Journal of Hazardous Materials 466 (2024) 133627

Contents lists available at ScienceDirect

Journal of Hazardous Materials

journal homepage: www.elsevier.com/locate/jhazmat

Distribution and release of PFAS from AFFF-impacted asphalt: How does it compare to concrete?

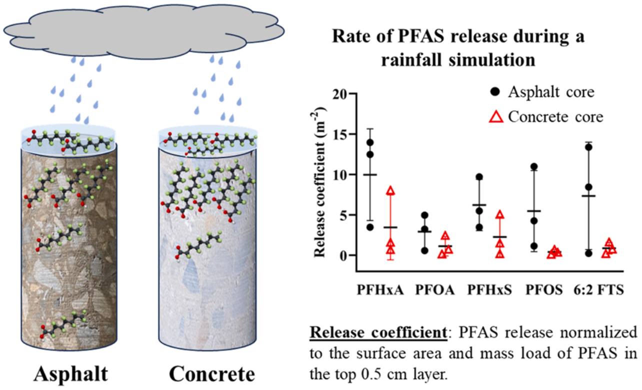
Yijing Li[a], Trent A. Key[b], Phong H.N. Vo[a,c], Scott Porman[d], Anita Thapalia[e], Jeffrey T. McDonough[f], Stephanie Fiorenza[g], Craig M. Barnes[h], Jochen F. Mueller[a], Phong K. Thai[a,*]

[a] Queensland Alliance for Environmental Health Sciences (QAEHS), The University of Queensland, QLD 4102, Australia
[b] ExxonMobil Biomedical Sciences Inc., Spring, TX 77389, USA
[c] Climate Change Cluster, Faculty of Science, University of Technology Sydney, 15 Broadway, Ultimo, NSW 2007, Australia
[d] Mobil Oil Australia, Melbourne, VIC 3008, Australia
[e] ExxonMobil Environmental and Property Solutions Company, Spring, TX 77389, USA
[f] ERM, Denver, CO 80202, USA
[g] Arcadis North America, Houston, TX 77042, USA
[h] Airservices Australia, 25 Constitution Avenue, Canberra, ACT 2601, Australia

HIGHLIGHTS

- First detail investigation of PFAS distribution and release from asphalt.
- Similar distribution profiles of PFAS in asphalt and concrete cores.
- 6:2 FTS and PFOS were predominant in impacted asphalt and concrete cores.
- Asphalt had a higher release rate of PFAS than concrete included in this study.
- Impacted asphalt could be a secondary source of PFAS into the environment.

GRAPHICAL ABSTRACT



ARTICLE INFO

Keywords:
PFAS
Concrete
Asphalt
Runoff
Release rate
Leaching

ABSTRACT

Aqueous film forming foam (AFFF)-impacted asphalt and concrete may serve as potential secondary sources of per- and polyfluoroalkyl substances (PFAS) to the environment through surficial leaching. We aimed to understand the vertical distribution and surficial release of PFAS from AFFF-impacted asphalt and concrete cores collected from various locations (~10–70 m distance between samples). Among the PFAS analyzed, 6:2 FTS was observed as having the highest concentration in the surface layer (0 – 0.5 cm) of concrete (225 μg kg^{-1}) and in the runoff from the concrete (2600 ng L^{-1}). PFOS was detected at the highest concentration in the surface layer (0 – 0.5 cm) of asphalt (47 μg kg^{-1}) and associated runoff (780 ng L^{-1}). The total mass of PFAS released during three rainfall simulations accounts for a fraction of the total mass in the surface layer (0 – 0.5 cm), ranging from 0.10 – 9.8% and 0.078 – 2.4% for asphalt and concrete cores, respectively. Asphalt exhibited a higher release rate

---

[*] Corresponding author.
E-mail address: p.thai@uq.edu.au (P.K. Thai).

https://doi.org/10.1016/j.jhazmat.2024.133627
Received 22 September 2023; Received in revised form 18 January 2024; Accepted 24 January 2024
Available online 26 January 2024

than concrete, demonstrated by the higher total release coefficient of PFAS (4 – 16 m^{-2}) compared to that of concrete cores (1 – 5 m^{-2}). These results suggested that, similar to concrete, AFFF-impacted asphalt may be a secondary source of PFAS to the environment.

