

Mobility of asbestos fibers below ground is enhanced by dissolved organic matter from soil amendments



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ABSTRACT

To prevent exposure of millions of people living near thousands of contaminated sites to asbestos, the sites are typically capped with soil and amendments, assuming asbestos mobility underground is negligible in all conditions. Here, we disprove this long-standing assumption and showed that the presence of certain types of dissolved organic matter (DOM) can facilitate the transport of the asbestos fibers through packed sand or soil columns. The column scale simulates asbestos transport without and without DOM, although asbestos transport at a longer scale through aquifer may vary because of soil heterogeneity. Our data shows that the extent of fiber transport in soil is affected by DOM quality: fulvic acid > humic acid > natural organic matter isolated from Suwanee River, a model terrestrial organic carbon. We attribute the results to the ability of DOM to affect aggregation of asbestos fibers and limit their attachment on soil. Thus, types of organic amendments applied on asbestos-contaminated sites may have an unintended consequence: transport of asbestos fibers to shallow groundwater to receiving streams or rivers, from where they could be resuspended in the air via irrigation or drying of the riverbed.

1. Introduction

Studies on exposure to asbestos fibers via air have received a lot of attention due to their implication to lung cancer and mesothelioma (Furuya et al., 2018; Nielsen et al., 2014; WHO, 2006), whereas exposure of asbestos via water received far less attention, despite increased risks of stomach cancer from ingested asbestos (Fortunato and Rushton, 2015). Furthermore, environmental exposure through water or soil has been assumed to be negligible (Fuller, 1977) with the premise that asbestos fibers should stick with soil and be filtered out of infiltrating water. However, recently asbestos fibers were found underground hundreds of meters away from their source (Buzio et al., 2000; Emmanouil et al., 2009; Buck et al., 2013), suggesting an alternative asbestos exposure pathway: shallow groundwater. Typically, asbestos-containing materials are buried in the soil to prevent erosion. However, smaller fibers may be produced in the waste piles by abrasion during transportation or storage. Owing to their smaller size, the fibers have a higher potential than bulk asbestos for transport in water. They could be transported from the waste piles via infiltrating water to groundwater. Use of contaminated groundwater for irrigation (Turci et al., 2016) and showering or indoor humidifiers (Roccaro and Vagliasindi, 2018) have been shown to increase asbestos inhalation risk (Fig. 1). Thus, it is critical to understand the

environmental conditions that increase the mobility of asbestos fibers from contaminated soil to groundwater.

A lack of research on asbestos fiber mobility in porous media limits our understanding of environmental factors that may trigger the transport of fibers in soil. Assuming asbestos fibers behave similarly to other mineral colloids in water, the fiber mobility in the soil can be predicted based on numerous colloid transport studies (Ryan and Elimelech, 1996). Colloids are transported through soil by infiltrating water and deposited on grain surfaces by settling, interception, and diffusion as predicted by colloid filtration theory (Elimelech, 1994). Thus, transport and removal of colloids can be affected by physical factors such as size (Pelley and Tufenkji, 2008), the shape of colloids (Seymour et al., 2013), and the distribution of pore sizes in soil (Bradford et al., 2002), and chemical factors such as pH (Bergendahl and Grasso, 1999), ionic strength (Tufenkji and Elimelech, 2005), and the presence of phosphates and dissolved organic carbon (Hofmann and Liang, 2007), which affect the interaction between colloids and soil.

