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(Article begins on next page)

Mobility of crocidolite asbestos in sandy porous media mimicking aquifer systems

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Abstract

Asbestos is widely recognized as being a carcinogen when dispersed in air, but very little is known about its exposure pathways in water and its subsequent effects on human health. Several studies have proved asbestos presence in groundwater but failed to assess its mobility in aquifer systems. This paper aims to fill this gap by studying the transport of crocidolite, an amphibole asbestos, through sandy porous media mimicking different aquifer systems. To this purpose, two sets of column test were performed varying the crocidolite suspension concentration, the quartz sand grain size distribution, and the physicochemical water parameters (i.e., pH). The results proved that crocidolite is mobile in quartz sand due to the repulsive interactions between fibres and porous media. The concentration of fibres at the outlet of the column were found to decrease when decreasing the grain size distribution of the porous medium, with a bigger impact on highly concentrated suspensions. In particular, 5-to-10- μm -long fibres were able to flow through all the tested sands while fibres longer than 10 μm were mobile only through the coarser medium. These results confirm that groundwater migration should be considered a potential exposure pathway while implementing human health risk assessment.

Environmental implication

Asbestos is widely recognized as a human carcinogen when inhaled, but its adverse effects in water are still being disputed. Since waterborne asbestos can be ingested or inhaled due to nebulization or vaporization, a precise evaluation of its mobility in the aquifer system - one of the largest sources of freshwater for human consumption - is essential to perform an accurate human health risk assessment and estimate an exposure concentration level. Our laboratory study has confirmed crocidolite (an amphibole asbestos) mobility and transport in porous media, confirming that groundwater is a potential asbestos migration pathway.

Keywords

Asbestos transport, Fibre mobility, Crocidolite, Groundwater, Column test

41 1 Introduction

42 Asbestos is the commercial term that indicates a group of six naturally occurring silicate
43 minerals with fibrous morphology. This group includes one serpentine phyllosilicate
44 (chrysotile) and five amphiboles, which are chain silicates (tremolite asbestos, actinolite
45 asbestos, anthophyllite asbestos, amosite, and crocidolite). In the past, these minerals have
46 been widely employed for industrial applications due to their valuable technological properties,
47 such as resistance to heat, fire, chemical and biological degradation. Nowadays their
48 application has been drastically decreased, or even banned by several countries (IBAS, 2023),
49 due to their adverse effects on human health. Indeed, the International Agency for Research
50 on Cancer classifies asbestos as carcinogenic of the first group (IARC, 2012) and it is widely
51 recognized as an air pollutant. For this reason, the European Union (EU) established an air
52 concentration limit in the workplace for respirable fibres, i.e. fibres with length $> 5 \mu\text{m}$, width $<$
53 $3 \mu\text{m}$ and aspect ratio (AR, length to width) > 3 (WHO, 1986), which are the ones considered
54 to have carcinogenic effects when inhaled. The limit is 100 fibres per litre (f/L) (Directive
55 2009/148/EC) and is applied to the average air concentration measured during an 8 hour work
56 shift. The same limit is also applied in the United States by the U.S. Occupational Safety and
57 Health Administration (OSHA, 2021).

58 In the last decades, the attention of researchers and regulators shifted from occupational to
59 environmental exposure scenarios. In particular, asbestos can be released by the erosion of
60 Naturally Occurring Asbestos (NOA) or by mining and industrial activities (e.g. Mensi et al.,
61 2015; Reid et al., 2007). An outdoor attention threshold of 1 f/L for ambient air was therefore
62 proposed by the EU in the air quality guidelines to regulate asbestos exposure in non-
63 occupational environments (WHO, 2000).

64 Up until recently, legislations on asbestos have however only focused on regulating its
65 presence in air, neglecting to consider migration pathways and subsequent human exposure
66 via water. This is partially due to the fact that the effects of waterborne asbestos ingestion on
67 human health are still unclear and have only recently begun to be studied by the scientific
68 community (Malinconico et al., 2022; Di Ciaula, 2017; Fortunato and Rushton, 2015). (WHO,
69 2021) The current literature identifies two main human exposure pathways for waterborne
70 asbestos: (i) direct ingestion of asbestos containing water or beverages (Cunningham and
71 Pontefract, 1971); (ii) inhalation of nebulised contaminated water droplets (e.g. Avataneo et
72 al., 2022; Roccaro and Vagliasindi, 2018) or of resuspended fibres after polluted water
73 vaporisation/evaporation. For the first scenario, the U.S. Environmental Protection Agency
74 (US-EPA) established a maximum contaminant level in drinking water of $7 \cdot 10^6$ f/L for fibres
75 longer than $10 \mu\text{m}$ (US-EPA, 2021) as a precautionary measure based on in vivo studies
76 (NTP, 1985). Instead, the legislation regarding airborne asbestos and its limits could be
77 applied to the second scenario, but it has been traditionally neglected, even if it can determine
78 a significant inhalation exposure.

79 Besides drinking water, also freshwater resources should be monitored, in particular
80 groundwater, which is one of the main sources of water for human consumption and
81 anthropogenic activities. Indeed, groundwater is at the basis of many agricultural and industrial
82 activities, as well as drinking water supply plants (e.g. Koumantakis et al., 2009). The presence
83 of asbestos in groundwater has been documented by several studies, which found fibres in
84 aquifers that are naturally rich in asbestos (Avataneo et al., 2021; Wei et al., 2013; Hayward,
85 1984; Oliver and Murr, 1977) or in areas where the mobilization of the fibres is further
86 enhanced by human activities, e.g. in the proximity of mines and mine tailing deposits (Turci
87 et al., 2016; Kashansky and Slyshkina, 2002; Buzio et al., 2000). More specifically, three
88 studies have investigated asbestos occurrence in groundwater close to the former chrysotile

