Naturally Occurring Asbestiform Minerals in Italian Western Alps and in Other Italian Sites

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Abstract

The natural occurrence of asbestos (NOA) in rocks and soil has been known for many years in several areas of the world, differently from the natural presence of asbestiform minerals. In Italy the mapping of NOA is mandatory according to the 2001 and 2003 regulations. The investigation, not yet concluded, has revealed that in Italy NOA are represented by chrysotile, tremolite asbestos with minor amount of actinolite asbestos and anthophyllite asbestos. A field survey carried out in Italian Western Alps (IWA), dealing with the naturally occurring of asbestiform minerals non-asbestos classified (NONA) and not regulated, started many years ago and is still ongoing. It revealed that the following kinds of asbestiform silicates are present (in decreasing order of frequency): asbestiform polygonal serpentine and asbestiform antigorite, asbestiform diopside, asbestiform carlosturanite, asbestiform

forsterite, asbestiform sepiolite, asbestiform balangeroite, and asbestiform talc. The asbestiform non-silicates brugnatellite and brucite have been rarely detected. Outside the IWA, asbestiform zeolite (erionite and offretite), asbestiform sodium amphibole (fluoro-edenite), and few other asbestiform silicates have been also detected. For some asbestiform minerals, the identification is problematic and needs the use of transmission electron microscope (TEM) combining either the imaging at high magnification as the electron diffraction and the chemical data. This investigation is particularly important to distinguish four kinds of asbestiform minerals (antigorite, polygonal serpentine, carlosturanite, and balangeroite) from chrysotile since only the last one is regulated. The issue is much more complicated by the intergrowth of different fibrous species on the scale of sub micrometre.

Keywords: NOA; naturally occurring non-asbestos classified asbestiform minerals (NONA); TEM-EDS identifications; fibre intergrowth

Introduction

In recent years and in several countries the health investigations dealing with asbestos moved from the occupational to environmental exposure (e.g., Baumann et al., 2015; Abakay et al., 2016; Noonan, 2017). According some authors in fact the exposure to low but continuous exposure, as in the case of inhabitants of houses next to the asbestos bearing rock outcrops (i.e. naturally occurring asbestos: NOA), could cause health problems (e.g., Luce et al., 2000; Bernardini et al., 2003). The same problem could concern the exposure to naturally occurring non-asbestos classified asbestiform minerals (NONA). For some of them, the carcinogenicity to high dose exposure is known as in the case of asbestiform fluoeredenite, asbestiform erionite, asbestiform winchite, and asbestiform richterite (e.g., Burragato et al., 2005, IARC, 2012). In addition to these last minerals, many others having an asbestiform morphology have been discovered over the years in different part of the world. The most

striking current example is represented by asbestiform antigorite. For many years this antigorite asbestiform variety has not been recognized as having a full identity and little information has appeared in few publications (e.g., Keeling et al., 2006).

But, exactly as it happens for the NOA, also the NONA can be dispersed in air both for natural causes (weathering, natural atmospheric agents, landslide) and for anthropogenic causes (e.g. excavation works).

If these asbestiform minerals are noxious following continuous low dose and/or sporadic high doses, it will be known only in many years, as it has happened for example for asbestos given the long latency time of related asbestos pathologies. For the instance, it would be a good practice to map the presence of these minerals in every country.

As it concerns Italy, the mapping of NOA is mandatory according to the March 23, 2001 Law, n. 93 and to the related March 18, 2003, Environment Ministry Decree, n. 101. The mapping of NOA must be carried out by each single Region through its own Environmental Protection Agency, using literature data, geological maps, historical information on research permits and mining concessions, reports and environmental monitoring carried out as part of environmental and strategic impact assessment procedures for the construction of infrastructural works, analytical certificates, mining activities of lithotypes suspected for the presence of asbestos, surveys for the Geological Cartography project at 1: 50,000 scale, specific investigations and surveys, possible inspections and sometimes collection of samples and laboratory analyses. Each year the individual EPA send any possible additional data to the Ministry of Health that integrates it to the aim of the evaluation of the necessary remediation works.