Unique physical and surface chemical properties of asbestos fibers could affect their transport in soil. Asbestos fibers have long aspect ratios with fiber length within the range of 100 μm (Skinner et al., 1988). Thus, they are expected to get trapped in soil pores due to physical straining (Bradford et al., 2006). As diffusion or transport of fibers along the

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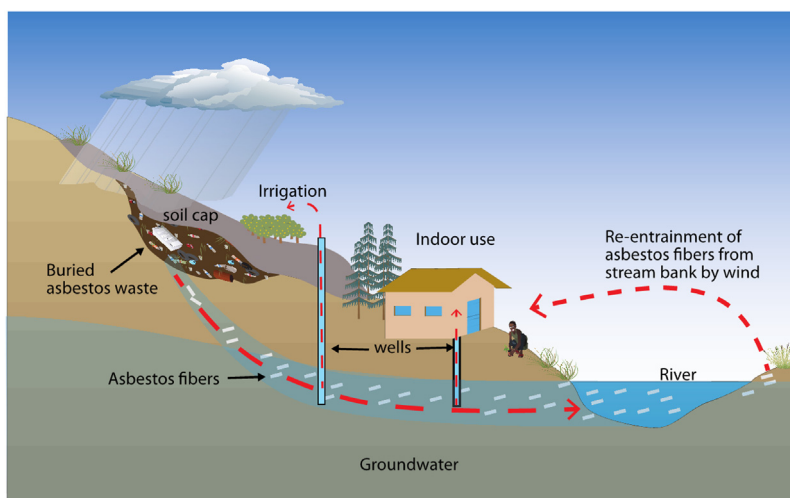


Fig. 1. Potential transport pathways of asbestos fibers in groundwater from contaminated sites.

longitudinal axis is expected to be higher (Wu et al., 2017), fibers can pass through pores that are smaller than the length but bigger than the width of fibers. Some asbestos minerals such as chrysotile—the most commonly used asbestos mineral—exhibit a net positive surface charge in the water around neutral pH because of an outer brucite-like layer (Pollastri et al., 2014). As natural soil has a net negative surface charge, electrostatic attraction between soil and chrysotile fibers can limit their mobility in soil. The surface charge, however, can be reversed with increasing pH or exposure to dissolved organic matter (DOM) (Morales et al., 2011), which in turn could increase their mobility.

DOM is ubiquitous in surface and subsurface environments. The concentration of DOM is expected to be high in the asbestos-contaminated site because the protective capped soil layer typically contains biosolids or compost to support vegetation cover (Li et al., 2014; Gonneau et al., 2017) and to support soil microorganisms, which can biodegrade asbestos using exudates (Mohanty et al., 2018). Yet, the ability of DOM to increase the transport of asbestos fibers has not been evaluated. Herein, we examine the effect of the presence of DOM on chrysotile fiber mobility in soil. By injecting chrysotile fiber suspensions in the presence of DOM through saturated soil or sand columns, we identify the potential culprit for asbestos fiber mobility in soil or groundwater.

2. Material and methods

2.1. Asbestos suspension

Chrysotile was chosen as the model asbestos mineral because it is the most commonly used asbestos type and ubiquitous in asbestos-contaminated sites (Li et al., 2014). To produce monodispersed chrysotile fibers, chrysotile ore (Globe, Arizona) was first ground in dry condition (Salamatipour et al., 2016) and then purified using ammonium chloride and heat to remove brucite impurities (Benson, 1982). The purified fibers were dispersed by an ultrasonic probe, and large fibers (length > 50 μm) were removed by settling the fibers with gravity for 30–45 min. Based on Stoke's law, 30 min is sufficient to settle particles larger than 50 μm assuming the particle density of asbestos is 2.6 g cm^{-3} . The smaller size fraction was used in this experiment due to their high potential for mobility compared to the bulk particle. The size also simulates silt and clay particles, which are typically eroded and transported from asbestos waste piles. The resulting suspensions were diluted with deionized water to lower the turbidity, equivalent to the fiber concentrations of $0.3 \pm 0.05 \text{ g L}^{-1}$, based on the calibration curve between fiber concentrations and absorbance or turbidity of samples. The concentration was determined based on a calibration curve of suspensions containing a known quantity of dried

asbestos fibers of a similar particle size range (Mohanty et al., 2018). To examine the effect of DOM on asbestos mobility, three types of organic carbon were used: fulvic acid, humic acid, and natural organic matter from Suwannee River (Green et al., 2015). Concentrated DOM prepared from all types of organic carbon fractions were spiked into the asbestos suspensions to adjust the concentration of DOM between 0–25 mg L^{-1} . The DOM stock solution (250 mg L^{-1}) was prepared by dissolving 25 mg of organic matter in 25 mL of ultrapure water raised to pH 11 and diluting the stock solution to 100 mL. Filtration of stock solution through 0.7 μm carbon fiber filter did not result in changes in absorbance indicating the absence of particulate organic carbon (>0.7 μm).