89 mine of Balangero, Italy (Avataneo et al., 2021; Turci et al., 2016; Buzio et al., 2000). Based
90 on analyses by Scanning Electron Microscopy coupled with Energy Dispersive Spectroscopy
91 (SEM-EDS), Buzio et al. (2000) detected an asbestos content of 1.00 ± 4.10 mg/L in
92 groundwater and reported that the presence of fibres was reduced by a factor 10 at a distance
93 of 5 km from the mine discharge. Similarly, Turci et al. (2016) monitored two wells in proximity
94 of the former mine and detected over 10^6 f/L in the sampling point close to the mine southern
95 tailings. Avataneo et al. (2021) instead reported a very variable asbestos content in wells or
96 piezometers located in the alluvial plain close to the former asbestos mine of Balangero.
97 Based on Transmission Electron Microscopy (TEM)-EDS, fibres up to $13 \mu\text{m}$ long were found
98 in a sampling point with a concentration of $6.7 \cdot 10^6$ f/L (corresponding to $2 \mu\text{g/L}$). Conversely,
99 a study conducted on three drainage pits located next to a Russian asbestos deposit showed
100 a very low asbestos concentration detected by means of Phase Contrast Optical Microscopy.
101 The maximum concentration was $0.99 \cdot 10^5$ f/L with fibres longer than $5 \mu\text{m}$ ranging between
102 9.82% to 44.58% of the total (Kashansky and Slyshkina, 2002). Studies conducted in the U.S.
103 in the '70 and '80 on samples collected from wells and springs and analysed by TEM-EDS
104 showed the occurrence of asbestos in groundwater, mainly caused by the leaching of
105 asbestos from the host rock formations. In particular, asbestos concentrations in the range
106 $2 \cdot 10^7$ - $2 \cdot 10^8$ f/L were found in California (Hayward, 1984), while values over $2 \cdot 10^9$ f/L (0.91
107 $\mu\text{g/L}$) were found along the Rio Grande Valley, New Mexico (Oliver and Murr, 1977). In the
108 Dayao region, China, a thin layer of outcrop crocidolite ore is spread on an area of around 200
109 km^2 , resulting in an average groundwater asbestos contamination of $8.6 \cdot 10^6$ f/L, detected by
110 SEM-EDS analyses (Wei et al., 2013). (e.g., Hu and Hubble, 2007; Al-Adeeb and Matti, 1984)

111

112

113 The above-mentioned studies only focused on the presence of asbestos in groundwater,
114 failing to consider the mobility and transport of fibres through the aquifer systems. Indeed, the
115 mobility of asbestos in the subsurface has always been neglected or considered irrelevant in
116 the literature (INAIL, 2022; Wallis et al., 2020; Paglietti et al., 2012). The contamination
117 scenarios mentioned in previous studies can therefore imply relevant environmental and
118 sanitary issues, especially if asbestos is able to migrate through porous media such as aquifer
119 systems. An accurate study of asbestos mobility and transport is therefore needed to
120 implement and integrate the knowledge of previous work to provide a comprehensive
121 assessment of the health risks connected to migration via groundwater.

122 The well-established knowledge on particle transport in aquifer systems can be adapted to the
123 case of asbestos. The colloidal transport in porous media is governed by advection, dispersion
124 and by physical and physicochemical interactions with solid phase. Physical factors include
125 the size and shape of the particles (Ting et al., 2021; Seymour et al., 2013; Pelley and Tufenkji,
126 2008), the characteristics of the porous medium and the flow conditions (Bradford et al., 2002).
127 Thanks to their typical form factor (length \gg width), fibres can flow through pores that are
128 smaller than their length but bigger than their width. On the other side, the elongation of the
129 particles associated with drag forces can lead to filtration due to mechanical and geometrical
130 interactions with the porous medium (Seymour et al., 2013). These interactions are further
131 increased by aggregation and tangling (Chequer et al., 2019; Wu et al., 2017). Water
132 chemistry, especially ionic strength and pH, can strongly influence the transport of particles
133 by modifying the intensity of the attractive/repulsive forces acting between the fibres and the
134 porous medium (Pulido-Reyes et al., 2022; Beryani et al., 2020; Tian et al., 2012; Gronow,
135 1986).

136 The transport of particles is also affected by the electrostatic interactions between the particles
137 and the porous media. Chrysotile, at neutral pH, is retained within the aquifer system due to
138 the attractive electrostatic interaction between the positive surface charge of the fibres
139 (Pollastri et al., 2014) and the net negative surface charge of the soil (Granetto et al., 2022).
140 A recent study confirmed that bare chrysotile is not mobile in porous media but that, in
141 presence of dissolved organic matter (DOM), its surface charge can be reverted thus allowing
142 the subsurface transport (Mohanty et al., 2021). As opposed to serpentine phyllosilicate,
143 amphiboles naturally exhibit a negative net surface charge in water at any pH (Pollastri et al.,
144 2014). For this reason, these minerals are expected to be mobile in subsurface environments,
145 due to the repulsive electrostatic interaction between the fibre and the negative surface of the
146 porous medium.

147 The goal of this study is to verify if amphibole fibres, i.e. a naturally negatively charged
148 asbestos, can be transported through saturated sandy aquifer systems without any surface
149 modifier (or DOM). To this purpose, laboratory transport tests in sand-packed columns were
150 performed using crocidolite as a representative amphibole asbestos. The tests were
151 performed by varying the crocidolite concentration, the sand grain size distribution and the
152 porewater composition with the aim to investigate: (i) to what extent crocidolite can be mobile
153 in sandy porous media; (ii) how the size of fibres and pores affect the crocidolite mobility; (iii)
154 if the groundwater physicochemical parameters influence the fibre transport.

155 2 Materials and methods

156 2.1 Asbestos suspensions

157 An asbestos suspension was prepared by adding crocidolite UICC (Union for International
158 Cancer Control), a well characterised standard (Kohyama et al., 1996), to deionized water to
159 obtain a 300 mg/L concentration. The crocidolite was dispersed in water applying sonication
160 and stirring for about 25 minutes. The crocidolite suspension was characterised by a pH of 7.7
161 and a zeta potential of -30.2 ± 0.4 mV in 0.01 M NaCl, measured by dynamic light scattering
162 (DLS Zetasizer Nano Z, Malvern Instruments Ltd., U.K.). A 1 mM MOPS buffer (MOPS, 3-N-
163 Morpholino propanesulfonic acid, Sigma-Aldrich) was added to stabilise the pH at 6-7.

164 Two batches were prepared:

- 165 • a high concentration suspension (hereafter referred to as HC) obtained by collecting
166 the supernatant of the 300 mg/L crocidolite suspension after 5 hours settling;
- 167 • a low concentration suspension (hereafter referred to as LC) obtained by filtering the
168 HC suspension through a coarse sand bed.

169 The batches were used to simulate two different scenarios: the asbestos transport near a
170 concentrated contamination source using the HC batch, and the fibre fate at medium to long
171 distance from the source thus using LC batch.

172 2.2 Porous media

173 Transport experiments were conducted in quartz sand (Dorsilit from Dorfner GmbH & Co.,
174 Germany). Three types of sand with different grain size distributions (Figure S1,
175 Supplementary Information-SI), defined respectively coarse, medium, and fine, were tested.
176 The measured values of d_{10} - d_{50} - d_{90} were 1.31-1.59-1.91 mm for coarse sand, 0.48-0.68-0.84
177 mm for medium sand and 0.26-0.49-0.58 mm for fine sand. Before use, the sand was
178 thoroughly cleaned to remove any residual impurities and colloids. The cleaning procedure
179 consisted of three cycles of washing and sonication with 100 mM NaOH, tap water and
180 deionized water, respectively. After cleaning, the sand was dried with a laboratory hot plate.