Currently, the map of the Italian NOA is published from the Ministry of the Environment and Protection of the Territory and the Sea; it is updated to the year 2018 and reports NOA sites only from three regions (Asbestos Mapping, 2018) but to the current state of scientific knowledge, NOA are

present in at least 7 other regions. This paper presents the different NOA and NONA until now detected in Italy.

Detected NOA and NONA

Firstly, owing to the different used definitions, it needs to define the meaning of the used words. According to the dimensional definition of the World Health Organization (WHO, 1997), many regulatory agencies and the literature (e.g. Belluso et al., 2017), in this note we use the definition listed below.

Fiber: inorganic particle with length ≥ 5 μm, width ≤ 3 μm, length/width (aspect ratio) $\geq 3:1$, parallel sides when seen in two dimensions, perpendicularly to fiber axis.

Fibril: a single mineral fiber which cannot be further separated longitudinally into smaller components (without losing the fibrous properties or appearances).

Asbestiform: adjective for fibers non asbestos classified having the "fiber" dimensions and at least one of the asbestos properties as flexibility, splitting etc.

Fiber bundle: parallel aggregate of mineral fibers.

In the scientific literature many researches touch the NOA topic. Some of them have been carried out specifically for mineralogical interests, others concern petrological, structural engineering or geology studies; sometimes the scientific investigations arise from minerals collectors. In any case, their identification is carried out by using different techniques, in some cases by using two or more than two in a complementary way. The most diffused identification techniques are X-ray powder diffractometry (XRPD), infrared spectroscopy (with Fourier transform), optical (OM) and electron microscopy (EM), usually scanning (SEM) and uncommonly transmission (TEM), coupled with energy dispersive spectrometry (EDS).

Based on scientific literature data, NOA are widespread in many areas of Italy and represented by chrysotile and tremolite asbestos (both very diffused), the less diffused actinolite asbestos and finally anthophyllite asbestos that is the much less diffused than others (e.g., Cavallo and Rimoldi, 2013; Gaggero et al., 2013; Vignaroli et al., 2013; Gaggero et al., 2017). The Figure 1 shows the 10 Regions where NOA are present in rocks, at the current state of knowledge. Crocidolite and amosite do not constitute NOA in Italy.

Unlike NOA, the mapping of NONA is not mandatory in Italy. Several spot investigations, carried out during both the asbestos investigation and specific mineralogical researches, show that NONA are present in many Italian areas (8 Italian Regions) and abundant in some places. Except Piedmont and Aosta Valley Regions which we will discuss later, the NONA identified in Italy are the following (in alphabetic order): asbestiform antigorite, asbestiform Ca-erionite (with Ca-levyne), asbestiform F-edenite, asbestiform gedrite, asbestiform Mg-horneblende, asbestiform offretite, asbestiform polygonal serpentine, asbestiform sepiolite (e.g., Cattaneo et al., 2011; Bloise et al., 2014;.Bloise et al., 2016; Bloise et al., 2017; Giordani et al., 2017; Lucci et al., 2018; Mattioli et al., 2018) The Italian Regions where these asbestiform minerals have been detected are shown in Figure 2.

Regarding the Italian Western Alps (IWA) Regions, i.e. Piedmont and Aosta Valley, a field survey dealing with NOA is carried out since 1980 and it is still ongoing (Baronnet and Belluso, 2002; Belluso et al., 2015; Leone, 2018; Paccagnella, 2018). More than 300 samples have been collected and analyzed by using XRPD coupled with SEM-EDS and/or TEM-EDS. As NOA, chrysotile, asbestos tremolite, and, in lesser amount, asbestos actinolite have been detected.

The investigation in this area revealed that not only NOA are present, but that there are many kinds of asbestiform silicates (sometimes in very high amount) of NONA, not regulated in Italy. Ten are the identified NONA and eight of these are magnesium-containing silicates (Table 1). The complete list is shown below, in decreasing order of frequency: asbestiform polygonal serpentine and

asbestiform antigorite (with very high frequency); asbestiform diopside, asbestiform carlosturanite, asbestiform forsterite, asbestiform sepiolite, asbestiform balangeroite, and asbestiform talc. Rare are asbestiform brugnatellite and asbestiform brucite, the only two non-silicates asbestiform minerals.