1. Introduction

Per- and polyfluoroalkyl substances (PFAS) are key components of PFAS-containing firefighting foams (e.g., aqueous film forming foam [AFFF]). These foams form a stable film over the liquid fuel which encourages cooling and discourages re-ignition [1]. AFFF has been utilized for firefighting training exercises and emergency responses in various locations, such as military bases, airports, firefighting training areas, and fuel storage facilities. The use of AFFF, particularly repeated and frequent use, can result in PFAS residues with concentrations as high as several mg g^{-1} in infrastructure materials, including concrete (e.g., training pads) and asphalt (e.g., driveways, pavements) [2-5].

Previous equilibrium experiments and rainfall simulations have demonstrated that the depletion of the most common PFAS associated with certain AFFF formulations, such as perfluorooctane sulfonate (PFOS) and perfluorohexane sulfonate (PFHxS), is slow [2,4]. Our recent assessment also suggests that the release of PFAS from AFFF-impacted concrete to surface runoff may occur for decades in some circumstances if mitigation is not performed [6].

Infrastructure materials that may be potentially impacted at AFFF sites include concrete and asphalt. While there have been studies monitoring the release of PFAS from concrete cores through water runoff, to our knowledge, no similar study has been conducted for asphalt [7], except one recent study that developed and applied new methods for PFAS analysis in asphalt samples [3]. This scarcity of information or knowledge gap on the leaching of PFAS from AFFF-impacted concrete and asphalt could impede effective management of PFAS from AFFF-impacted sites [7]. Further, this knowledge would be useful to evaluate the appropriateness of mitigation or remediation of PFAS leaching from AFFF-impacted concrete and asphalt.

In this study, we investigated the distribution of PFAS in both AFFF-impacted asphalt and concrete as well as the dynamic release of PFAS in response to rainfall simulations in laboratory settings. Using those data, we aimed to confirm the release profile of PFAS from impacted concrete, and for the first time, explore the profile of PFAS release from asphalt. The differences between the distribution and release of PFAS in asphalt versus concrete were also assessed.

2. Materials and methods

2.1. Asphalt and concrete samples

Three asphalt cores (A1, A2, and A3; d = 95 mm) and three concrete cores (C1, C2, and C3; d = 65 mm) were collected from various locations (~10–70 m distance between samples) at an AFFF-impacted site (Fig. S1) using a core drill. To obtain the PFAS concentration profiles along the depth of cores, samples of asphalt and concrete were collected by drilling the sidewall of the cores at various depths down to the bottom (Table S3), similar to the practice of previous studies [2,4,5]. For each depth, 5 to 7 locations around the circumference of the cores were drilled (i.e., core sidewall drilling) and samples were combined to obtain a representative sample of each depth.

2.2. Release kinetics from rainfall simulations

Laboratory-based rainfall simulations were performed based on procedures described by Thai et al. [4]. A PVC ring (d = 75 mm for asphalt cores; d = 56 mm for concrete cores) was sealed on the surface of each core by a silicone sealant (Selleys). A 2 mm depth of pH = 7 MilliQ water was poured into the PVC ring (equivalent to 9 mL for the asphalt cores and 5 mL for the concrete cores) and drawn out completely within 2 min. This process was repeated 30 times, resulting in a simulated rainfall of 60 mm in one hour (i.e., a rainfall intensity of 1 mm min^{-1}).

A total of three rainfall simulations were conducted on each core with different drying periods (from one day to two weeks) between any two rainfall events to simulate alternating wetting and drying conditions in the field. A blank experiment was conducted in parallel by pouring pH = 7 MilliQ water (74 mL, equivalent to a water depth of 30 mm) into a PVC ring (d = 56 mm) sealed to aluminum foil with the same silicone sealant (Selleys). The blank sample was collected after ponding for 48 h.