181 Prior to packing the columns, a known quantity of dry sand was rehydrated in a 1 mM MOPS
182 solution and degassed with a vacuum bell.

183 The zeta potential of each porous medium was measured in 0.01 M NaCl solution by DLS
184 (Zetasizer Nano ZS90, Malvern Instruments Ltd., U.K.) and was equal to -46.8 ± 1.0 mV, -
185 49.1 ± 2.6 mV and -52.5 ± 0.6 mV, respectively for coarse, medium and fine sand.

186 The hydrodynamic parameters of the porous media, namely effective porosity and dispersivity,
187 were determined by fitting a breakthrough curve (BC) of a conservative tracer (i.e. 10 mM
188 NaCl in 1 mM MOPS). The estimated effective porosity was 47%, 43% and 39% for coarse,
189 medium and fine sand, respectively. While the dispersivity was 0.47 mm for the coarse sand,
190 0.20 mm for the medium sand and 0.10 mm for the fine sand. The BCs and the parameters
191 are reported in Figure S2 and Table S1 of SI, respectively.

192 2.3 Column test setup, sampling, and injection procedure

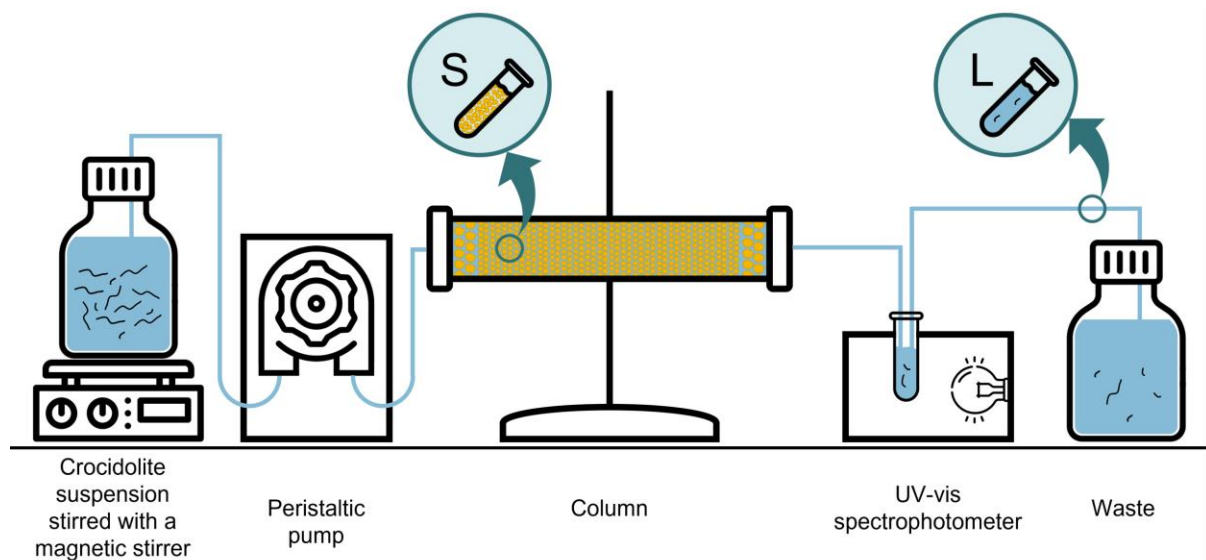
193 The experimental setup is outlined in Fig. 1. A column with internal diameter of 1.6 cm and
194 total length of 11 cm was wet packed as follows: coarse sand was used to create a 0.5 cm
195 drain at the column inlet and outlet; coarse, medium or fine sand, depending on the specific
196 experiment conditions, were used to pack the 10 cm porous medium to be tested.

197 The crocidolite suspension, continuously stirred, was pumped into the saturated column with
198 a peristaltic pump at a flow rate of 0.22 ± 0.01 mL/min, corresponding to a Darcy velocity of
199 1.56 ± 0.09 m/d. The injection schedule was applied for all the tests as described below.

- 200 (i) Preconditioning: 10 pore volumes (PV) of a 1 mM MOPS solution to equilibrate the sand
201 column.
- 202 (ii) Injection: 6 PV of crocidolite suspension (HC or LC) to test the asbestos transport.
- 203 (iii) Flushing: 3 PV of a 1 mM MOPS solution to flush the system.
- 204 (iv) Release: 3 PV of a solution containing 1 mM MOPS and 100 mM NaOH (pH 13) to induce
205 the detachment of the fraction of fibres that were reversibly deposited on the porous media
206 during phase (ii).

207 During the experiments, the concentration of crocidolite suspension at the column outlet was
208 measured online, via optical density measurements using a UV-vis spectrophotometer
209 (Specord S600, Analytik Jena, Germany) equipped with a flow-through cell (Hellma,
210 Germany). A linear relation between absorbance and crocidolite concentration was found at a
211 wavelength of 285 nm (Figure S3-S4-S5, SI). Throughout the paper “C” will be used to refer
212 to the absorbance of crocidolite concentration measured by the spectrophotometer for outlet
213 liquid samples, whereas “C₀” to refer to the absorbance concentration at the inlet.

214 During the column tests “L” liquid samples (Fig. 1) were collected at the column outlet every 1
215 PV. At the end of each test, the column was dissected and porous medium “S” samples (Fig.
216 1) were collected every 1 centimetre to determine the mass of fibres filtered out by column.
217 Both “L” and “S” sample types were then analysed by means of SEM-EDS.



218

219 Fig. 1. Schematic representation of the experimental setup used for crocidolite transport test;
 220 S and L represent, respectively, the solid and liquid sample location.

221 2.4 SEM-EDS analyses

222 Both liquid “L” and solid “S” samples were collected during each column test and analysed by
 223 means of SEM-EDS. An aliquot (dependant on suspension turbidity) of the liquid samples was
 224 first filtered on a polycarbonate membrane (25 mm diameter, 0.1 μm porosity) using a vacuum
 225 filtration system. To prevent the sedimentation and agglomeration of fibres, samples were
 226 sonicated for 8 minutes before filtration. As regards solid samples, porous medium aliquots
 227 were resuspended in 10 mL deionized water and sonicated for 8 minutes to ensure the
 228 complete detachment of crocidolite from the sand fraction. A 0.1 mL aliquot of the supernatant
 229 was then filtered on polycarbonate membranes with the same procedure applied for liquid
 230 samples. All the filtering membranes were left to dry at room temperature, adequately covered
 231 to avoid any contaminations. Then, membranes were mounted on aluminium sample holders
 232 using graphite tape and coated by a conductive graphite layer.

233 All liquid and solid samples membranes were analysed by means of a JEOL JSM IT300LV
 234 SEM with W emitter, coupled with an EDS Oxford INCA Energy 200 X-act SDD thin window
 235 detector. 0.1 mm^2 of each membrane surface was scanned acquiring 0.003 mm^2 images with
 236 a resolution of 32 pixel/ μm .

