Volatilization Potential of Per- and Poly-fluoroalkyl Substances from Airfield Pavements and during Recycling of Asphalt

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Abstract: Per- and poly-fluoroalkyl substances (PFAS) in water are typically present in their ionic (nonvolatile) forms; however, these can transition to their nonionic (volatile) forms when in contact with organic solvents and organic matrices. In particular, when PFAS are dissolved in organic solvents such as residues left from firefighting foams, fuels, and bitumen present in asphalt, the equilibrium between ionic and nonionic forms can trend toward more volatile nonionic forms of PFAS. We assessed the volatility of common PFAS based on calculated and available experimental data across ambient temperature ranges experienced by airfield pavements and at elevated temperatures associated with reworking asphalts for reuse. Volatilities are shown to be comparable to hydrocarbons in the semivolatile range, suggesting that volatilization is a viable loss mechanism for some PFAS that are nonvolatile in water. The present study points to future investigative needs for this unexplored mass loss mechanism and potential exposure pathway. Environ Toxicol Chem 2022;41:2202–2208. © 2022 Commonwealth of Australia. Environmental Toxicology and Chemistry published by Wiley Periodicals LLC on behalf of SETAC.

Keywords: Perfluoroalkyl substance; Air pollution; Atmospheric transport; Fate and transport; Hazardous waste sites

INTRODUCTION

Per- and poly-fluoroalkyl substances (PFAS) are widespread in the environment and thought to pose threats to human and ecological health (Giesy & Kannan, 2001). Implications of PFAS contamination can include negative effects on fetal development, learning and behavior in children, adult fertility, hormonal balance, and liver function (National Toxicology Program, 2016). High concentrations of PFAS were present in in aqueous film-forming foams (AFFFs) used in periodic firefighting training for more than four decades. There has been a strong focus on assessing PFAS partitioning into groundwater and soil to assist in defining such risks (Davis et al., 2021; Li et al., 2018), with some investigation undertaken on the volatility of PFAS (Roth et al., 2020) as a human health risk pathway. Little has been done to investigate PFAS mass losses due to volatilization, with only a limited number of PFAS volatilization (Sima & Jaffé, 2021) or thermal desorption (Longendyke et al., 2022) studies in the literature and none to the knowledge of the authors that assess volatilization losses from residual solvents present in both firefighting foams and bitumen in airport asphalts.

With airports across the world previously using AFFF for historic firefighting and training activities, there is potential for airport runways and tarmacs to have been in contact with PFAS through the use of AFFF as well as waste material from the maintenance of tarmacs (White & Boston, 2019). Many of the estimated 400 airports in Australia likely have potential PFAS contamination based on AFFF use for fire suppression in the period 1960–2004. In Canada, an investigation of 2071 airport and heliport sites, using a decision tree approach, found that 7% of these sites were deemed to have likely PFAS contamination, with another 13% potentially impacted (Milley et al., 2018). In the United States, 172 PFAS-contaminated sites have been identified at military bases in 40 states (EWG Environmental Working Group, 2020; PFAS Project Lab, 2021). Based on these examples, it is likely that many of the world’s airport runways and tarmacs have been exposed to and possibly contaminated with PFAS. Airport runways have an average surface life of approximately 10 years, and resurfacing requires 8000–24,000 tonnes of new asphalt, constituting a significant volume of waste asphalt to be recycled or landfilled. The reuse of asphalts and repairs of runways contaminated with PFAS...
AFFF formulations will have a differing composition compared to historically used AFF (Houtz et al., 2013). We used common PFAS generally considered to be nonvolatile in water to estimate indicative volatilization behaviors and mass partitioning characteristics.

