ratio (length to diameter) of >3:1 (asbestos fibers);

- the asbestos fibers with a length greater than 5 μm, diameter smaller than 3 μm, and length to diameter ratio equal to or greater than 3:1, identified on the basis of their chemical composition (regulated asbestos fibers);

- all the visible non-asbestos fibers regardless of their length identified on the basis of their chemical composition, with an aspect ratio (length to diameter) of >3:1 (fibers other than asbestos);

- the non-asbestos fibers with a length greater than 5 μm, diameter smaller than 3 μm, and length to diameter ratio equal to or greater than 3:1, identified on the basis of their chemical composition (regulated fibers other than asbestos).

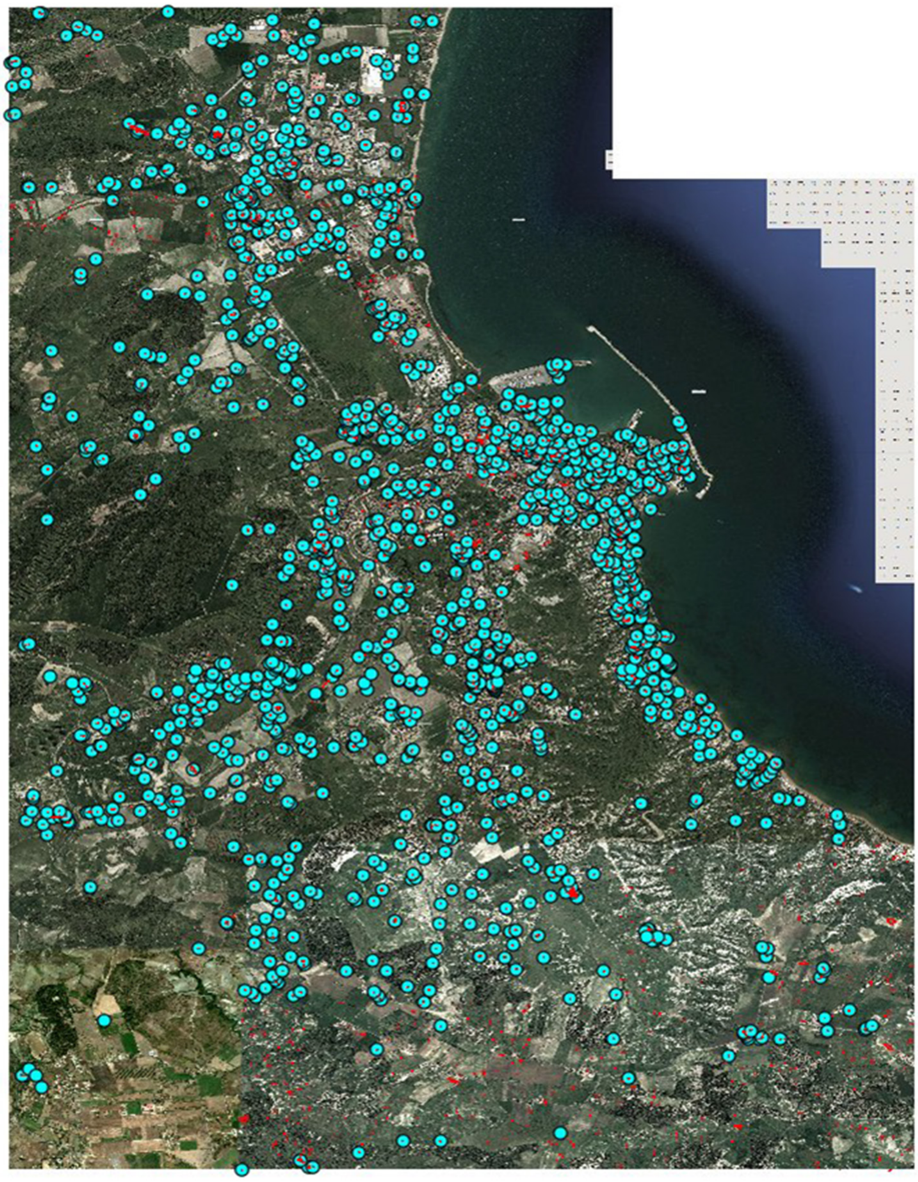

Fig. 4 - Eternit roofings in Crotone (UTM: 684088.402, 4328119.858) and Strongoli area (UTM: 676947.629, 4348633.024) (from DataGeo).

All fibers lying within the counting area (area of the field at 2.000X, as equivalent to the TV screen placed in the position; we read about 450 fields.) were counted as one fiber. We follow the M.D. 06/09/1994 both for the working conditions of the SEM and the counting of the fibers. Fibers riding the edges of the screen were counted as a half fiber. A split fiber was considered as a fiber. Fibers in a cluster were counted individually if they were sufficiently distinct, even at high magnification, as long as they complied with the dimensions shown in the definitions. The count of the total mineral fibers had been carried out over the entire area of the sampling filters (Tab. 2).