2.3. Chemical analysis

Concrete samples (approximately 0.4 g) were spiked with a mixture of isotopically labelled PFAS (2 ng, or 10 μL at 0.2 ng μL^{-1}), followed by thorough mixing using a roller mixer and ultrasonic solvent extraction with two cycles of 2% ammonium/methanol (w/w). Water samples were spiked with a mixture of isotopically labelled PFAS (1 ng, or 5 μL at 0.2 ng μL^{-1}) prior to solid phase extraction (SPE) with Strata X-AW cartridges (100 mg/ 6 mL). Due to the significant decrease in PFAS concentrations in water samples from the later phase of rainfall simulation [4], two to three water samples collected consecutively after 20 min into the rainfall simulation were combined to increase the concentration factor of SPE. Detailed extraction procedures for concrete and water samples are provided in the Supporting Information (S1.2).

All extracts were analyzed with high performance liquid chromatography (Nexera HPLC, Shimadzu Corp., Kyoto Japan) coupled to a tandem mass spectrometer (SCIEX Triple Quad 6500 +, Concord, Ontario, Canada). The targeted chemicals include two perfluorocarboxylic acids (PFCAs): perfluorohexanoic acid (PFHxA) and perfluorooctanoic acid (PFOA), two perfluorosulfonic acids (PFSAs): PFHxS and PFOS, and 6:2 fluorotelomer sulfonate (6:2 FTS). Detailed information about instrumental analysis can be found in the SI (S1.3).

To provide quality assurance and quality control (QA/QC), procedural blank samples (every 20 samples), duplicate samples (every 10 samples), and samples spiked with native standards (every 20 samples) were included in each batch (20 – 40 samples) and processed in the same way as regular samples. QA/QC results are provided in the SI (Table S5). In the blank samples, PFOA, 6:2 FTS, and PFOS were not detected, and PFHxA and PFHxS were below the limit of detection. These suggest there is no/negligible release of the investigated PFAS from the silicone sealant used in the experiment.

2.4. Estimation of mass of PFAS in the surface layer and release coefficient of PFAS

The total mass load of individual PFAS in the top 0.5 cm layer ($M_{0.5}$) is estimated based on Eq. (S1) in the SI (S1.5). For the ease of comparing the surficial leaching of PFAS among different cores, the total release coefficient (m^{-2}) is calculated according to Eq. (S2), which represents the PFAS release normalized to the surface area and $M_{0.5}$. However, it should be noted that PFAS in layers deeper than 0.5 cm may also contribute to the surficial release of PFAS.

3. Results and discussion

3.1. Vertical concentration profiles of PFAS in asphalt and concrete cores

The vertical concentration profiles of three major PFAS compounds in asphalt and concrete cores are shown in Fig. 1 with additional two

PFAS compounds shown in the SI (Figs. S2 and S3). The vertical profiles in the concrete cores resemble those observed in previous studies [2,4], featuring the highest concentrations at the top 0.5 – 1 cm layer followed by a decrease in concentration for the lower depths. Asphalt A2 also displayed an overall decreasing trend in PFAS concentrations with depth, except for increases at a depth of 4 – 5 cm and the bottom of core (Fig. 1A, B, and C, Fig. S2A and B). Meanwhile, asphalt A1 showed a trend of initially increasing and then decreasing PFAS concentrations with depth, with the highest concentrations observed at the 2 cm depth (Fig. S2). The discrepancies between PFAS profiles of the two asphalt cores could be attributed to the heterogeneity of asphalt collected from different areas of the site. The increase in concentrations from the surface to the middle layers of A1 implies downward transport of PFAS after AFFF exposure. The mechanisms remain unclear with regards to the increase in PFAS concentrations at the depth of 4 – 5 cm and the bottom of A2. It could be due to the lateral seepage of AFFF-impacted runoff from the surrounding area or other unidentified factors [4]. Variations of PFAS vertical profiles in concrete cores have been observed in other recent studies [5,8].

The highest concentration observed in the concrete cores was of 6:2 FTS, reaching up to 225 μg kg^{-1}, followed by PFOS (152 μg kg^{-1}) and PFHxS (65 μg kg^{-1}) (Fig. S3). The highest concentration in the asphalt cores was of PFOS (47 μg kg^{-1}), followed by PFHxS (11 μg kg^{-1}) and 6:2 FTS (3.3 μg kg^{-1}) (Fig. 1). The presence of high concentrations of 6:2 FTS, PFOS, and PFHxS in the investigated cores suggests that both fluorotelomer- and PFOS-based AFFF products were historically used at the study site.