237 For liquid samples, crocidolite was counted regardless of length, width, or aspect ratio after
 238 verifying the chemical composition. Concentration in f/L was then calculated considering the
 239 volume of sample filtered through the porous membrane, following the method proposed by
 240 the Regional Agency for the Protection of the Environment of Piedmont (Italy) (ARPA-
 241 Piemonte, 2021). In addition to the fibre number, the SEM image analysis allowed to measure
 242 the length and width of the fibres and to estimate their volume and mass by approximating
 243 their shape to a cylinder and considering a crocidolite density of 3.37 g/cm^3 (US-EPA, 1983).
 244 The conversion of number concentration (f/L) into mass concentration (mg/L) was performed
 245 adapting an Italian guideline for massive samples investigations (DM 06/09/1994).

246 The quantitative analysis of liquid samples was performed only during the injection step (phase
 247 ii in section 2.3). A qualitative analysis was instead performed on the liquid samples collected
 248 during the release step (phase iv in section 2.3) to gain insights on the dimensional and
 249 morphological characteristics of the crocidolite released as a result of the pH variation of the
 250 flushing solution. As far as concern the “S” samples, SEM-EDS analyses were performed to

251 obtain qualitative data on the amount of crocidolite retained in different segments of the
252 column.

253 Throughout the paper “F” will be used to refer to crocidolite concentration in number (f/L)
254 measured by SEM-EDS, while “M” to crocidolite concentration in mass (mg/L). “F5” indicates
255 the number concentration of fibres with AR > 3, width < 3 µm and length > 5 µm, “F10”
256 indicates the number concentration of fibres with AR > 3, width < 3 µm and length ≥ 10 µm.
257 Subscript “0” is added when the concentration refers to the injected suspensions.

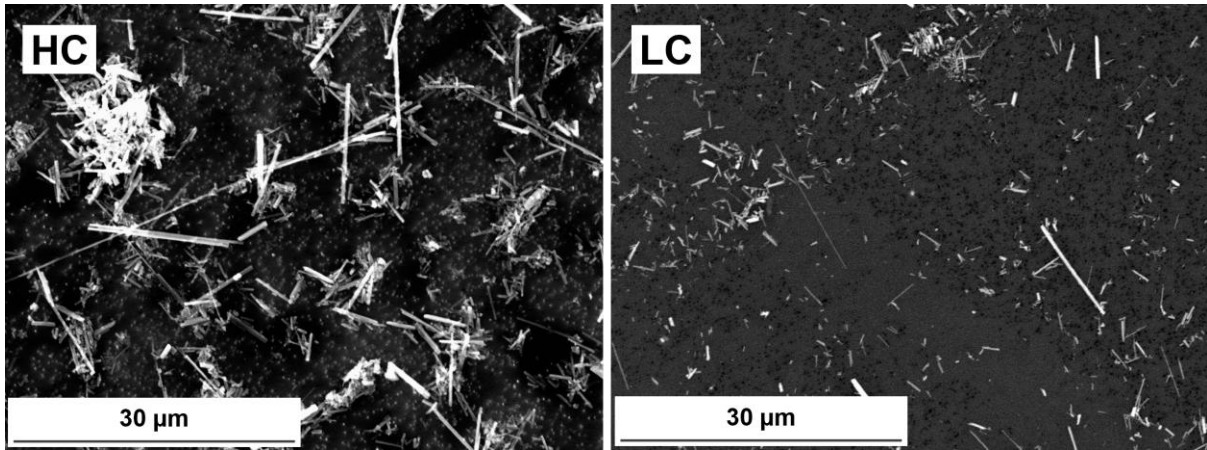
258 3 Results and discussion

259 3.1 Characterisation of asbestos suspensions

260 The first batch, HC, shown in Fig. 2, was obtained by collecting the supernatant of the 300
261 mg/L crocidolite suspension after a 5 h settling. The suspension had an average concentration
262 in number of $2.9 \cdot 10^{11}$ f/L (as shown in Table 1), which was evaluated by analysis of SEM
263 images. The corresponding mass concentration of 92.9 mg/L was estimated as described in
264 section 2.4. The mean value of F5 was $1.6 \cdot 10^{10}$ f/L (5.5% of the total) and the mean value of
265 F10 was $2.8 \cdot 10^9$ f/L (1.0% of the total). The maximum AR value was 102, while the average
266 value was 10. The HC size distributions are shown in Figure S6 (SI).

267 The second batch, LC, shown in Fig. 2, was obtained by filtering the HC suspension through
268 a coarse sand bed using the setup described in section 2.3. The resulting suspension had a
269 concentration in number of $9.9 \cdot 10^{10}$ f/L, shown in Table 2, corresponding to a mass
270 concentration of 5.5 mg/L, similar to the mass concentration detected in aquifers in NOA-rich
271 areas (Buzio et al., 2000). The F5 concentration was $1.1 \cdot 10^9$ f/L (1.1% of the total) and the
272 F10 concentration was $2.1 \cdot 10^8$ f/L (0.2% of the total). The AR mean value was 8 with a
273 maximum of 92. The LC size distributions are shown in Figure S6 (SI).

274 It must be noted that, despite the number concentration of fibres in the HC suspension is
275 comparable to LC one ($2.9 \cdot 10^{11}$ f/L for the HC and $9.9 \cdot 10^{10}$ f/L for the LC), a substantial mass
276 concentration difference between the two suspensions is observed (92.9 mg/L for the HC and
277 5.5 mg/L of the LC). This discrepancy between the number and mass concentration is justified
278 by the different fibre size distribution in the two samples. In particular, the F5 value is higher
279 in the HC suspension than in the LC one. These fibres contribute significantly to the total
280 asbestos mass in the samples; indeed, the F5 amount is one order of magnitude higher in the
281 HC suspension ($1.6 \cdot 10^{10}$ f/L) than in the LC one ($1.1 \cdot 10^9$ f/L). On the contrary, the LC
282 suspension contains a high number of shorter fibres, which do not contribute significantly to
283 the total asbestos mass in the samples.



284

285 Fig. 2. SEM pictures of tested crocidolite suspensions. HC) high concentration suspension;
 286 LC) low concentration suspension.

287 **3.2 Column transport tests**

288 Two sets of transport tests, conducted by injecting crocidolite suspensions in 1D columns filled
 289 with sand material, were performed to probe the mobility of the fibres mimicking two realistic
 290 scenarios. The first set was conducted to simulate the mobility of a highly concentrated
 291 suspension (HC) in an aquifer system close to the source of contamination, the second to
 292 investigate the transport of a low concentrated suspension (LC) in a contaminated plume far
 293 from the release zone. The following sections describe the influence of asbestos size and
 294 concentration, and of the porous media granulometry, and of physicochemical parameters on
 295 the crocidolite mobility.