To adjust the geochemical conditions of the suspensions, pH, ionic strength, and DOM concentration were varied. The pH was adjusted between 3–12 by adding a buffer solution containing 1 mM MOPS buffer (MOPS, 3-N-morpholino propanesulfonic acid) and small quantities of concentrated HCl or NaOH. To examine whether the organic buffer has any effect on asbestos mobility, pH was also adjusted with only HCl and NaOH. To examine the effect of ionic strength, NaCl was added to the solutions to adjust the ionic strength to values between 0.1 mM–100 mM. To examine the effect of other ions that may be present in soil leachate, mobility experiments were repeated using soil leachate or suspensions containing phosphate (1 and 5 mg L^{-1}). To isolate the effect of confounding factors, we examine the effect of geochemical factors including pH, ionic strength, dissolved organic carbon quantity, soil leachate, and phosphate concentration in sand columns. All soil column experiments were conducted at pH 7 due to their relevance to the most natural soil pH.

2.2. Asbestos mobility experiments

Coarse quartz sand (ASTM 20-30) and soil collected from BoRit, PA—a site previously contaminated with asbestos-containing material—were used as porous media for this study. Sand or sieved (< 2 mm) soil was packed in glass columns (2.54 cm ID and 15 cm length) and flushed with deionized water at 12 cm h^{-1} or 2 pore volumes per hour using a peristaltic pump for 24 h from bottom to top to displace air, maintain saturation, and remove any loosely attached natural colloids. Solution without asbestos, adjusted to a specific pH, ionic strength, with and without specific DOM, was injected from the bottom of the column to equilibrate the packed sand or soil with the geochemical composition of the asbestos suspension. After flushing 10 pore volumes of the asbestos-free solution, 4 pore volumes of asbestos suspension with and without DOM was injected through the columns, followed by injection of 4 pore volumes of asbestos-free solution to flush fibers from pore water. The effluents were collected at 10 mL

fractions and analyzed for asbestos fibers. Control experiments without asbestos were conducted to measure the background turbidity under the same experimental conditions. The difference in turbidity of samples collected in experiments with and without asbestos fibers was used to quantify the approximate concentration of asbestos fibers in the effluent. This method eliminates the contribution of soil particles and any background asbestos present originally in the soil to effluent turbidity values and helps estimate the percentage of injected asbestos transported through sand or soil columns.

Effluents were analyzed for pH and optical density (Hach DR 2700). The optical density of suspensions at 595 nm was used as a surrogate measure of asbestos concentration in water. At this wavelength, the color of the suspension from any added DOM has a negligible (<0.1 %) effect on the optical density of the suspension. This method assumes that any increase in absorbance during the experiments was due to the presence of asbestos fibers or particles in the effluent. To confirm the presence of asbestos fibers, the effluent samples were concentrated to 10x concentration. Briefly, nearly 10 mL suspension in 15 mL centrifuge tube was centrifuged at 5000 g for 10 min, and 9 mL supernatant was discarded, leaving behind settled asbestos with 1 mL of effluent. A drop of concentrated (10x) suspension was dried and analyzed for fiber morphology using a scanning electron microscope with an energy-dispersive X-ray spectroscope (SEM-EDX). The asbestos samples were not coated with gold before SEM analysis. To measure any change in surface charge of chrysotile fibers in the presence of DOM, electrophoretic mobility of asbestos fibers at neutral pH was measured with and without the presence of humic acid, fulvic acid, and natural organic matter (10 mg/L), and corresponding zeta potential was estimated (Pollastri et al., 2014).