**METHODS**

We assessed the potential volatilization of PFAS from pavements contained in organic residues as well as the volatilization of PFAS-contaminated RAP when recycled in hot asphalt mixes. The vapor pressure of some selected anionic PFAS (perfluorohexanoic acid [PFHxA], perfluoroheptanoic acid [PFHpA], perfluorooctanoic acid [PFOA], perfluorononanoic acid [PFNA], perfluorodecanoic acid [PFDeA], perfluoroundecanoic acid [PFUnA], perfluorododecanoic acid [PFDoA], perfluorobutane sulfonic acid [PFBS], perfluorohexane sulfonic acid [PFHxS], perfluorooctanoate sulfonic acid [PFOS], and perfluorooctane sulfonamide [PFOSA]; Table 1) commonly found at airports (Milley et al., 2018) and selected hydrocarbons (undecane and tetradecane; Table 1) at various temperatures (20–200 °C) were estimated using the Clausius-Clapeyron equation:

\[
\ln\left(\frac{P_2}{P_1}\right) = \frac{\Delta H_{\text{vap}}}{R} \times \left[\frac{1}{T_2} - \frac{1}{T_1}\right]
\]

(1)

In Equation 1, \(\Delta H_{\text{vap}}\) is the enthalpy of vaporization (joules per mole), \(R\) is the universal gas constant (8.3145 J mol\(^{-1}\) K\(^{-1}\)), \(P_1\) and \(P_2\) are the vapor pressures at two temperatures, and \(T_1\) and \(T_2\) are the corresponding temperatures (Kelvin). The Clausius-Clapeyron equation was used to estimate vapor pressures of pure PFAS and a common method (as well as methods based on the Clausius-Clapeyron equation) used to estimate vapor pressures of pure substances with limitations (Olsen & Nielsen, 2001). Other methods to estimate vapor pressures were not investigated because comparison of the calculated vapor pressures using the Clausius-Clapeyron equation and experimental vapor pressures for the PFAS used in the present study over the temperature ranges investigated were in good agreement for the broad purposes of the study.

The enthalpy of vaporization was not available in most instances and was calculated using experimentally available vapor pressures and boiling points with Equation 1. Parameters used to calculate vapor pressures for the selected PFAS and the resulting PFAS concentrations in bitumen that would result from asphalt contaminated with PFAS are shown in Table 1 as well as undecane and tetradecane.

Concentrations of PFAS in the gas phase at equilibrium with the pure PFAS as well as PFAS present as a contaminant in hot asphalt mixes were calculated using the ideal gas law:

\[
P V = n R T
\]

(2)