296 **3.2.1 HC transport experiments**

297 The first set of column tests was performed injecting the HC suspension through three types
 298 of quartz sand with different grain size distributions (coarse, medium and fine). The
 299 breakthrough curve (Fig. 3) of the experiment in coarse sand showed C/C_0 values greater than
 300 20% during the whole injection step, with a maximum value of 30% observed at about 2 PV.
 301 Lower values, namely 2.2% and 0.3%, were respectively found for the medium and fine sand
 302 after 3 PV from the start of the injection. A higher retention of the crocidolite is observed when
 303 reducing the grain size distribution of the sand. This trend is also confirmed by the number
 304 concentration determined by SEM-EDS analyses (Table 1), which shows F values for the
 305 coarse, medium and fine sand equal to $5.7 \cdot 10^{10}$ f/L, $2.1 \cdot 10^9$ f/L, and $7.8 \cdot 10^8$ f/L respectively,
 306 for the liquid samples L2 collected after 3 PV at the outlet of the column. The corresponding
 307 M values, 3.69 mg/L for coarse, 0.22 mg/L for medium and 0.16 mg/L for fine sand, reflect the
 308 same tendency (Table 1). These results indicate that, despite a large amount of the injected
 309 asbestos is filtered out by the sandy media, a non-negligible fraction of the fibres is still mobile.

310 These results prove that crocidolite with a negatively charged surface can be transported,
 311 even in the absence of DOM, through negatively charged quartz sands. However, since only
 312 fibres longer than $5 \mu\text{m}$ (characterized by width $< 3 \mu\text{m}$ and $AR > 3$) are considered dangerous
 313 to health if inhaled, it is of crucial importance to also assess the geometric characteristics of
 314 the fibres. As reported in Table 1, the average concentration of fibres longer than $5 \mu\text{m}$ (F_{50})
 315 in the injected HC suspension was equal to $1.6 \cdot 10^{10}$ f/L, the 17.5% of which consisted in fibres
 316 even longer than $10 \mu\text{m}$ (F_{100} equal to $2.8 \cdot 10^9$ f/L). All the outlet samples are characterized
 317 by a F_5 count well above 10^7 f/L and more specifically in the range $2.1 \cdot 10^7$ - $5.3 \cdot 10^8$ f/L (Table
 318 1). The concentrations of these samples are comparable or higher than the ones found in
 319 suspensions used by Avataneo et al. (2022) and by Roccaro and Vagliasindi (2018), who
 320 reported that waterborne values of $4.4 \cdot 10^7$ f/L and in the range $7.9 \cdot 10^3$ - $2.5 \cdot 10^4$ f/L,

321 respectively, can cause an airborne contamination above the 1 f/L attention threshold (WHO,
322 2000), whether the water-to-air migration of fibres is triggered under specific conditions.

323 Fibres up to 18 μm were found in outlet samples of coarse and medium sand tests (see L_{max}
324 in Table 1), while fibres up to 7 μm were measured in the fine sand test outlet. In particular,
325 the 0.8% (coarse), 0.5% (medium) and 0.2% (fine) of F_{50} were recovered at the column outlet
326 after 3 PV of suspension injection (L2, Fig. 4). Conversely, fibres longer than 10 μm were
327 found only in sample L1 at the outlet of the coarse and medium sand columns. In these two
328 samples the same F_{10} concentration value was found, $1.1 \cdot 10^8$ f/L, which is higher than the
329 EPA maximum contaminant level for drinking water ($7 \cdot 10^6$ f/L for fibres longer than 10 μm).
330 The results of the column tests performed at HC highlight that fibres longer than 10 μm barely
331 migrate, while crocidolite with length between 5 and 10 μm might be substantially transported
332 through sandy aquifers.

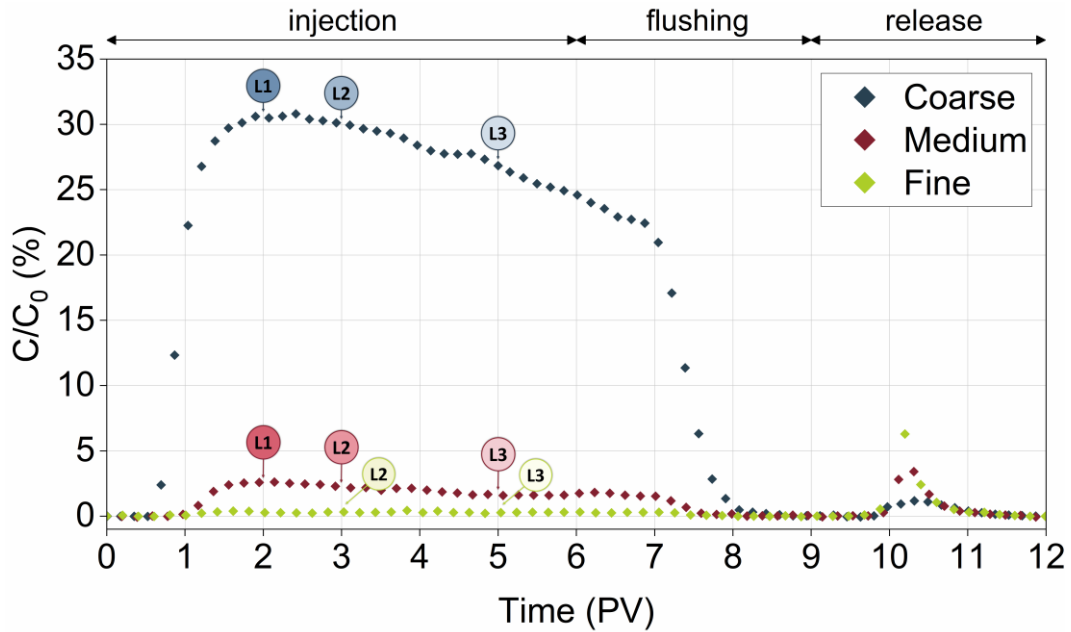
333 As far as concern transport mechanisms, the particles deposition in porous media is
334 considered the result of both physical (i.e. mechanical filtration of thick particles in small pores)
335 and physicochemical processes (i.e. particle attachment on the porous medium surface due
336 to attractive particle-grain interaction forces). The first mechanism is typically assumed
337 irreversible, meaning that no particle remobilization is expected upon a change of the pore
338 water chemistry. As for attachment, instead, a variation of hydrochemical conditions (e.g. pH
339 increase) may induce a detachment of the deposited particles due to the reduction of the
340 attractive forces between the particles and the sand grains. In the case of crocidolite, physical
341 filtration in small pores is expected to play a major role for the longer fibres. (Elimelech,
342 1995)(Bradford et al., 2002)(Lin et al., 2021) This is confirmed by SEM-EDS analysis of porous
343 media sample collected at the first column centimetre for each tested sand (coarse, medium,
344 and fine). All three samples (HC_S1_Coarse, HC_S1_Medium, HC_S1_Fine) presented a
345 high number of fibres longer than 5 μm and even longer than 10 μm (Fig. 5). Moreover, visual
346 inspection confirmed that the length of trapped fibres increases when decreasing the sand
347 grain size distribution.