As far as quantities are concerned, asbestiform carlosturanite and asbestiform balangeroite are very abundant. As regards the quantities of asbestiform antigorite and asbestiform polygonal serpentine, is not possible to define the amount because these two minerals are always intergrown with others fibrous minerals, asbestos and non asbestos classified.

For some asbestiform minerals, the certain identification is not so easy because they show similar (at times even equal) characteristics on a macroscopic scale. As it concerns NOA and NONA in IWA Regions, on the macroscopic scale in most cases it is not possible distinguish among chrysotile, asbestiform antigorite, asbestiform polygonal serpentine, asbestiform carlosturanite, and asbestiform balangeroite. Figure 3 shows an example of this similarity on the eye scale, both in outcropping rocks and after picking fibers from the rock, between asbestiform antigorite (Figs. 3a and 3b) and chrysotile (Fig. 3c). The bundles of fibres are flexible and their characteristics are very similar to each other. Also the colour is similar because chrysotile containing a little amount of iron has not white colour but cream to light brown.

Often it needs the use of the transmission electron microscope (TEM) combining either the imaging at high magnification as the selected area electron diffraction (SAED) and the chemical data (EDS).

This investigation is particularly important to distinguish four kinds of NONA and specifically asbestiform antigorite (Fig. 4), asbestiform polygonal serpentine (Fig. 4), asbestiform carlosturanite, and asbestiform balangeroite, from chrysotile (Fig. 4) since only the last one is regulated, being asbestos classified. The issue is very complicated because usually, on the scale of little thousands or

even hundreds micrometers, the fibers of two to four mineral species are intergrown (owing to solid state transformations) as it can see in Fig. 4.

Based on the detailed investigation by TEM-EDS, practically each bundle of fibers turns out to be made up of at least 2 fibrous mineral species, except in the case of asbestiform sepiolite, which on the other hand appears to have only intergrown with an organic phase (aliphatic hydrocarbons: Giustetto et al., 2014).

Conclusions

In Italy NOA and NONA are diffused in several areas. NOA in Italy are represented only by four mineral species; among them only chrysotile and tremolite asbestos are abundant and diffused whereas actinolite asbestos is less diffused and anthophyllite asbestos is rare. NONA in Italy are constituted by 15 mineral species, many more than NOA, although they have not the same diffusion and abundance.

Detailed investigation on NOA and NONA, even if not completed, were conducted only in Piedmont and in Aosta Valley Regions and lead to reveal many different NONA in rocks and in the same bundle of fibers. Owing to the intergrowth on the sub-micrometric scale of different fibrous species, many times only the TEM-EDS investigation allows to detect all the mineral species and for certain phases only this kind of techniques allows to obtain a certain identification, as for example to distinguish among asbestiform antigorite, asbestiform polygonal serpentine, and chrysotile, therefore from non-asbestos and asbestos classified minerals.

Finally, many issues arise from the shown data as the relevance of the correct and representative sampling, the importance of the use of the technique suitable to identify the content of these composite materials (i.e. the bundle of fibers), the need of the extended surveys for extensive investigations where there are rocks that may contain mineral fibers.

172 The investigation by TEM-EDS of fiber bundles collected around the world may give surprises
173 like those from Italian Western Alps rocks.
174

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TABLE CAPTIONS

Table 1. List of NONA in decreasing order of finding frequency, detected in Piedmont and Aosta Valley Regions, North-Western Alps, Italy and the ideal chemical- formula.

