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Inhalation risks of wind-blown dust from biosolid-applied agricultural lands: Are they enriched with microplastics and PFAS?

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Abstract

Application of wastewater biosolids as a fertilizer on agricultural fields may release pollutants such as microplastics and per- and polyfluoroalkyl substances (PFAS) into the air and pose a serious inhalation risk. This article quantifies the total PFAS and microplastics present in biosolids, discusses their potential transport by wind, and highlights research needs to estimate inhalation risks of PFAS and microplastics in biosolids. Analyzing published data, we found that the dust from biosolid-applied land could be enriched with microplastics and PFAS. Microplastics are more susceptible to suspension by wind than natural soil particles. Future studies should measure microplastics and associated PFAS in dust and biosolids and quantify the exposure risks.

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Keywords

Biosolids, Microplastics, pfas, Wind transport, Inhalation toxicity.

Introduction

Sludge from municipal wastewater treatment plants (WWTPs) is widely applied as biosolids to agricultural fields as part of sustainable use of fertilizers [1,2]. Biosolids are used as fertilizer because they contain a high concentration of nitrogen, phosphorous, organic carbon, and other essential elements, which can have beneficial effects on soil quality and crop production [1,3]. Furthermore, unlike commercial fertilizers, biosolids release nutrients slowly, thereby reducing the eutrophication risk [4]. However, biosolids could also contain toxic pollutants present in wastewater including microplastics [5–7], heavy metals [8], antibiotic resistance genes [9]), organic pollutants [10], and per- and polyfluoroalkyl substances (PFAS) [11]. Among these pollutants, microplastics, and PFAS are very persistent with a half-life exceeding several decades [12–14]. Thus, they are expected to accumulate in soil. In the US, more than 51% of the biosolids are currently applied on land, which may serve as a secondary source of these persistent pollutants. A recent analysis of worldwide data reveals that biosolids applied to agricultural fields can release up to 1080 trillion microplastics into the environment [6]. Microplastics found in wastewater could contain pollutants including PFAS [15]. Application of biosolids could increase particle-bound PFAS such as microplastics with adsorbed PFAS in soil. Thus, preferential emission of these contaminated microplastics by wind, owing to their lighter weight than other soil particles, may lead to enrichment of PFAS and microplastics in the fugitive dust. Climate change (e.g., more frequent droughts, crop failures, and fires), and lack of proper soil conservation measures have rendered agricultural soils highly susceptible to accelerated soil erosion [16,17]. Thus, it is critical to understand the fate, accumulation, and release of PFAS and microplastics in biosolids applied agricultural soils.

The fate of PFAS and microplastics accumulated on biosolid-applied land is unclear. PFAS can adsorb onto organic carbon, metal oxides, and clay minerals in subsurface soil [18,19], can enter crops via root [20–22]. The remaining amount can be washed off by stormwater runoff or infiltrate into the ground [23] if they are not degraded. Some precursors can also be oxidized to produce PFAS in the subsurface [18]. On the other hand, microplastics, owing to their large size, can be filtered from infiltrating water and remain in the topsoil [24]. Thus, these pollutants can be assumed to accumulate in topsoil, where they may pose inhalation risk due to preferential entrainment of lighter particles such as microplastics by the wind. Several studies have confirmed a high concentration of both microplastics and PFAS in dust samples [25,26]. Inhaling

microplastics can cause breathing irritation and oxidative stress in lung tissues, along with general inflammatory responses in airways and bronchi [27]. Chronic exposure can also cause death [28]. Similarly, inhalation of PFAS can cause acute lung toxicity and inhibit lung surfactant function [29]. Thus, future studies should estimate the inhalation risk of both microplastics and PFAS from biosolids applied to land. In this study, we reviewed 27 peer-reviewed journal articles that reported microplastic and PFAS concentrations in biosolids and analyzed 8 articles that reported PFAS concentrations in dust samples to evaluate their transport potential by the wind. Furthermore, we suggest specific areas of research, which can further improve our understanding of the aerial transport of these pollutants.

How much microplastics and PFAS are present in biosolids?