In Equation 2, \(P\) is the gas pressure, \(V\) is the gas volume, \(n\) is the number of moles, \(T\) is the temperature (Kelvin), and \(R\) is the universal gas constant (0.0821 atm L mole\(^{-1}\) K\(^{-1}\)). Rearranging the formula \((nV = P/R)\) allowed concentration to be calculated in the gas phase as moles per liter.
| Selected PFAS and hydrocarbons | Abbreviation | Molar mass (g/mole) | pK_a ranges in water | Boiling point (°C) | Experimental (calculated) vapor pressures (kPa) | Enthalpy of vaporization (J/mole) | PFAS mole fraction in recycled asphaltb-d |
|-------------------------------|--------------|--------------------|----------------------|-------------------|-----------------------------------------------|---------------------------------|---------------------------------------------|
| Perfluorohexanoic acid        | PFHxA        | 314.1°             | -0.16 to <1.6f       | 157°              | 0.00825 (0.00825) 11.55°C, 101.325 (101.325) 157 °C | 65,914h                        | 6.08 × 10⁻⁸                     |
| Perfluoroheptanoic acid       | PFHpA        | 364.1°             | -0.19 to <1.6f       | 176°              | 0.0177 (0.01773) 25 °C, 101.325 (101.325) 176 °C | 63,786h                        | 5.24 × 10⁻⁸                     |
| Perfluorooctanoic acid        | PFOA         | 414.07°            | -0.5 to 3.8°         | 190.5°            | 0.128 (0.185) 59.25°C, 1,602 (1.602) 94.9 °C, 2.69 (2.59) 103.94 °C, 22.2 (20.7) 148.6 °C, 96.5 (102) 190.8 °C | 61,547h                         | 4.61 × 10⁻⁸                     |
| Perfluorononanoic acid        | PFNA         | 464.08°            | -0.21 to <1.6f       | 203.12°           | 1.22 (1.12) 99.63 °C, 3,120 (3.12) 117.66 °C, 15.2 (14.2) 150.71 °C, 101 (101) 203.12 °C | 63,027h                        | 3.82 × 10⁻⁸                     |
| Perfluorodecanoic acid        | PFDeA        | 514.1°             | -0.21 to 2.6°        | 218.88°           | 3.129 (3.13) 129.56 °C, 62.1 (59.9) 202.93 °C, 101 (100) 218.88 °C | 64,142h                        | 3.71 × 10⁻⁸                     |
| Perfluoroundecanoic acid      | PFUnA        | 564.1°             | -0.21 to 2.6°        | 237.65°           | 0.616 (0.616) 112.04 °C, 4.51 (3.83) 149.22 °C, 34.5 (29.9) 200.68 °C, 101.3 (101) 237.65 °C | 66,457h                        | 3.38 × 10⁻⁸                     |
| Perfluorododecanoic acid      | PFDoA        | 614.1°             | -0.21 to 3.1°        | 247.36°           | 0.856 (0.856) 127.58 °C, 2.99 (2.52) 149.6 °C, 2.99 (2.52) 149.6 °C, 22.07 (18.6) 197.44 °C, 101.3 (101) 247.36 °C | 69,118h                        | 3.11 × 10⁻⁸                     |
| Perfluorobutane sulfonic acid | PFBS         | 300.1°             | 0.14—<0.3f           | 211°              | 0.00357 (0.00357) 25 °C, 101.325 (101.325) 211 °C | 66,155h                        | 6.36 × 10⁻⁸                     |
| Perfluorohexane sulfonic acid | PFoS         | 400.12°            | 0.14—<0.3f           | 238.5°            | 0.000613 (0.00061) 25 °C, 101.325 (101.325) 238.5 °C | 71,377h                        | 4.77 × 10⁻⁸                     |
| Perfluorooctane sulfonic acid | PFOS         | 500.13°            | 0.14—<1f             | 249°              | 0.000266644 (0.00030) 25 °C, 0.800 (0.800) 133 °C, 101.325 (101.325) 249 °C | 73,593h                        | 3.82 × 10⁻⁸                     |
| Perfluorooctane sulfonamide   | PFOSA        | 499.15°            | 6.24—6.522          | 210°             | 0.012 (25 °C), 101.325 (101.325) 210 °C | 58,533h                        | 3.82 × 10⁻⁸                     |
| Undecane                      | nC11         | 156.13°            | —                    | 195.9°            | 0.055 (0.055) 25 °C, 101.325 (101.325) 195.9 °C | 41,910h                        | 1.22 × 10⁻⁷                     |
| Tetradecane                   | nC14         | 198.39°            | —                    | 253.535°          | 0.002 (0.00039) 25 °C, 101.325 (101.325) 253.535 °C | 71,300h                        | 9.62 × 10⁻⁸                     |

 abnormal; t normal; c calculated; s standard.

- Selected PFAS and hydrocarbons
- Abbreviation
- Molar mass (g/mole)
- pK_a ranges in water
- Boiling point (°C)
- Experimental (calculated) vapor pressures (kPa)
- Enthalpy of vaporization (J/mole)
- PFAS mole fraction in recycled asphalt

*Average of boiling points.
*Based on 0.001 mg/kg of PFAS in recycled asphalt resulting from using 20% reclaimed asphalt pavement contaminated with PFAS at 0.005 mg/kg.
*Bitumen average molecular weight of 1050 g/mole (Lesueur, 2009).
*National Center for Biotechnology Information (2021).
*Interstate Technology Regulatory Council (2021).
*Zhang et al. (2020).
*Calculated using the Clapeyron equation from experimental vapor pressures at various temperatures (in bold) and boiling point vapor pressure (101.325 kPa).
*Steele et al. (2002).
*Kaiser et al. (2005).
*Predicted boiling point.
*Bhatarai and Gramatica (2011).