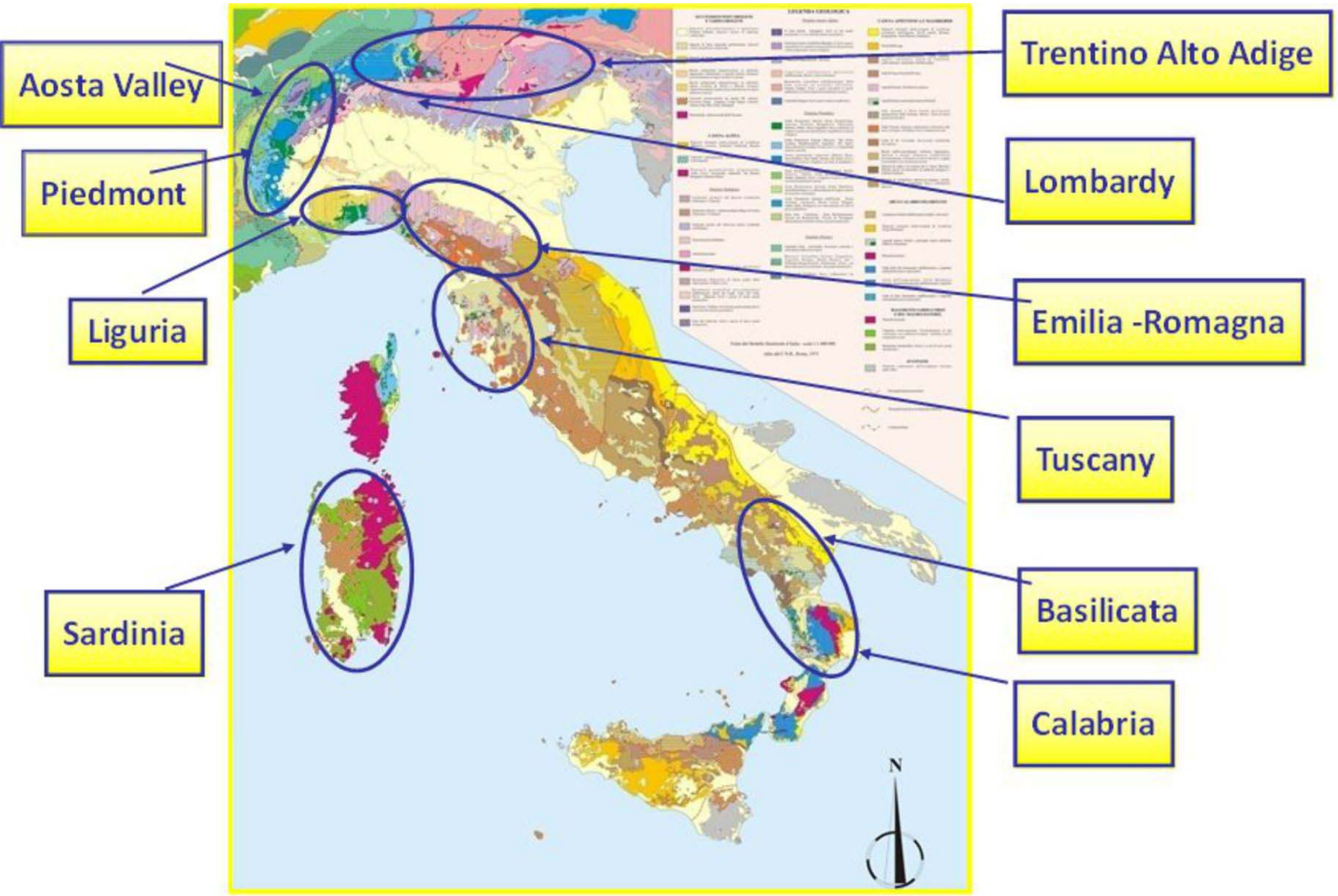
Table 2. Parallel intergrowths of asbestos and asbestiform minerals after investigations by TEM-EDS. The most abundant asbestiform minerals (main) and the intergrown minerals (subordinate) in the same bundle are listed in the left and in the right column, respectively. (asbestos are indicated in italic- Italian legislation 277/91).