Compared to the concentrations of similar PFAS compounds present in the surface of concrete cores reported in previous studies [2,4], the results for concrete and asphalt analyzed in this study are much lower. The comparatively lower concentrations of PFAS in this study are likely attributable to smaller volumes and less frequent historical use of AFFF, though site-specific conditions such as environmental weathering and meteorological conditions (e.g., annual rainfall) may also contribute. Cores from Baduel et al. [2] and Thai et al. [4] came from former firefighting training grounds (FTG) where larger volumes of AFFF were historically used at a greater frequency.

We acknowledge that site-specific factors can influence the amount of PFAS retained within AFFF-impacted concrete and asphalt. Nevertheless, our findings provide valuable information on the expected variability of PFAS within concrete and asphalt at different sites and their corresponding leaching potential, which is lacking in the literature [5]. This variability is an important consideration when performing an environmental risk assessment or when considering the cost-benefit of attempting to mitigate PFAS leaching from concrete or asphalt.

3.2. Comparison of release kinetics between asphalt and concrete cores from rainfall simulations

We selected a total 60 mm rainfall depth with a constant rainfall

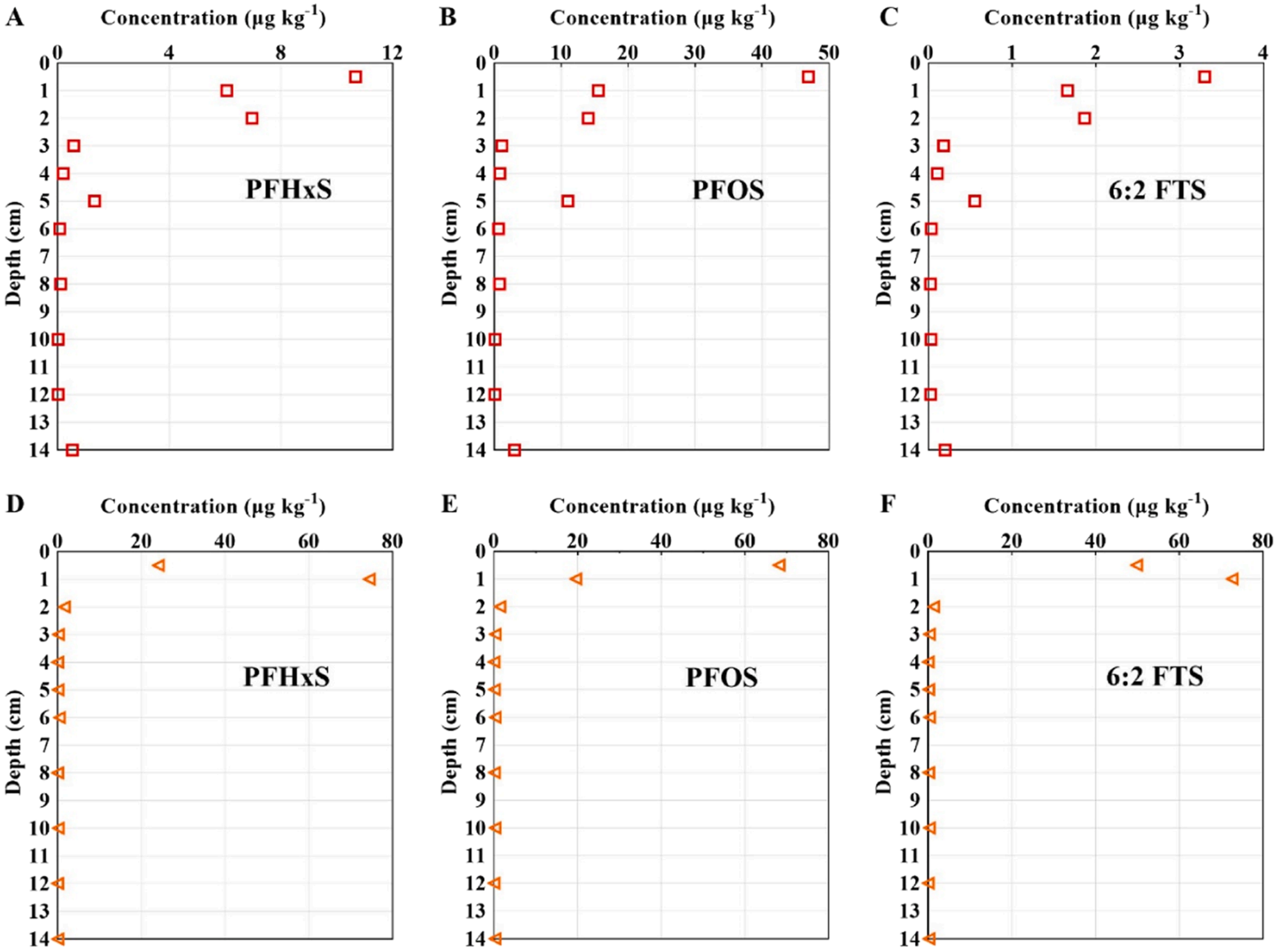


Fig. 1. Vertical concentration profiles of PFHxS, PFOS, and 6:2 FTS in asphalt core A2 (A, B, and C in the upper panel) and concrete core C2 (D, E, and F in the lower panel).

intensity of 1 mm min^{-1} for our experimental design. While the total rainfall depth of 60 mm could be considered an extreme case in reality, it is necessary for our study design and research questions. By simulating a rainfall with a constant rainfall intensity (1 mm min^{-1}) for 60 min and with high frequency sampling, we achieved the objectives of this research, which were to understand the behavior and kinetics of PFAS leaching from asphalt and concrete. Further, our findings can be interpolated to any rainfall depth within the range of 2 to 60 mm. Understanding PFAS leaching under different rainfall durations is essential to develop long-term, robust management strategies under different weather scenarios.

The concentration of five PFAS compounds in runoff water from both asphalt and concrete surfaces during rainfall simulations exhibited an exponential decreasing trend (Figs. 2, S4–S7). Concentrations of PFAS decreased rapidly in the first 10 min of rainfall (equivalent to 10 mm of rainfall depth at the rainfall intensity of 1 mm min^{-1}) followed by a slower decline and eventually reaching a plateau in the remaining duration of the rainfall simulation. The observed runoff profiles of PFAS from concrete cores are consistent