pK_a = dissociation constant.
To calculate the equilibrium gas-phase concentrations of PFAS and hydrocarbons that would partition from the bitumen component of the asphalt, and assuming the aggregate in the asphalt does not impact on partitioning, Raoult’s law was used:

\[ P_{\text{PFAS}} = X_{\text{PFAS}} \times P^o_{\text{pure PFAS}} \]  

In Equation 3, \( P_{\text{PFAS}} \) is the vapor pressure of the PFAS compound based on its mole fraction in the solvent, \( X_{\text{PFAS}} \) is the mole fraction of PFAS compound in the solvent, and \( P^o_{\text{pure PFAS}} \) is the vapor pressure of the pure PFAS compound.

Estimated equilibrium gas-phase concentrations were calculated for pure individual PFAS in their nonionic forms and for two pure semivolatile hydrocarbons (undecane and tetradecane) over a temperature range (20–60 °C). Estimated equilibrium gas-phase concentrations were calculated for PFAS-contaminated recycled asphalt (0.001 mg/kg for individual PFAS components based on containing 20% asphalt contaminated with PFAS at 0.005 mg/kg) over a temperature range used to prepare bitumen hot mixes (120–200 °C; Shell Bitumen, 2015). The use of a concentration of 0.001 mg/kg for individual PFAS compounds in the recycled asphalt was based on the lowest PFAS reuse screening levels for soils in Victoria, Australia (Environmental Protection Authority Victoria, 2019). Bitumen (the organic solvent in the asphalt) is a black viscous petroleum product remaining after the distillation/refining of crude oil at temperatures of 350–380 °C (Shell Bitumen, 2015). Conventional bitumen used for roads typically softens at 45–60 °C. Bitumen is used to bind the aggregate in asphalt as well as to provide flexibility to asphalt paving (Shell Bitumen, 2015).

RESULTS AND DISCUSSION

The volatilization of PFAS components is dependent on the abundance of its nonionic and ionic forms, and the forms present can be affected by the solvent in which the PFAS is dissolved. Solvents are generally classified by their polarity based on their dielectric constants as polar >50, semipolar 20–50, or nonpolar 1–20. Water is the most polar solvent, with a dielectric constant of 78.54; and in water PFAS like perfluorocarboxylic and perfluorosulfonic acids are generally present as moderate to strong acids in their nonvolatile ionic forms (Interstate Technology Regulatory Council, 2021; Vieker et al., 2013). In contrast, perfluorosulfonamides would be present as a mixture of nonvolatile ionic forms and volatile nonionic forms. However, when dissolved in less polar organic solvents, the proportion of nonionic PFAS present can increase to the extent where nonionic PFAS can volatilize. Examples of dissociation constants (pKₐ) in organic solvents for the PFAS investigated in the present study were not available. Smaller, more acidic PFAS such as trifluoroacetic acid and trifluoromethane sulfonic acid with low pKₐ values in water have much higher pKₐ values in acetonitrile, a semipolar solvent with a dielectric constant of 36 (Kütt et al., 2018). Trifluoroacetic acid and trifluoromethane sulfonic acid in water have low pKₐ values of 0.5 (Serjeant & Dempsey, 1979) and −5.9 to −14 (Gutowski & Dixon, 2006), respectively; and in acetonitrile there is a large increase in pKₐ values of 12.65 and 2.6 (Eckert et al., 2009), respectively. In the case of trifluoroacetic acid, 100% would be present as the volatile nonionic form in acetonitrile. Nonpolar organic solvents, such as ethylene glycol butyl ether (dielectric constant of 9.87 [Sengwa et al., 2006] because no dielectric data were available for diethylene glycol butyl ether) and bitumen (dielectric constant of 2.7 [Shell Bitumen, 2015]) would likely result in larger increases in pKₐ than semipolar solvents such as acetonitrile. Literature pKₐ values for PFAS in water (Table 1) show a range of values for perfluorocarboxylic acids that is significantly larger compared to other PFAS types. Perfluorocarboxylic acids in water have been reported to increase in pKₐ values with increasing chain length (C4–C11; Moroi et al., 2001). This suggests that the perfluorocarboxylic acids considered in the present study (≥C8 carbon chain lengths) will likely have pKₐ values greater than that of trifluoroacetic acid. As such, the PFAS in the present study have pKₐ values in water (Table 1) high enough to be present mainly in their volatile nonionic forms in nonpolar solvents such as diethylene glycol butyl ethers, bitumen, and petroleum.