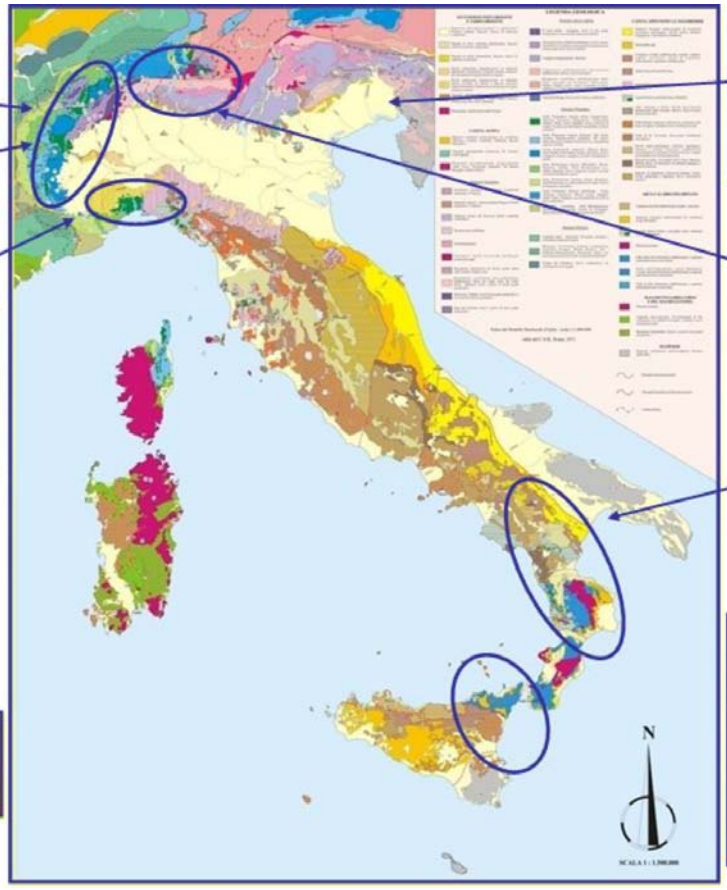
FIGURE CAPTIONS

Figure 1. Italian regions and mineral species of naturally occurring asbestos (NOA) in rocks: chrysotile, tremolite asbestos, less diffused actinolite asbestos, and rare anthophyllite asbestos.

Figure 2. Italian regions and mineral species of naturally occurring non-asbestos classified asbestiform minerals (NONA) in rocks: asbestiform antigorite, asbestiform Ca-erionite with Ca-levyne, asbestiform F-edenite, asbestiform gedrite, asbestiform Mg-horneblende, asbestiform offretite, asbestiform polygonal serpentine, asbestiform sepiolite. For Aosta Valley and Piedmont Regione, details are in the text.**Figure 3.** Macroscopic similarity of mineral fibre bundle. A) Vein filled of asbestiform antigorite. B) Fibrous bundles of asbestiform antigorite. C) Fibrous bundles of chrysotile.

Figure 4. TEM image of cross-section of an asbestiform composite made of axially-textured intergrown fibrils of asbestiform antigorite (atg) with minor asbestiform polygonal serpentine (PS) and chrysotile (chrys).


Aosta Valley
Piedmont
Liguria
Sardinia
Trentino Alto Adige
Lombardy
Emilia -Romagna
Tuscany
Basilicata
Calabria
N