with those reported by Thai et al. [4] despite concentrations being lower (in concrete and the associated runoff). The peak concentrations of PFAS were observed at the beginning of the rainfall event, suggesting that PFAS are more amenable to leaching into the initial runoff compared to the later phase of the rainfall event. These results are consistent with the “first flush” effect, where the initial runoff volumes receive disproportionally higher loads and have much higher concentrations compared to subsequent runoff volumes [9].

Among the three rainfall simulations for asphalt cores, the first rainfall event resulted in the highest percentage of released PFAS relative to the mass of PFAS in the top 0.5 cm (Fig. S8). For instance, the maximum percentage of PFOS released from the three asphalt cores were 4.8%, 2.3%, and 1.6% in the first, second, and third rainfall simulations, respectively. Similarly, concrete core C1 also exhibited the highest release percentage of PFOS during the first rainfall simulation (0.09%) compared to second (0.04%) and third (0.04%) rainfall simulations. In contrast, concrete core C2 exhibited a more comparable release percentage during the first and third rainfall events, 0.18% and 0.24%, respectively. With the highest percentage of observed PFAS release occurring within the first rainfall simulation and with a consistent observation of a rapid decline over the course of each rainfall simulation (Fig. 2), the overall mass release of PFAS to a watershed may be negligible. Therefore, mitigation of PFAS leaching from concrete and/or asphalt may only be required if stormwater compliance in a "first flush sample" is jeopardized.

Thai et al. [4] also found similarity in initial PFAS concentrations during five successive rainfall simulations on concrete cores, although at higher concentrations (μg L^{-1}) compared to concentrations (ng L^{-1}) observed in our study. They attributed the initial similarity to a “wicking” effect, in which PFAS in the subsurface are transported upward with water to the surface of the concrete, and subsequently remain on the surface while water evaporates, making them susceptible to release during subsequent rainfall events [4]. Assuming PFAS released during the second and third rainfall simulations are attributed to the wicking effect, the released mass could be expected to be correlated to the total mass in the top surface. As shown in Fig. S9A, there is a strong positive correlation ($r^2 = 0.999$) between the total mass of PFAS released from asphalt cores during the second and third rainfall simulations and the mass of PFAS in the top 0.5 cm of asphalt cores. The correlation was not as strong for concrete cores (Fig. S9B), which could be attributed to the differences in permeability and/or chemistries associated with asphalt versus concrete materials.