3. Results and discussion

3.1. Dissolved organic matter enhanced asbestos mobility in sand and soil columns

Using three types of organic carbon from the same sources, we showed that the presence of DOM increased asbestos mobility in soil columns (Fig. 2) and sand columns (Fig. S1, Supporting Materials). The columns experiments where no DOM was used showed no detectable breakthrough curves in sand columns (Fig. S1) and soil columns (Fig. S2). When DOM was not used in the suspension, the effluent turbidity in columns without and with asbestos was similar, indicating limited to no asbestos mobility without DOM. In contrast, when DOM was used, effluent turbidity in columns with asbestos was significantly higher than effluent turbidity without DOM, indicating transport of asbestos through soil columns was enhanced in the presence of DOM. Subtracting the area below the breakthrough curves in control columns (without asbestos) from the area below the columns with asbestos, we estimated the recovery or net amount of asbestos transported through columns in the presence of each type of DOM. The fraction transported in the presence of FA, HA, and NOM are $10.4\% \pm 0.2\%$, $4.4\% \pm 0.8\%$, and $0.4\% \pm 0.8\%$, respectively. The result indicates that NOM has the least effect on asbestos mobility compared with humic or fulvic acids.

SEM image of asbestos fibers size in the effluent showed that some fibers with a size of nearly $10\ \mu\text{m}$ were transported through columns (Fig. 3), indicating pore size in sand columns were large enough to permit passage of large asbestos fibers, especially along with the fiber c- or z-axis. Smaller fibers were appeared lighter compared to large and thicker fibers (dark) due to excess charging under the electron beam during SEM image acquisition. It should be noted that effluent samples were concentrated by a factor of 10 by centrifugation before evaporating a drop of concentrated asbestos suspension on SEM grids. Thus, fibers were aggregated on the SEM grid. Therefore, the result should not be used to quantify aggregation state and any change in the size distribution of fiber during transport through sand or soil columns.

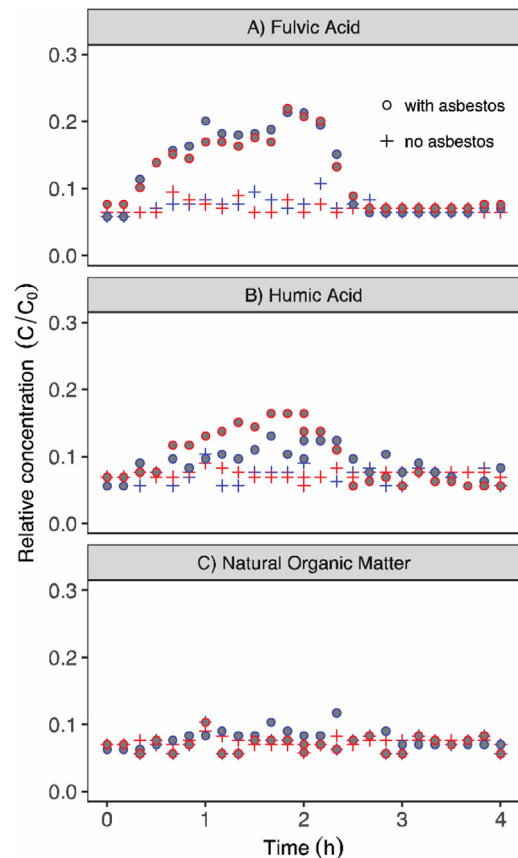


Fig. 2. Transport of asbestos fibers through duplicate soil columns, in the presence of 10 mg/L of (a) fulvic acid (FA), (b) humic acid (HA) or (c) natural organic matter (NOM). The ionic strength (I) was adjusted to 10 mM and the pH was adjusted to 7. Duplicate experiments were conducted for each condition: with or without asbestos in the influent. The data for control columns where no DOM was used is provided in Figure S2.