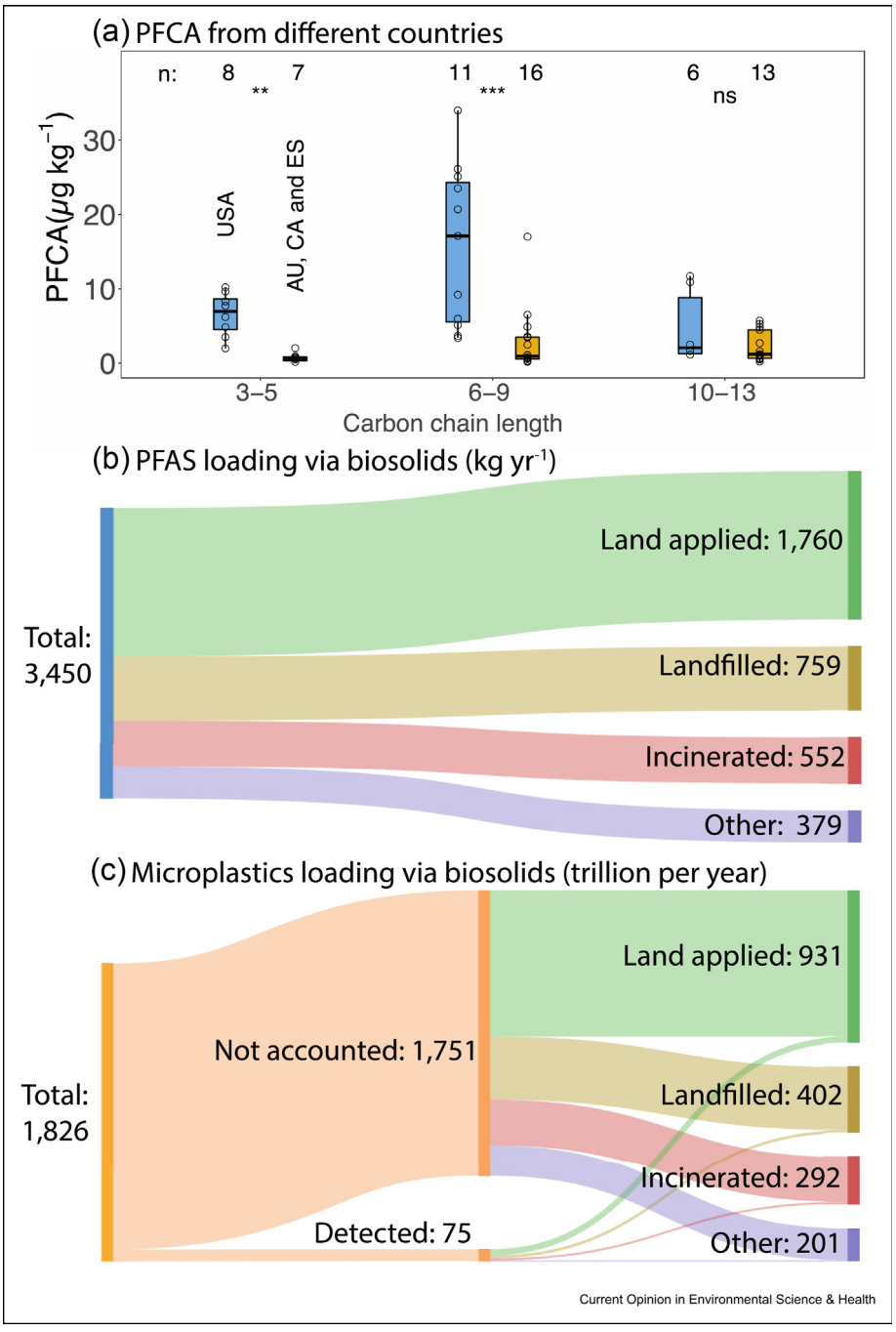
Several studies provided evidence of the presence of microplastics in biosolids and preferential enrichment of microplastics in dust emitted from biosolid-applied land [21,30–32]. Surveying 20 research articles published after 2015, we estimate that microplastic concentration in sludges ranges between 800 and 41,000 pieces per kilogram. However, the common detection methods used to count microplastics in biosolids may underestimate or even exclude many microplastics smaller than 10 μm [6]—the fraction that could pose a greater risk for human and animal health [33–35]. Due to the difficulty in the detection of smaller plastics, the concentration of microplastics in biosolids can be underestimated [6]. Comparing the fraction of total microplastics removed from wastewater to microplastics detected in biosolids, the same study found that microplastics detected in biosolids constitute only 4% of the removed microplastics, indicating most of them in biosolids evade detection potentially due to change in size or surface properties. Of those detected, most particles are of fiber shape indicating a higher concentration of fibers are available for resuspension via wind [24,30,36]. Indeed, a higher concentration of fibers was frequently found in dust samples [37,38], indicating fiber-shaped plastics could also be more susceptible to resuspension by the wind. For instance, based on the orientation or contact points on soil grain, fibers may experience drag or shear force more than smaller fragments if a greater portion of the fiber is dangled into the air from soil surface. However, further study is needed to confirm this hypothesis. Nevertheless, a high concentration of fibers in the dust sample could have other unintended health risks. For instance, the aspect ratio or shape of asbestos, another particulate air pollutant with type I carcinogenicity, plays a critical role in developing lung disease [39,40]. Thus, future studies should examine if the shape of microplastics has any correlation with toxicity in the lungs.