Aosta Valley

Piedmont

Liguria
asbestiform sepiolite
asbestiform antigorite

Sicily
asbestiform F-edenite

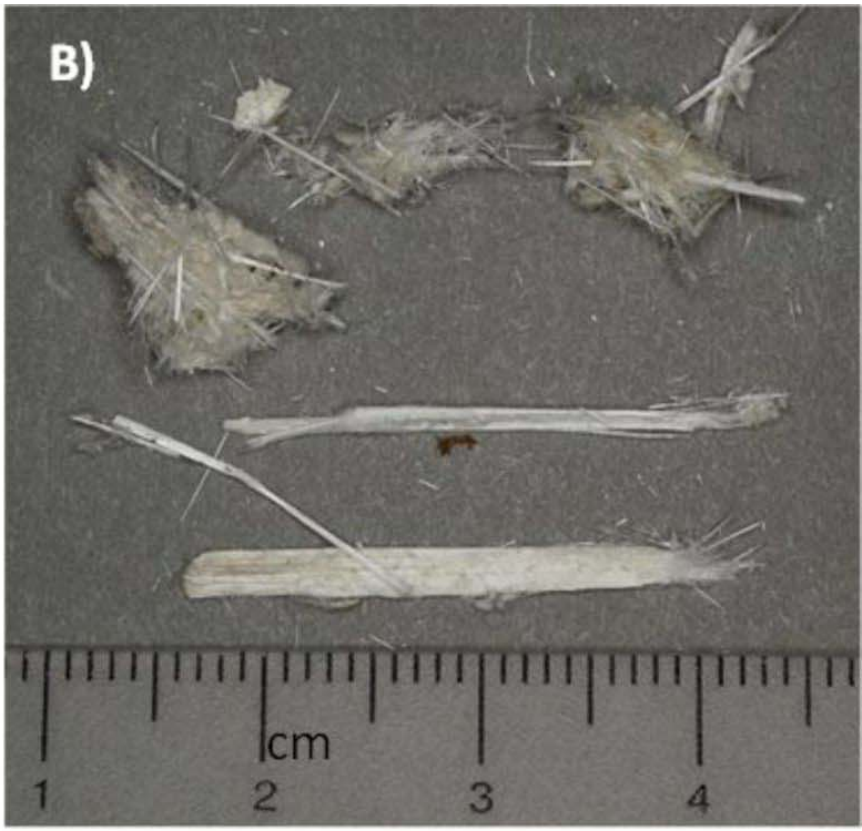
Veneto
asbestiform offretite
asbestiform Ca-erionite + Ca-levyne

Lombardy
asbestiform antigorite

Basilicata
asbestiform polygonal serpentine

Calabria
asbestiform antigorite
asbestiform polygonal serpentine
asbestiform gedrite
asbestiform Mg-horneblende


A)


B)
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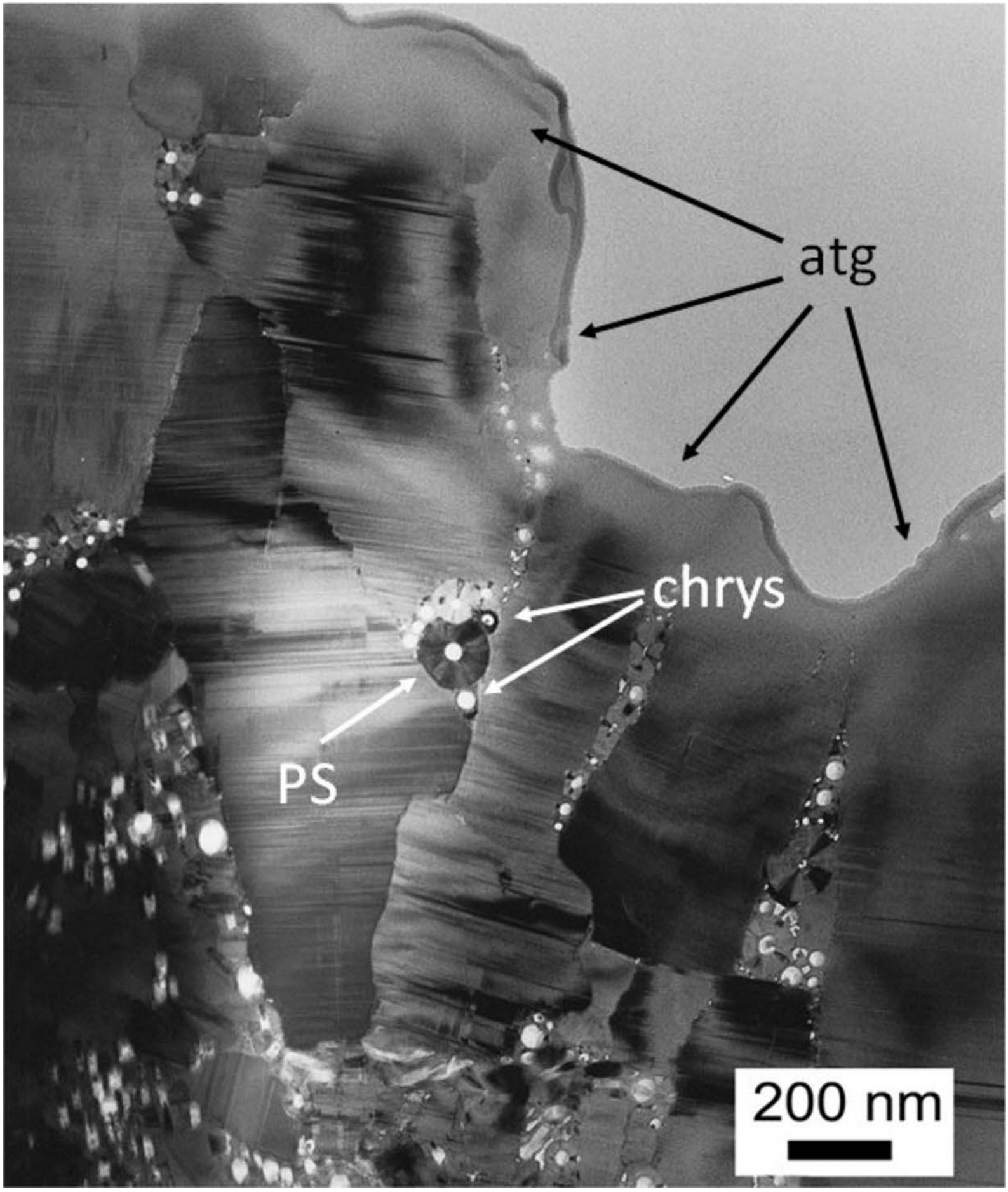
atg
chrys
PS
200 nm