4. RESULTS AND DISCUSSIONS

For all investigated site, the type of asbestos phases identified are chrysotile (Fig. 6a) and amphibole asbestos (Fig. 6b). Most of the fibres from a morphological point of view integrated by the EDS chemical analyses they can be identified as chrysotile (Fig. 6a) while, Si vs. Mg/(Mg+ Fe2+) diagram (Hawthorne and Oberti, 2007) allowed to classify all the analysed amphibole asbestos as tremolite (Fig. 6b). In addition, mineral fibers of various nature other than amphibole and chrysotile fibers were discovered. Results of SEM analyses show that in order of decreasing abundance, mineral fibers are represented by: silicate glass, consisting of more than 65-70% SiO_2 (glass wool), organic materials, calcite, individual or aggregated into bundles (Fig. 6 c,d), gypsum and halite. The needle-fiber calcite likely derive from altered cement building (Miriello et al., 2013). In almost all samples we found ceramic fibers with rigid and compact shape and smooth surface.

They have been grouped into the “regulated fibers other than asbestos” (Tab. 2).

Sampling	Station of measurement	UTM (m) X	UTM (m) Y	Description
Crotone	CP1 (background)	681329	4334247	Villa Ermenegilda
	CP5	683656	4327931	Alfieri school
	CP7	681526	4330594	Consortium for Industrial Development
	CP8	681080	4331119	Regional Agency for Development and Services in Agriculture (ARSSA)
Strongoli	SP3	677753	4348421	School in Pianette
	SP6	683224	4328344	Lucifero school
	SP1 (background)	677331	4348692	Strongoli football field

Tab. 1 - Map reference points of the sampling stations.

Fig. 5 - Low-flow sampling system used for the campaign Strongoli 1 at CP1 (a) and SP3 (b) stations.

Within this group we included the "glass fibers", owing this name to their fibrous shape and to the fact that they are completely made up by SiO_2 (Fig. 6c).

The highest concentration of regulated fibers was found at the site of the Regional Services and Development in Agriculture (ARSSA), with 0.15 ff/l (CP8), while the lowest were discovered at the school "Alfieri" in Crotone: 0.01 ff/l (CP5) (Tab. 2). All the other sites have intermediate values between the two. Most of the samples contain asbestos fibres with concentration lying below 0.1 ff/l, which is the value proposed by Gualtieri et al. (2009) as the concentration limit for asbestos fibers in ambient living environments.

However, we exclude that asbestos fibers come from naturally occurring asbestos inasmuch NOA has no outcrops in the study area (Figs. 1, 2).

Anthropogenic pollution is more likely since people live close to sources of asbestos containing materials (ACMs) such eternit (Fig. 4). In fact, eternit (cement-asbestos) is one of the asbestos-based products widely spread in the study area. In the whole, the low number of fibers and asbestos suggest that the asbestos-cement roofing (eternit) are still in good condition and that, following the enactment of Law No. 257/1992 and DM

Sampling	Station of measurement	Code of Sample	Fibers	Regulated fibers	Asbestos fibers	Regulated asbestos fibers	Fibers other than asbestos	Regulated fibers other than asbestos	Total Regulated ff/L	Total asbestos ff/L
Crotone	CP1 (background)	CP1	358	250	1	1	326	218	0.0665	0.0003
	CP8	CP8	478	318	32	33	471	312	0.1593	0.0107
	CP7	CP7	160	110	1	1	159	109	0.0363	0.0003
	CP5	CP5	67	18	0	0	49	18	0.0117	0.0000
Strongoli	SP3	SP3	96	53	1	1	101	53	0.0331	0.0000
	SP1 (background)	SP1	124	41	4	2	120	39	0.0392	0.0017
	SP6	SP6	113	54	0	0	59	54	0.0392	0.0000

Tab. 2 - Fiber minerals count under SEM, following Gualtieri et al. (2009).