We attributed DOM-facilitated transport of asbestos fibers to alteration in aggregation or dispersion of asbestos fibers in pore water by DOM, which could have made the fibers move through soil pores with limited interaction with soil grains. We observed a lack of settling of fibers in fulvic acid solution in 24 h and formation of stratified layers in the absence of DOM tested in this study, indicating that the addition of DOM increase the stability of the fiber suspension (Supporting Material).

DOM can also alter the surface charge of chrysotile fibers and change their interaction with the soil. Zeta potential measurement indicates that the zeta potential of chrysotile was reversed from net positive ($2.34 \pm 0.57\ \text{mV}$) to negative ($-44.45 \pm 13.21\ \text{mV}$) in the presence of all DOM at a concentration of 10 mg/L at pH 7. Thus, electrostatic repulsion between DOM-coated chrysotile and soil could partially explain an increase in the fiber mobility in the presence of DOM. We attributed the increase in transport of asbestos fibers to a decrease in interaction of asbestos fibers with soil and an increase in stability of asbestos suspension in the presence of DOM, similar to transport of other positively charged minerals such as hematite, goethite, and magnetite (Philippe and Schaumann, 2014). However, surface charge reversal alone may not explain the observed increase in asbestos mobility in sand or soil, partly because an increase in pH from 3–12 did not have any effect on asbestos mobility in sand columns (Fig. S3). An increase in pH beyond the point of zero charges of chrysotile is expected to reverse the surface charge of chrysotile from net positive to net negative and increase their transport through the sand. However, we did not observe an increase in transport of asbestos fibers at pH 12, suggesting that pH does not affect asbestos mobility. Exposure to phosphate, which can also reverse surface charge, and soil leachate,

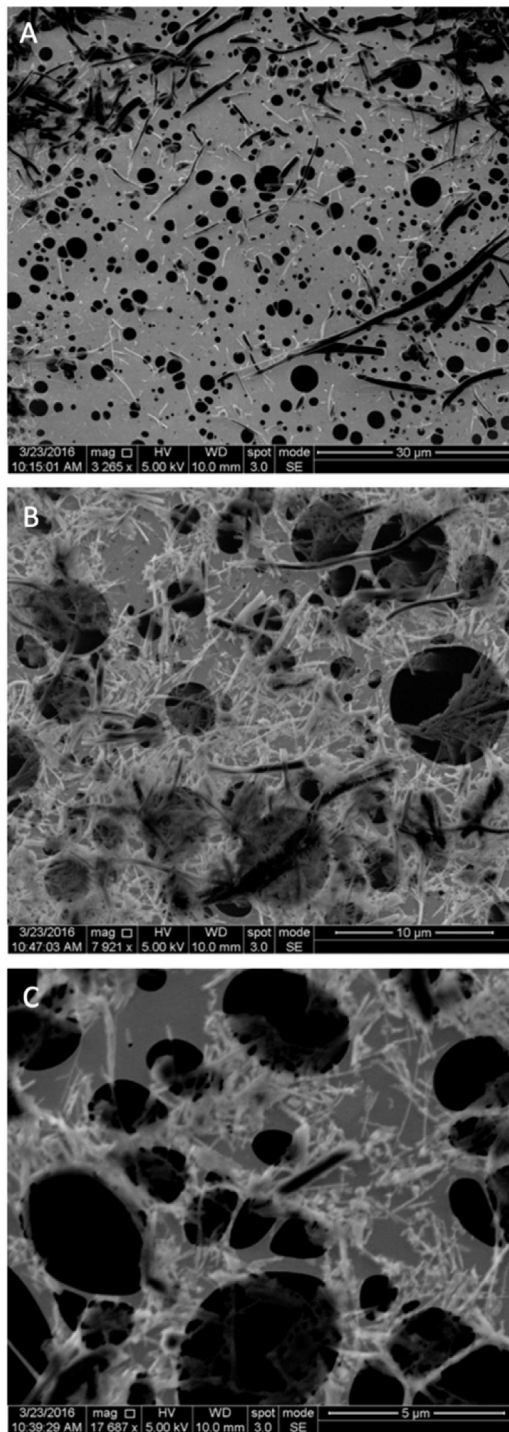


Fig. 3. SEM picture of chrysotile fibers in influent (A) and effluents of sand (B) and soil columns (C). Note the scale of the bar is different in each figure.