348 Another evidence confirming the occurrence of mechanical filtration is related to the evolution
349 of the outlet concentration, which decreases over time once the filtered fibres start to clog the
350 pores. This can be observed by the breakthrough curves reported in Fig. 3 where the
351 absorbance ratio decreases from 30% to 22% for the coarse sand and from 2.5% to 1.5% for
352 the medium sand. However, the decrease in absorbance is not evident for the fine sand,
353 probably due to the low absorbance value detected. The increased filtration efficiency over
354 time is also confirmed by the SEM-EDS analysis of the liquid samples collected at the column
355 outlet, shown in Fig. 4. The graph shows a clear decrease over time (from sample L1 to sample
356 L3) of the F_5 values for all three quartz sands. In particular, the F_5/F_{50} ratio decreases from
357 3.8% to 0.4% in coarse sand, from 2.7% to 0.2% in the medium sand, and from 0.18% to
358 0.16% in the fine one.

359 The release of reversibly attached fibres was induced by increasing the pH of the flushing
360 water (Pulido-Reyes et al., 2022; Beryani et al., 2020; Tosco et al., 2009). As shown by the
361 BCs in Fig. 3, a significant release peak is observed during the NaOH flushing for all three
362 sand types. The peak height is inversely proportional to the sand grain size distribution,
363 suggesting that physicochemical processes are more intense in the finer sand than in the
364 coarser one due to the greater surface area of the porous medium in fine sand. A liquid
365 sample was also collected during the release phase of the coarse sand column experiment
366 and a SEM picture was acquired (Figure S7, SI). The image demonstrates the presence of
367 fibres in the eluate, some of them with length of about 5 μm . This result indicates that, even

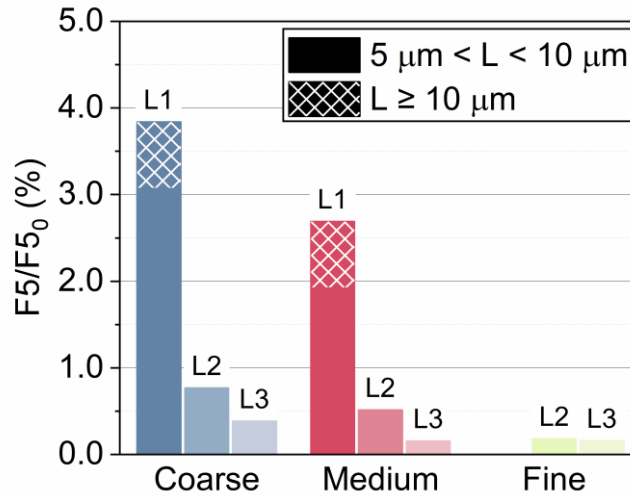
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if initially retained by the porous medium, fibres may be remobilized if there is a change in the aquifer hydrochemical conditions.



370

371 Fig. 3. Experimental breakthrough curves of normalized absorbance concentration of HC
372 crocidolite suspension during transport experiments performed in quartz sand with different
373 granulometry (coarse, medium, fine). Labels correspond to the liquid samples (L) collected at
374 the column outlet and analysed by SEM-EDS.



375

376 Fig. 4. Bar chart of concentration of fibres longer than 5 µm ($F5$) in liquid samples collected at
377 the column outlet (L1, L2, L3) as a percentage of concentration of fibres longer than 5 µm of
378 the HC suspension ($F5_0$) for the three tested sands (coarse, medium, fine); solid fill) the portion
379 of fibres longer than 5 µm and shorter than 10 µm; pattern fill) the portion of fibres longer than
380 (or equal to) 10 µm.

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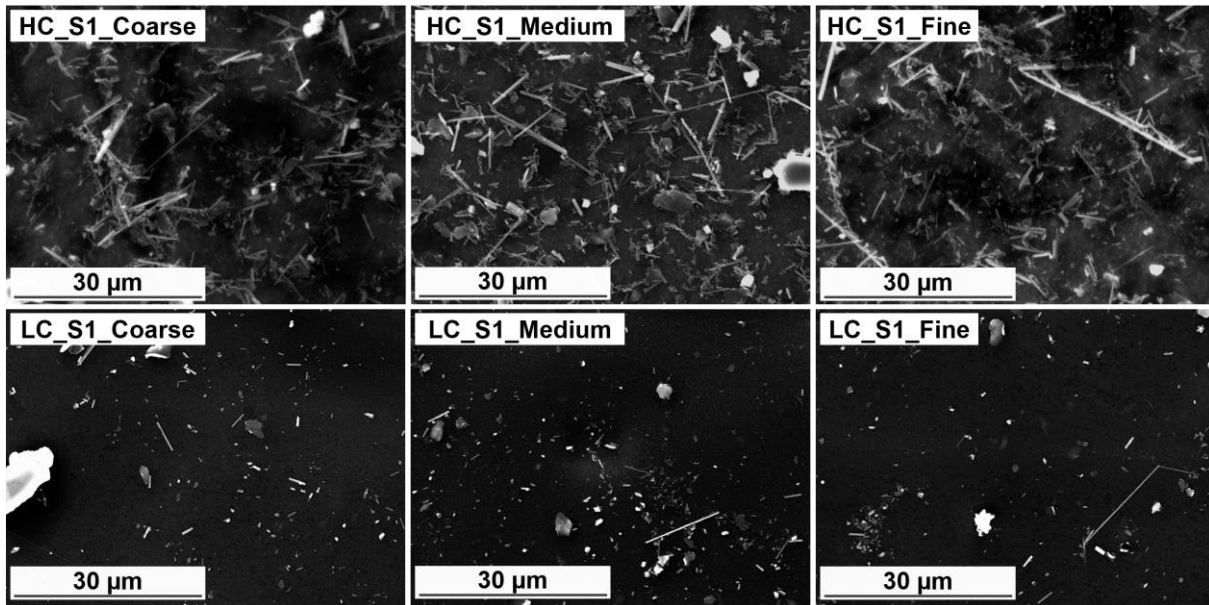
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386 Table 1. F (f/L) and M (mg/L) concentrations of crocidolite in the inlet (HC) and in liquid
 387 samples collected at the column outlet (L1, L2, L3) during transport tests performed with the
 388 high concentration suspension (HC). F5, number concentration of fibres longer than 5 µm, and
 389 F10, number concentration of fibres longer than 10 µm values, are also reported. Dimension
 390 ranges (L=length, W=width) of detected fibres are reported for each sample. Additional details
 391 regarding quantitative results obtained on SEM-EDS analyses are reported in Table S2 (SI).