The total mass of PFAS compounds released during three rainfall simulations accounts for a fraction of the total mass in the surface layer (0 – 0.5 cm), ranging from 0.10 – 9.8% and 0.078 – 2.4% for asphalt and concrete cores, respectively (Table S1). These data suggest that both asphalt and concrete impacted by AFFF can act as a secondary source of PFAS released into the environment, but the release is small in comparison to that retained within the asphalt and concrete, may continue for a long time, and ultimately may only influence first flush stormwater compliance rather than meaningfully impact the larger watershed. It is important to note that the estimation of PFAS mass in the top 0.5 cm was based upon discrete samples collected using core sidewall drilling, a simplified equation, and did not take into consideration the heterogeneity of PFAS distribution in asphalt and concrete cores, which has been demonstrated by Vo et al. [8].

Asphalt exhibited greater variations in release coefficients of PFAS compared to concrete cores (Fig. 3), suggesting a higher degree of heterogeneity and/or different release mechanisms. The average release coefficient for each rainfall simulation was higher in the three asphalt cores than that in the concrete cores, although the difference was not statistically significant ($p > 0.05$, Table S2). Furthermore, the average total release coefficient of PFAS during three rainfall simulations for asphalt cores was in the range of 4 – 16 m^{-2} (Table S1), which is higher than that for concrete cores ranging from 1 – 5 m^{-2}.

Among the concrete samples, C1 demonstrated the highest total cumulative released mass for five PFAS compounds combined (41 μg m^{-2}), followed by C2 (13 μg m^{-2}) and C3 (3.1 μg m^{-2}). The higher mass of PFAS in the surface layer of C1 could explain its higher mass release in the runoff (Table S1). However, despite C2 demonstrating a higher cumulative release for all targeted PFAS than C3, C2 and C3 exhibited similar levels of PFAS mass in the top 0.5 cm. These results suggest that the release of PFAS from concrete is affected by other factors in addition to the total mass of PFAS.

The higher average total release coefficient of PFAS from asphalt determined in this study (4 – 16 m^{-2}) compared to concrete (1 - 5 m^{-2}) suggests that PFAS may be more readily leached from asphalt than concrete. However, it should be noted that these differing rates could be due to differing conditions of the asphalt and concrete surfaces at this site. For example, the amount and frequency of AFFF is assumed to be higher for concrete samples within the loading area compared to asphalt

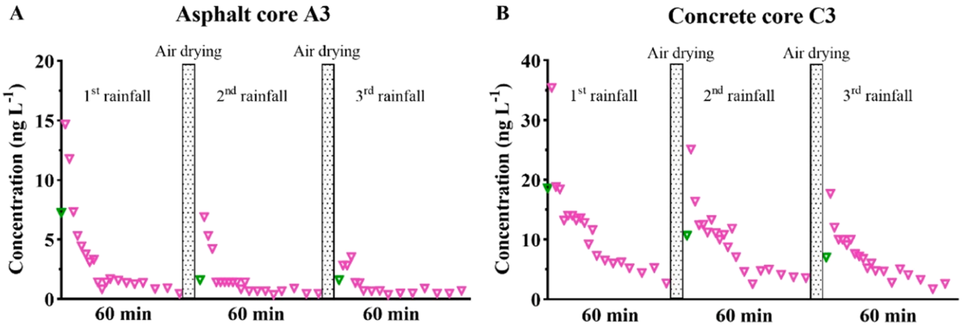


Fig. 2. Concentration response of PFOS in runoff water during three rainfall simulations on asphalt (A) and concrete cores (B). The green symbols indicate the PFOS concentration in the water collected at the beginning of the rainfall simulation (i.e., the pH = 7 MilliQ water was poured on the surface and drained out immediately).