Calculated and experimental PFAS equilibrium gas-phase concentrations over a temperature range of 20–60 °C are shown in Figure 2. These are based on experimental (literature) and calculated vapor pressures and show good agreement. Equilibrium concentrations of PFAS in the gas phase ranged from 37 mg/m³ (PFOS) to 2371 mg/m³ (PFHxA) at 20 °C and from 1218 mg/m³ (PFOS) to 53,650 mg/m³ (PFHxA) at 60 °C. In comparison, the hydrocarbon equilibrium concentrations in the gas phase ranged from 19 mg/m³ (tetradecane) to 10,282 mg/m³ (undecane) at 20 °C and from 566 mg/m³ (tetradecane) to 71,308 mg/m³ (undecane) at 60 °C. This would suggest that the hydrocarbons (undecane and tetradecane) and the selected PFAS (in their nonionic forms) would likely be rapidly lost to volatilization on pavements at ambient temperatures. The semivolatile hydrocarbons undecane and tetradecane were included in the present study to represent volatile compounds with volatilities that encompass those of the selected PFAS (Figure 2). This implies that the volatility of the selected PFAS in their nonionic forms and the semivolatile hydrocarbons undecane and tetradecane may have comparable losses to volatilization when exposed on airfield pavements (dependent on mol fractions). Undecane and tetradecane, which are kerosene-range hydrocarbons, would be readily lost to volatilization at ambient temperatures over short time periods based on kerosene-range hydrocarbon losses (Fingas, 1997), especially if exposed as thin films on surfaces such as pavements.

Possible scenarios for volatilization of PFAS could be from pavements where PFAS components are taken up into organic solvents originally contained in some AFFFs such as diethylene glycol n-butyl ether. On evaporation of water over a period of days from the diluted AFFF, PFAS could be concentrated in high–boiling point organic solvent residues. Such a process can lead to more nonionic PFAS being available for volatilization as a result of less dissociation when present in organic solvents (Kütt et al., 2018).
Estimated equilibrium gas-phase concentrations of PFAS associated with new hot asphalt mixes up to 200 °C (Shell Bitumen, 2015) containing RAP (20%) contaminated with PFAS (0.005 mg/kg) are shown in Figure 3. For example, calculations for PFAS partitioning from PFAS-contaminated hot mix asphalt (at 200 °C) indicate PFAS equilibrium gas-phase concentrations ranging from 0.0849 mg/m³ (PFOS) to 2.62 mg/m³ (PFHxA) and hydrocarbon equilibrium gas-phase concentrations (Undecane and tetradecane).

**Figure 2:** Plot of equilibrium gas-phase concentrations (based on calculated vapor pressure data) versus temperature (ambient temperature range 20–60 °C) for selected pure PFAS and hydrocarbons. Equilibrium gas-phase concentrations calculated from the Clausius-Clapeyron equation, Raoult’s law, and experimental vapor pressures obtained from the literature (Table 1). PFAS = per- and poly-fluoroalkyl substances; PFHxA = perfluorohexanoic acid; PFHpA = perfluorooctanoic acid; PFNA = perfluorononanoic acid; PFDeA = perfluorodecanoic acid; PFUnA = perfluoroundecanoic acid; PFDoA = perfluorododecanoic acid; PFBS = perfluorobutane sulfonic acid; PFHxS = perfluorohexane sulfonic acid; PFOS = perfluorooctane sulfonic acid; PFOSA = perfluorooctane sulfonamide; nC11 = undecane; nC14 = tetradecane.