		F (f/L)	F5 (f/L)	F10 (f/L)	M (mg/L)	L _{min} (µm)	L _{max} (µm)	W _{min} (µm)	W _{max} (µm)
HC		2.9E+11	1.6E+10	2.8E+09	92.91	0.520	17.790	0.077	0.766
Coarse sand	L1	6.3E+10	5.3E+08	1.1E+08	3.81	0.292	18.138	0.044	0.554
	L2	5.7E+10	1.1E+08	-	3.69	0.311	6.205	0.062	0.560
	L3	3.7E+09	5.3E+07	-	0.56	0.330	7.343	0.083	0.466
Medium sand	L1	6.3E+09	3.7E+08	1.1E+08	2.25	0.517	18.248	0.069	0.532
	L2	2.1E+09	7.0E+07	-	0.22	0.436	6.151	0.056	0.348
	L3	2.1E+09	2.1E+07	-	0.22	0.404	6.468	0.075	0.405
Fine sand	L2	7.8E+08	3.0E+07	-	0.16	0.532	6.047	0.079	0.407
	L3	8.9E+08	2.6E+07	-	0.17	0.435	7.351	0.069	0.473

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393

394 Fig. 5. SEM pictures of the porous media samples collected from the first column centimetre
 395 after the transport test with: HC suspension for coarse sand (HC_S1_Coarse), medium sand
 396 (HC_S1_Medium), fine sand (HC_L1_Fine); LC suspension for coarse sand (LC_S1_Coarse),
 397 medium sand (LC_S1_Medium), fine sand (LC_S1_Fine).

398 3.2.2 LC transport experiments

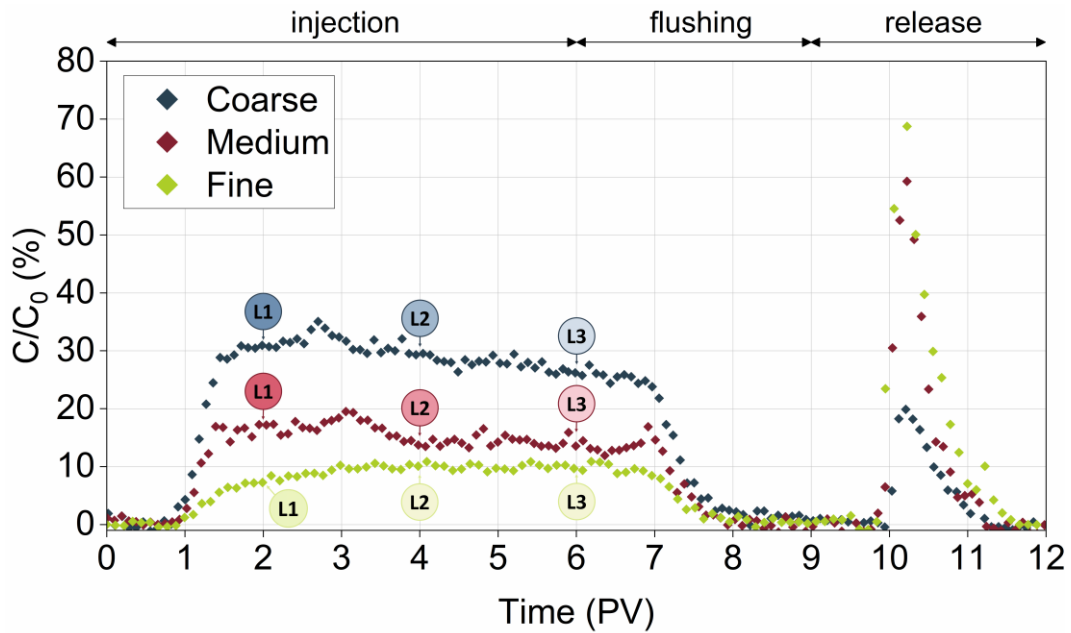
399 The second set of column tests was carried out injecting the LC suspension through the three
 400 porous media. The resulting normalized absorbance curves are shown in Fig. 6. Similarly to
 401 what observed for HC suspension, the BCs show an increase of the crocidolite retention when
 402 reducing the grain size distribution of the sand. However, overall, the crocidolite low

403 concentrated suspension shows a much higher mobility than the one observed during the HC
404 experiments. The mobility increase is particularly pronounced for tests performed in the
405 medium and fine sand, where the maximum value of C/C_0 increases from 1.1% (HC) to 18.4%
406 (LC) for medium sand and from 0.5% (HC) to 9.7% (LC) for fine sand. The higher mobility of
407 the LC suspension is probably due to the lower F_{50} value compared to HC one, in terms of
408 both absolute number ($1.1 \cdot 10^9$ f/L for LC and $1.6 \cdot 10^{10}$ f/L for HC) and percentage on total
409 (1.1% for LC and 5.5% for HC). The lower content of long fibres reduces the probability of
410 porous medium clogging, which would have resulted in an increase of the filtration efficiency
411 over time. This is confirmed by the shape of the BCs in Fig. 6 that, differently from what
412 observed during the HC experiments, remain relatively constant during all the injection
413 phases.

414 From the results of the SEM-EDS analysis, F_5 values range between $1.1 \cdot 10^7$ and $1.5 \cdot 10^8$ f/L
415 for the outlet liquid samples (Table 2), which are of the same order of magnitude as the HC
416 tests. Interestingly, due to a lower porous media clogging, a F_{10} amount higher than the EPA
417 threshold for drinking water was found in the L1 and L3 samples of the coarse sand (Table 2).
418 The L2 liquid sample of the coarse sand test has a concentration of fibres longer than $5 \mu\text{m}$
419 abnormally lower than L1 and L3. This difference can be occurred due to an incorrect sampling
420 or sample preparation. Looking at the normalized F_5 value over time (Fig. 7) a net decreasing
421 trend was not observed for the LC tests, in contrast with what observed for HC set in Fig. 4.
422 This is a further evidence of a minor porous media clogging.

