State of the science and meta-analysis of crop uptake of per- and polyfluoroalkyl substances (PFAS)

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Introduction

Per- and polyfluoroalkyl substances (PFASs) comprise a group of synthetic chemicals that are detected in a wide variety of commercial and industrial products (e.g., cleaners, textiles, paints, fire-fighting foams). PFASs have been detected in the environment worldwide, due to their widespread use and persistence (EPA 2016). In 2002, 3M stopped production of perfluorooctane sulfonic acid (PFOS) and perfluorooctanoic acid (PFOA); since that time, other companies pledged to decrease production and use of PFASs, including perfluorooctanoic acid (PFOA) (3M 2017; DuPont 2017; EPA 2007, 2017). Despite efforts toward reduction in the release of PFASs, they continue to be detected in air (Barber et al. 2007; Harada et al. 2005b, 2006; Kim and Kannan 2007), dust (Kubwabo et al. 2005; Moriwaki et al. 2003), surface water (Boulanger et al. 2004; Kannan et al. 2005; Kim and Kannan 2007; Nakayama et al. 2007; Simcik and Dorweiler 2005; Sinclair et al. 2006), groundwater (Xiao et al. 2015), soil (Xiao et al. 2015), and sediment (Zareitalabad et al. 2013), as well as in the blood of the general U.S. population in 1999-2000 and 2003-2004 (Calafat et al. 2006b, 2007a, 2007b; De Silva and Mabury 2006; Kuklenyik et al. 2004; Olsen et al. 2003a, 2003b, 2004c, 2005, 2007a). More recent biomonitoring data indicate widespread human exposure, but levels of PFOS and PFOA appear to be declining (Olsen et al. 2008; Kato et al. 2011). However, levels of other PFASs, such as perfluorononanoic acid (PFNA), appear to be increasing (Kato et al. 2011). The major human exposure pathways for PFASs are not yet fully understood, though it is expected that dietary intake is the primary source of exposure for most people (ATSDR 2015; EPA 2016; Domingo 2012; Domingo and Nadal 2017).

Exposures to PFASs through the consumption of vegetables has been hypothesized to be a non-negligible source of PFASs intake (Herzke et al. 2013; Domingo 2012; Halldorsson et al. 2012; Braun et al. 2016; Vieira et al. 2013). Agricultural soil PFAS input sources may vary, but are generally thought to originate from biosolids (Lu et al. 2012), aerial deposition (Stemmler and Lammel 2010; Wallington et al. 2006; Paustenbach et al. 2006), and irrigation water (Blaine et al. 2014). Crop uptake of PFASs has been reported in corn, wheat, potato, oats, carrots, and cucumbers that were grown in PFASs-spiked soils (Blaine et al. 2013; Stahl et al. 2009; Lechner and Knapp 2011; Yoo et al. 2011). Yoo et al. reported a transfer of PFASs to grass from industrially contaminated biosolid-amended soils (Yoo et al. 2011). To our knowledge, there are no published reports of crop uptake of PFASs using municipal biosolids with detected PFAS.

Thus, the objective of this study is to provide a state-of-the science review on PFAS uptake in plants intended for human and animal consumption and to conduct a screening level human health risk assessment for consumption of PFAS based on the following: i) in which crops PFASs have been detected, ii) at what concentrations are PFASs detected, iii) which PFASs have been previously detected in crops, and iv) where in the plant PFASs tend to accumulate, if at all. We describe known sources of PFASs deposition in soil and anticipated source deposition trends, based on commercial use patterns.

Materials and methods

A literature search was performed based on the Boolean search, "PFOA OR PFOS OR PFAS OR "perfluoroalkyl" AND plant OR crop OR uptake factor OR bioaccumulation factor. A total of 288 records were identified. Of these, nine studies were selected for evaluation (Blaine et al. 2013, 2014; Yoo et al. 2011; Stahl et al. 2008; Krippner et al. 2015; Zhao et al. 2013; Lee et al. 2013; Gottschall et al. 2016; Lechner et al. 2011). These studies were selected based on i) the reported concentration of PFOS or PFOA in a crop; ii) reported non-detect of PFOS or PFOA in a crop and the reported limit of detection; and iii) the reported soil concentration of PFOA or PFOS measured from the same soil as where the crops were grown. One study was excluded, as maize was grown in solution to measure uptake kinetics and plant tissue concentration was not presented at the 1 mg/L exposure level (Wen et al. 2013).

Results and discussion

We captured over 140 data points, of which approximately 50% were comprised of human edible plants or crops intended for livestock consumption. Over 85% of all data points were for either shoots (e.g. celery shoot, wheat shoot) or aerial plant components (e.g. Bermuda grass, corn grain, corn stover, Kentucky blue grass, lettuce leaves, maize kernel, maize straw, maize ears, oat grain, oat straw, pea fruit, perennial wheatgrass, pumpkin stalk, pumpkin flower, pumpkin leaf, pumpkin fruit, tall fescue, tomato fruit, wheat grain, and wheat straw). The remainder were components below ground surface (e.g. potato tuber, potato peel, radish root, pumpkin root, wheat root). Bioaccumulation factors (BAFs), defined as the PFAS concentration in plant divided by the PFAS concentration in soil, for the current data set ranged from 0 to 6.20 for PFOA and 0 to 0.98 for PFOS.

Limited data were available for other, shorter carbon chain PFASs, with the exception of PFHxA (perfluorohexanoic acid and PFHxS (perfluorohexanesulfonic acid). BAF for PFHxA and PFHxS ranged from 0.12 - >10.0 (median 2.47) and 0.006 to 2.9 (median 0.40), respectively. PFHxS has an elimination half-life in humans of several years and has been widely detected in serum samples in the United States and Australia (Calafat et al. 2007; Kärrman et al. 2006). Across all PFASs of variable carbon chain length, serum concentrations of many PFAS have decreased since 2000; however, PFNA (perfluorononanoic acid) serum concentrations have inexplicably increased (Anderson et al. 2008). No studies were located that reported plant concentrations of PFNA or BAFs.

Our analyses showed that PFASs accumulate in crops for human or livestock consumption. However, the BAFs represent a wide range that does not seem to follow a linear dose response. Field site- and chemical-specific physico-chemical factors undoubtedly play a large role in the resulting BAFs. Collectively, these results indicate that the long-term use of PFAS-containing irrigation or biosolids as a soil amendment may result in human intake of PFASs through consumption of crops or consumption of meat or milk fed to livestock who consumes these crops. Further studies will investigate the implications for human health risk as a result of consuming crops, meat, or milk that contain PFASs.

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