NONA	Ideal chemical formula
asbestiform polygonal serpentine, asbestiform antigorite	$Mg_3Si_2O_5(OH)_4$
asbestiform diopside	$CaMgSi_2O_6$
asbestiform carlosturanite	$(Mg,Fe,Ti)_{21}[Si_{12}O_{28}(OH)_4](OH)_{30}\bullet H_2O$
asbestiform forsterite	Mg_2SiO_4
asbestiform balangeroite	$(Mg,Fe,Mn)_{21}O_3(OH)_{20}(Si_4O_{12})_2$
asbestiform sepiolite	$Mg_4Si_6O_{15}(OH)_2\bullet 4H_2O$
asbestiform talc	$Mg_3Si_4O_{10}(OH)_2$
asbestiform brugnatellite	$Mg_6Fe(CO_3)(OH)_{13}\bullet 4H_2O$
asbestiform brucite	$Mg(OH)_2$

Table 1. List of NONA in decreasing order of finding frequency, detected in Piedmont and Aosta Valley Regions, North-Western Alps, Italy and the ideal chemical- formula.

MAIN MINERAL	SUBORDINATE MINERALS
chrysotile + asbestiform polygonal serpentine	asbestiform antigorite, asbestiform diopside, asbestiform carlosturanite, asbestiform forsterite, asbestiform balangeroite
asbestiform antigorite	*chrysotile* + asbestiform polygonal serpentine, *asbestos tremolite*, asbestiform carlosturanite
asbestos tremolite (asbestos actinolite)	*chrysotile* + asbestiform polygonal serpentine, asbestiform antigorite, asbestiform talc
asbestiform diopside	*chrysotile* + asbestiform polygonal serpentine, asbestiform antigorite, asbestiform carlosturanite, asbestiform balangeroite, asbestiform brugnatellite
asbestiform carlosturanite	*chrysotile* + asbestiform polygonal serpentine, asbestiform diopside, asbestiform antigorite, asbestiform forsterite, asbestiform brucite
asbestiform forsterite	*chrysotile* + asbestiform polygonal serpentine, asbestiform carlosturanite
asbestiform balangeroite	*chrysotile* + asbestiform polygonal serpentine, asbestiform diopside
asbestiform brugnatellite	asbestiform diopside
asbestiform brucite	asbestiform diopside
asbestiform sepiolite	"organic matter"

Table 2. Parallel intergrowths of asbestos and asbestiform minerals after investigations by TEM-EDS. The most abundant asbestiform minerals (main) and the intergrown minerals (subordinate) in the same bundle are listed in the left and in the right column, respectively. (asbestos are indicated in italic- Italian legislation 277/91).