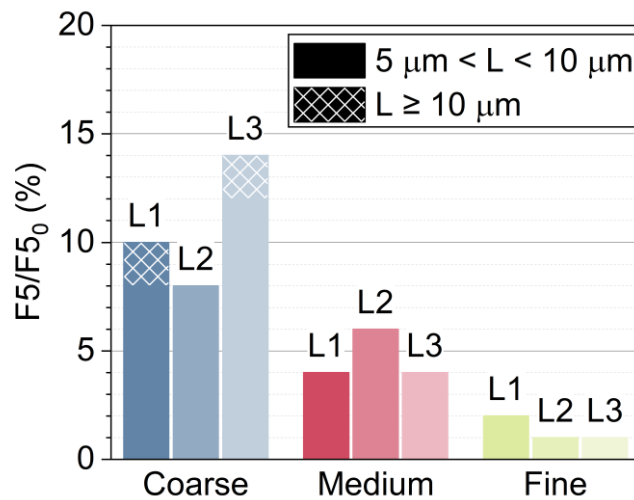
423 As for the HC experiments, a high pH flushing was performed at the end of the test to
424 discriminate between reversible and irreversible fibre deposition. Despite the smaller amount
425 of fibre retained in the column during the injection phase (i.e. higher BCs), the release peaks
426 for the LC set result to be higher than for the HC tests. The absorbance peak varies from
427 18.0% to 68.8% for the tests performed with the LC suspension and from 1.2% to 6.3% for the
428 HC one. This result suggests that asbestos transport may be influenced not only by
429 mechanical filtration, but also by physicochemical reversible processes.

430 The SEM images of the porous media portions sampled from the first centimetre of each
431 column (LC_S1_Coarse, LC_S1_Medium, LC_S1_Fine in Fig. 5) revealed the presence of
432 fibres in all the samples. More specifically, they showed the presence of fibres longer than $5 \mu\text{m}$
433 and $10 \mu\text{m}$, but in a smaller amount compared to the HC samples. This is further evidence
434 that clogging is lower for LC suspension compared to HC, due to the lower number of fibres
435 longer than $5 \mu\text{m}$ trapped in the sand. Moreover, being reversible processes higher for LC
436 suspension, most of the shorter fibres were flushed during the NaOH release phase and
437 therefore are not visible in Fig. 5. The SEM pictures of the liquid samples collected during the
438 release phase (Figure S8, SI) show the presence of crocidolite with length up to $5 \mu\text{m}$,
439 particularly in medium and fine sand tests.



440

441 Fig. 6. Experimental breakthrough curves of normalized absorbance concentration of LC
 442 crocidolite suspension during transport experiments performed in quartz sand with different
 443 granulometry (coarse, medium, fine). Labels correspond to the liquid samples (L) collected at
 444 the column outlet and analysed by SEM-EDS.



445

446 Fig. 7. Bar chart of concentration of fibres longer than 5 μm (F5) in liquid samples collected at
 447 the column outlet (L1, L2, L3) as a percentage of concentration of fibres longer than 5 μm of
 448 the LC suspension ($F5_0$) for the three tested sands (coarse, medium, fine); solid fill) the portion
 449 of fibres longer than 5 μm and shorter than 10 μm ; pattern fill) the portion of fibres longer than
 450 (or equal to) 10 μm .

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456 Table 2. F (f/L) and M (mg/L) concentrations of crocidolite in the inlet (LC) and in liquid samples
 457 collected at the column outlet (L1, L2, L3) during transport tests performed with the low
 458 concentration suspension (LC). F5, number concentration of fibres longer than 5 μm , and F10,
 459 number concentration of fibres longer than 10 μm values, are also reported. Dimension ranges
 460 (L=length, W=width) of detected fibres are reported for each sample. Additional details
 461 regarding quantitative results obtained on SEM-EDS analyses are reported in Table S3 (SI).

		F (f/L)	F5 (f/L)	F10 (f/L)	M (mg/L)	L _{min} (μm)	L _{max} (μm)	W _{min} (μm)	W _{max} (μm)
LC		9.9E+10	1.1E+09	2.1E+08	5.49	0.191	13.223	0.049	0.404
Coarse sand	L1	4.3E+09	1.1E+08	2.1E+07	1.14	0.435	11.830	0.104	0.528
	L2	5.8E+09	2.1E+07	-	0.93	0.354	5.044	0.070	0.551
	L3	6.5E+09	1.5E+08	2.1E+07	1.18	0.317	14.530	0.070	0.762
Mediu m sand	L1	5.4E+09	4.2E+07	-	0.43	0.243	8.821	0.070	0.379
	L2	3.8E+09	6.3E+07	-	0.39	0.267	9.189	0.066	0.423
	L3	2.3E+09	4.2E+07	-	0.28	0.386	6.683	0.088	0.325
Fine sand	L1	3.7E+09	2.1E+07	-	0.75	0.311	5.144	0.062	0.543
	L2	2.8E+09	1.1E+07	-	0.38	0.352	5.256	0.063	0.517
	L3	1.7E+09	1.1E+07	-	0.19	0.320	5.532	0.072	0.418

462 4 Conclusion

463 This study investigated the transport mechanism of waterborne crocidolite in saturated porous
 464 media mimicking sandy aquifers. Crocidolite was selected to represent amphibole asbestos
 465 behaviour, which exhibits negative net surface charge in water at any pH. Although asbestos
 466 mobility in subsoil has been generally neglected (or considered negligible), the results here
 467 presented show that bare crocidolite can be mobile in negatively charged porous media such
 468 as quartz sand aquifers.

469 Our results show that highly concentrated crocidolite suspensions generally determine lower
 470 breakthrough concentrations at the outlet of the columns due to the clogging of the porous
 471 medium which is induced mainly by the mechanical filtration of the longer fraction of the fibres.
 472 The decrease of the grain size distribution of the porous medium determines a strong
 473 decrease of the output concentrations further confirming the important role of mechanical
 474 filtration. On the contrary the injection of low concentration suspensions corresponds to a
 475 higher fibre recovery (if normalized to inlet concentration). Under these conditions, the
 476 decrease of the grain size of the porous media determines a less pronounced decrease in
 477 outlet concentrations. The transport is thus less influenced by the mechanical filtration (due to
 478 the reduced number of long fibres) but it is still dependent on the physicochemical interactions
 479 occurring under unfavourable deposition conditions (since the fibres and collectors are
 480 characterized by the same surface charge).

481 By characterizing the fibre suspensions sampled from the column outlet, we demonstrated
 482 that the medium and fine sandy media can retain most of the fibres longer than 10 μm , while
 483 5-to-10- μm -long fibres can easily flow through. These results, obtained under specific
 484 laboratory conditions, might suggest that groundwater extracted downstream a contamination
 485 source is potentially safe to drink, but precautions should be taken to avoid water vaporization
 486 or fibre volatilization. In coarser aquifer systems also fibres longer than 10 μm can migrate

487 downwards a source of contamination determining a potential hazard for direct oral intake of
488 contaminated groundwater.

489 In addition, our results indicate that a change in the water physicochemical parameters, e.g.
490 a pH increase, can remobilize fibres primarily attached to the porous media, proving that
491 waters considered safe to use can become potentially harmful after natural or human induced
492 events that cause alterations of the groundwater geochemistry.

493 This study indicates that groundwater migration is a possible exposure pathway that should
494 be included in human health risk assessment evaluations and that further studies have to be
495 conducted to elucidate the risk induced by exposure to asbestos contaminated drinking water.

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