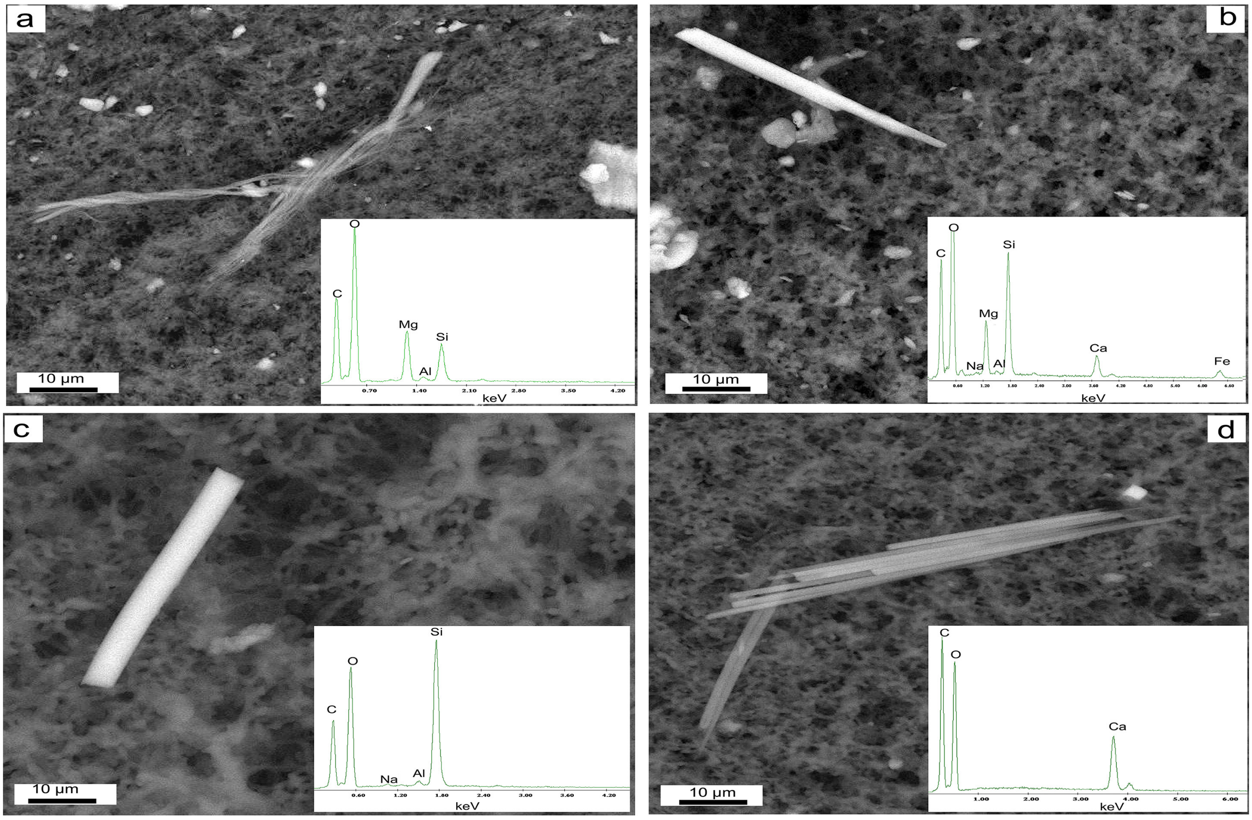

Fig. 6 - Mineral fibres of chrysotile (a), tremolite (b), silicatic glass (c), and calcite (d) with the relative energy-dispersive spectrometry (EDS) point analysis.

06/09/1994, the disposal of the same has had the desired effect. Furthermore, it was noted that among the samples of background level and those of polluted sites there are not substantial differences.

Therefore, the measured concentrations may be considered as a general level of asbestos in the atmospheric dust of Crotone (so-called "background level").

Anyway it must be considered that the small quantities of fibers are lower than the theoretical method detection limits of the SEM techniques for identification and counting of asbestos fibers (about 1 f/l for SEM see for example Steen et al., 1983). Moreover, it is worth remembering that the global scientific community also acknowledged the fact that there is no evidence of a threshold level of exposure to asbestos fibres below which there is no risk of mesothelioma (Chiappino et al., 1991; Ramazzini, 2010). In addition, samples from the Regional Development and Services in Agriculture (A23-CP8) shown the presence of lead fibers. Lead can be related to anthropogenic origin, like traffic, or associated to dispersion of lead-rich ceramic frit (glaze material) in the air. The latter origin is supported by the presence in the same samples of lead chromate ($PbCrO_4$) impregnated fiber-glass.

As a matter of fact, there are both epidemiological and experimental indications that trace elements such as Cr and Pb may provoke lung cancer (Nemery, 1990; IARC, 1993; Chen et al., 2003; Shekhawat et al., 2015) and some researchers have claimed that fibres morphology may play a passive role in producing diseases as carriers of trace elements (Bloise et al., 2016c).

The presence of glass fibers could be ascribed to fibers of quartz or cristobalite, which are respectively considered as a raw material or a high-temperature product of ceramic industry. In 1988 the International Agency for Research on Cancer classified fiberglass, rock wool, slag wool, and ceramic fibers as Group 2B, i.e. possibly carcinogenic to humans.

However, in 2002 it reassigned fiberglass, rock and slag wool, and continuous glass filaments to Group 3, not classifiable as carcinogenic to humans and this somehow leaves the issue open to debate.

5. CONCLUSIONS

This work shows concentration and mineralogical composition of the airborne fibers collected in the urban areas of Crotone and Strongoli. The monitoring of asbestos had been conducted from May to September 2012, by using a low-flow sampling system. All the sampled sites display a low number of regulated fibers such asbestos (chrysotile and tremolite).

Despite the high presence of asbestos roofings in the analyzed areas, the level of mineral fibers in air particulate is low than expected. The highest concentration of regulated fibers had been found at the site of the Regional Services and Development in Agriculture (ARSSA), with 0.1593 ff/l, while the lowest were discovered at the school "Alfieri" in Crotone: 0.0117 ff/l.

It seems unlikely environmental pollution as the NOA outcrops are many kilometers distant. Anthropic pollution due to roofs containing asbestos (eternit) is more likely and the measured concentrations can be considered as background level of asbestos in atmospheric dust in Crotone and Strongoli. Moreover, since there is convincing evidence of a relationship between lung cancer mortality and cumulative Cr and Pb exposure, the toxicity of fibrous minerals could be increased by harmful effect of these elements which have been found in more of the analyzed samples.

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