which may contain some organic carbon, did not alter asbestos mobility through sand columns. Thus, we assume that DOM can affect asbestos fiber behavior such as aggregation in the suspension and its interaction with soil or sand surface, both of which can affect asbestos mobility.

3.2. Dissolved organic matter quality is critical for asbestos fiber mobility

The breakthrough concentration of asbestos fibers decreased in the following order: fulvic acid > humic acid > natural organic matter (Fig. 2). Integrating the area underneath the breakthrough curve and

subtracting the area of control experiments (no asbestos), we estimated that on average 10.4 %, 4.4 %, and 0.4 % of applied asbestos fibers were transported in the presence of fulvic acid, humic acid, and natural organic matter, respectively. These results confirmed that DOM quality is critical in determining the extent to which they can increase asbestos mobility in soil. It should be noted that the pH of suspension (7.0 ± 0.2) was buffered using 1 mM or 209 mg L⁻¹ MOPS, which is also another type of DOM. Yet, effluent turbidity did not increase, indicating MOPS did not affect the mobility of asbestos fibers. A lack of fiber mobility in the presence of 209 mg L⁻¹ MOPS buffer further confirmed that DOM quality is critical for fiber mobility. However, the characteristic property of DOM or specific quality that may make it effective for asbestos mobility is not clear. Our study shows that negative charge density may be one attribute of DOM that could affect asbestos transport. Fulvic acid, humic acid, and natural organic carbon contain carboxyl or other negatively charged functional groups, which can bind with positively charged surfaces via adsorption or complexation. Based on the properties of these well-characterized organic carbons, fulvic acid has higher negative charge density than humic acid, and humic acid has higher negative charge density than natural organic carbon (Green et al., 2015). Mechanism of DOM adsorption on inorganic colloids highly depends on the characteristics of colloids and DOM characteristics such as molecular structure, charge, hydrophobicity, molecular weight, and conformation (Philippe and Schaumann, 2014). Thus, future studies should examine the relative importance of the specific DOM qualities on asbestos mobility.

3.3. Environmental implications

These results may have profound consequences on the asbestos mobility in soils and groundwater, which in turn could increase asbestos exposure to millions of people living near asbestos-contaminated sites. For instance, if the DOM released from compost and biosolids from the capped soil layer increases the release of asbestos fibers from the contaminated soil, it could increase the fiber transport to surface water via shallow groundwater. Thus, this alternative asbestos exposure route via groundwater should not be ignored, and organic amendment types should be selected based on their potential impact on asbestos mobility. Caution should be practiced in extrapolating the results of this bench-scale study to field sites because of the difference in the scale of transport at the field site (Leij et al., 2016), the grain size distribution of soil and asbestos, types of asbestos minerals, and DOM concentration and quality (Lyon et al., 2011).

4. Conclusions

Our results disproved the prevailing assumption that asbestos fibers are immobile in soils and could not move in groundwater in all conditions. We showed that exposure to certain types of DOM at the contaminated sites can increase asbestos mobility. The extent to which DOM increases asbestos mobility depended on the DOM quality: fulvic acid > humic acid > natural organic matter. We surmised that a combination of increased stability of asbestos fiber suspension and decreased interaction between the fiber and soil or sand grain in the presence of DOM could increase the mobility of asbestos fibers through soils.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary material related to this article can be found, in the online version, at doi:<https://doi.org/10.1016/j.hazl.2021.100015>.

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