We analyzed 7 studies that measured perfluoroalkyl carboxylic acid (PFCA) concentrations in biosolids from multiple wastewater treatment plants in the US, Canada, Australia, and Spain (list in the Supplementary Material) and observed that biosolids in the US have a significantly higher amount of PFCA than that produced in other countries (Figure 1). This could be due to the continued use of PFAS precursors in domestic and industrial products in the US [41] or more sensitive methods used to detect PFAS in the reported study. Consequently, PFAS concentrations in biosolids have not decreased even after the use of long-chain PFAS was phased out in 2002 [11,42]. A high concentration of PFCA in the USbiosolids indicates a greater risk of PFAS exposure using those biosolids in agriculture than those produced in other reported developed countries. Thus, biosolid applications should be regulated based on the amount of PFAS found in them [42]. A previous study estimated that up to 3450 kg of PFAS may be released into the environment via biosolids each year in the US [11]). Based on the usage of biosolids reported by the US Environmental Protection Agency (EPA), 1760 kg of PFAS could be deposited on land via land application annually, from where they could spread into the environment via stormwater runoff and wind and infiltrate into groundwater [18]. As most PFAS are retained within subsurface or root zone, they can be taken up by plants or crops and enter the food chain [43]. Among all these modes of exposure, exposure via air is a direct threat to human health as there is little to no mitigation measure.

Why do microplastics and PFAS-associated with microplastics pose increased inhalation risk compared to other natural particles?

Many studies have directly measured microplastic concentration suspended in air [38,44,45] and deposited on land [46,47] or trapped on tree canopies [48]. Furthermore, microplastics have been repeatedly found in remote areas and higher elevations far away from their source [49–53], indicating they are highly susceptible to be airborne. A theoretical framework for microplastic transport by wind has been proposed [54]. Briefly, microplastics can be released into the air by direct resuspension or by the disintegration of biosolid aggregates via saltation. As plastics, depending on type and manufacturing process, are 30–60% lighter than natural soil minerals, microplastics are more likely to be entrained up by wind. Interparticle forces between plastic particles and soil grain and soil moisture content could all play a critical role in determining the number of microplastics emitted from biosolid-applied land under given conditions. Therefore, future studies should evaluate how microplastic type, shape, and biosolid characteristics such as moisture content could affect the resuspension of microplastics by the wind.