**Figure 3:** Plot of equilibrium gas-phase concentrations (based on calculated vapor pressure data) versus temperature (20–200 °C) for PFAS-contaminated recycled asphalt (0.001 mg/kg) using hot asphalt mixes (120–200 °C). Equilibrium gas-phase concentrations calculated from the Clausius-Clapeyron equation, Raoult’s law, and experimental vapor pressures obtained from the literature (Table 1). PFAS = per- and poly-fluoroalkyl substances; PFHxA = perfluorohexanoic acid; PFHpA = perfluorooctanoic acid; PFNA = perfluorononanoic acid; PFDeA = perfluorodecanoic acid; PFUnA = perfluoroundecanoic acid; PFDoA = perfluorododecanoic acid; PFBS = perfluorobutane sulfonic acid; PFHxS = perfluorohexane sulfonic acid; PFOS = perfluorooctane sulfonic acid; PFOSA = perfluorooctane sulfonamide; nC11 = undecane; nC14 = tetradecane.
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Concentrations ranging from 0.0778 mg/m³ (tetradecane) to 0.539 mg/m³ (undecane). Therefore, the hydrocarbons (undecane and tetradecane) and the selected PFAS would likely be rapidly lost to volatilization from hot asphalt. This assumes that the PFAS are present in their nonionic forms once taken up in the hot bitumen, with the hot bitumen behavior akin to that of a nonpolar organic solvent. Importantly, considerable reductions in bitumen viscosity of approximately two orders of magnitude occur from both 20 to 60 °C and from 60 to 200 °C, respectively (Cardone et al., 2014). This may facilitate increased transfer of PFAS into the hot bitumen during recycling and bitumen in asphalt pavements under ambient conditions. Thus, PFAS present in a hot asphalt mix could volatilize when heated at temperatures used to prepare and lay asphalt (120–200 °C; Shell Bitumen, 2015).

The common PFAS assessed in the present study have volatilities (when present in organic solvents and in their nonionic forms) comparable to a narrow range of semivolatile hydrocarbons. Most of the PFAS in the present study had volatilities within a narrow range of semivolatile (undecane and tetradecane) hydrocarbons and would likely be completely removed as a result of volatilization processes over short time periods based on assessments of kerosene-range hydrocarbon losses (Fingas, 1997). This suggests that any remaining AFFF (and associated PFAS) on pavements that were not washed off (spillover, cleaning, or rainfall) or that penetrated the pavements where it was used or PFAS present in hot asphalt mixes would result in complete loss of PFAS to the environment via volatilization (depending on the chemical and physical properties of the individual PFAS and their mole fraction).

**CONCLUSION**

A range of common PFAS (PFHxA, PFHpA, PFOA, PFNA, PFDeA, PFUnA, PFDoA, PFBS, PFHxS, PFOS, and PFOSA) encompassing anionic, cationic, and zwitterionic forms have been assessed for mass losses due to volatilization when present in their nonionic forms in organic solvents resulting from AFFF usage. The assessment indicates that the selected PFAS would be volatile when in their nonionic forms in organic solvents, such as petroleum fuels, diethylene glycol butyl ethers (present in AFFF), and bitumen (present in asphalt). This could be either as thin AFFF films on pavements, as bitumen in asphalt pavements, and during heating when recycled in new asphalt. Evaporation over time of the water present in AFFF on pavements will increase the organic solvent content originally present in AFFF and increase the proportion of nonionic PFAS available for volatilization. Volatilization of PFAS can be significant in some circumstances and may be comparable to semivolatile hydrocarbons found in kerosene or Jet Fuel A1. This has implications for release to the air under ambient temperatures and heated asphalt conditions, which has the potential to reduce PFAS mass in asphalt, but may create greater potential exposures to the atmosphere. This assessment focused on common PFAS where experimental data and inferred data are more readily available and will differ depending on new and historically applied AFFF formulations. Health implications across the broad spectrum of PFAS are dependent on a broader suite of PFAS being investigated and a weighting across individual PFAS seen as hazardous when potentially released to the environment (National Toxicology Program, 2016).

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**Author Contributions Statement**—Trevor Bastow, Greg Davis, Grant Douglas: Conceptualization, Methodology; Writing—reviewing & editing.

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