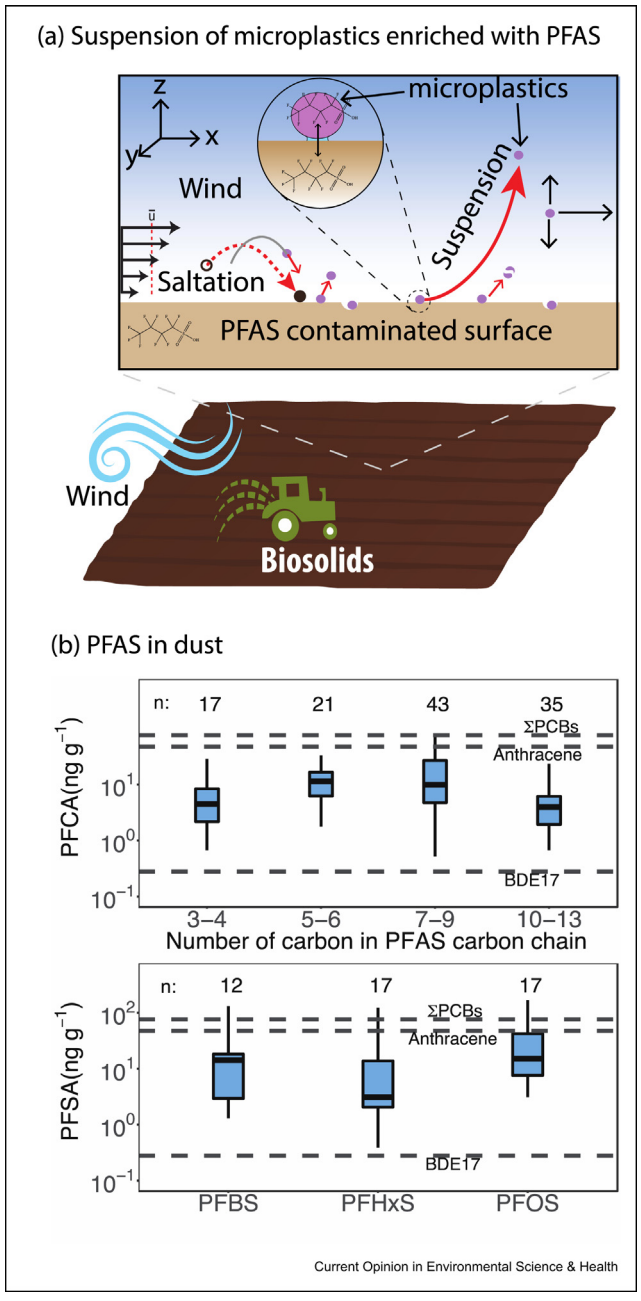
Figure 1



(**a**) Perfluorocarboxylic acid concentrations in biosolids from the USA and other countries (Australia, Canada, and Spain). '**' and '***' symbols indicate a *p-value* less than 0.01 and 0.001, respectively. ns denotes non-significant. (**b**) The total amount of PFAS deposited on land via biosolid application. (**c**) amount of PFAS released into the environment via biosolids, of which only a small fraction of them is detected, and the rest of them are too small (<10 μm) to detect using current technologies, therefore are not accounted for.

As most PFAS are non-volatile, with an exception of volatile precursors, PFAS concentration in the air could be dominated by PFAS associated with dust or microplastics. Analyzing eight studies that measured the PFAS concentration in dust samples from indoor environments, we show that a significant fraction of total PFAS measured in the air are associated with dust, aerosol, and other particulate matter, and the long-chained PFAS are enriched in the dust (Figure 2b). Our analysis also reveals that an increase in PFAS

Figure 2



(**a**) Mechanism of microplastic emission from the land where biosolids are applied. (**b**) PFAS concentrations in dust emitted from land-based on 8 studies (Supplementary Material). The dashed lines refer to concentrations of ΣPCB, Anthracene and BDE-17 reported in the literature [56,57].

chain length increases their association with dust. Increased affinity of long-chained PFAS with solid surfaces explained this observation [55]. The PFAS concentrations observed in these studies are comparable to other legacy organic pollutants typically found in the dust (Figure 2b): polycyclic aromatic hydrocarbons (PAH), polybrominated diphenyl ethers (PBDE), and polychlorinated biphenyls (PCBs) [56,57]. The analysis indicates that PFAS has a similar potential of aerial transport as other organic pollutants, even though PFAS exposure via inhalation did not receive a similar level of attention as other organic pollutants.

Biosolids can also release fine particles or colloids when subjected to natural drying and freeze–thaw cycles, which can carry PFAS to subsurface and groundwater at a concentration higher than the EPA advisory limits. Previous laboratory studies [58,59] show that the presence of colloids released by these natural cycles can increase the PFAS concentration in water samples by 3–15 times. The same colloids can be suspended by the wind. Thus, microplastics in biosolids can be enriched with long-chained PFAS, similar to the other organic or inorganic particles or colloids. However, PFAS concentration on microplastics in biosolids is rarely measured. Furthermore, most airborne particles have a size of less than 1 μm, and microplastics of size less than 10 μm are difficult to detect due to the limitation of the optical microscope. Thus, future studies should examine the concentration of nano or submicron size plastic particles in dust samples and the concentration of attached PFAS on them.

Needs for future research

Analyzing eight studies that measured the PFAS concentration in dust samples from indoor environments, we show that a significant fraction of total PFAS measured in the air is associated with dust, aerosol, and other particulate matters, and the long-chained PFAS are concentrated in the dust. Analysis of the reported studies confirmed that the dust released from biosolids could be enriched with microplastics and PFAS. Therefore, the application of biosolids may increase the inhalation risks of microplastics and PFAS. Thus, future studies should quantify the concentration of both pollutants in dust emitted from biosolid-applied land. Detection of smaller microplastics (<10 μm) in biosolids is challenging, and the concentration could vary by orders of magnitude based on sample collection method, sample volume, filter cutoff size used to isolate plastic particles from water, organic digestion method, density stratification solution, counting methodology, and method detection limit [6]. Estimating microplastics in biosolids often require complex sample preparation procedures and the use of expensive microscopes, thereby limiting the usability of the method in the agriculture field. Thus, it is critical to develop a simple, rapid method of isolating and quantifying microplastics from environmental samples. Microplastics obtained from field studies are shown to have significantly higher PFAS concentrations than those measured in laboratory settings [60]. The result indicates that aging could affect PFAS adsorption on microplastics. Future studies should examine PFAS adsorption and desorption on

aged and fresh plastic surfaces to understand the fate of adsorbed PFAS on microplastics. Microplastics with PFAS could pose higher risks than uncontaminated microplastics. Thus, the lung toxicity of microplastics should be evaluated as a function of adsorbed PFAS in addition to the shape of microplastics, and the age of microplastics.

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 article.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.coesh.2021.100309.

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** of outstanding interest

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This study estimated PFAS accumulation by microplastics in the laboratory and the field and proved that microplastics in the environment can accumulate significantly higher PFAS concentrations than microplastics in the lab.