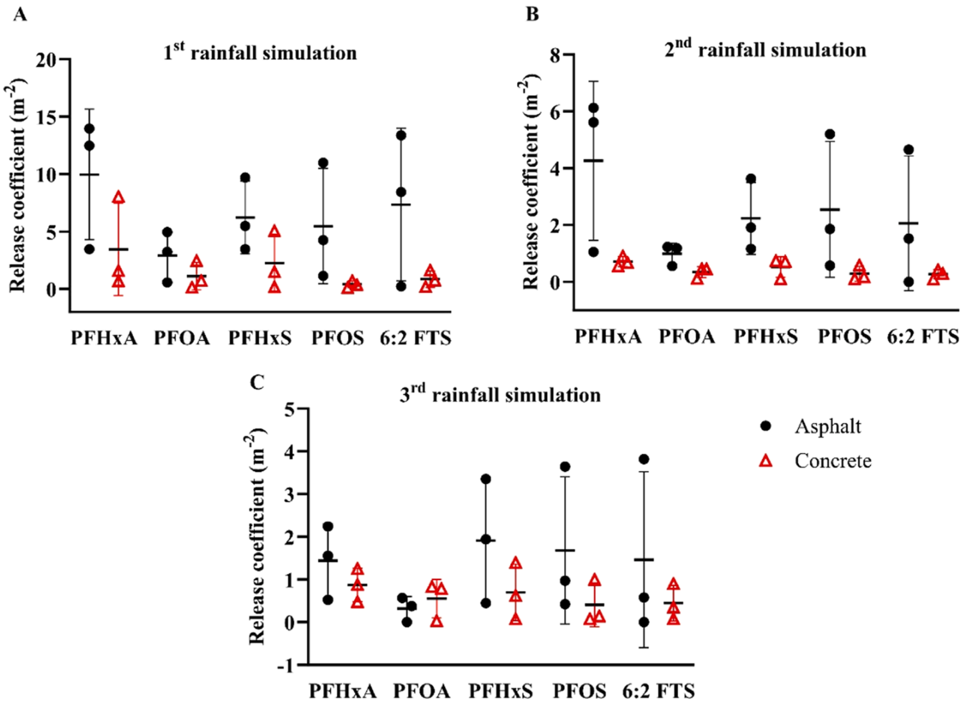


Fig. 3. Release coefficient (m^{-2}) of target PFAS in asphalt and concrete cores during 1st (A), 2nd (B), and 3rd (C) rainfall simulations. The middle line represents the mean value and the error bar indicates the standard deviation (SD).

samples in the surrounding area. Additionally, the location of the concrete cores was historically covered by a canopy until the demolition of the loading area. To the best of the authors' knowledge, the location of the asphalt cores has always been uncovered. These differences, as well as other potential undetermined factors, may have implications on the release coefficients of PFAS from concrete and asphalt. More study and evaluation is necessary to consider how these differences may influence the comparative release coefficients of PFAS from asphalt and concrete surfaces.

3.3. *Future perspective*

There are uncertainties and limitations of estimating PFAS mass in surficial samples. Obtaining a higher resolution mass distribution of PFAS in concrete and asphalt would be beneficial and may provide valuable insights into the release behavior of PFAS. Further, the controlled laboratory conditions of the rainfall simulation did not consider characteristics of natural rainfall events, such as droplet size distribution, kinetic energy, and contact time. Other factors (e.g., rainfall intensity, slope, and surface roughness) can affect the release of PFAS in the field. Therefore, it would be of value to further investigate the relationship between these factors and PFAS storage and release from concrete and asphalt using both laboratory- and field-scale efforts.

4. Conclusions

In this study, we have demonstrated that both AFFF-impacted asphalt and concrete surfaces may serve as secondary sources of PFAS to runoff. Our results showed concrete samples have a higher PFAS mass concentration than asphalt samples, which could be due to differing site-specific AFFF use patterns (amount, frequency) on concrete and asphalt surfaces, differing environmental weathering scenarios, or other currently undetermined differentiating factors. Further, the surficial release of PFAS was observed to be slower from concrete samples than asphalt samples (although not statistically significant), which may suggest potential differences in storage and release mechanisms between these materials. Our findings highlight the value of characterizing both AFFF-impacted concrete and asphalt hardscapes at the field-scale, as well as the need for further studies to evaluate material properties to help describe and distinguish storage and release mechanisms between concrete and asphalt.

CRediT authorship contribution statement

Key Trent: Writing – review & editing, Funding acquisition, Conceptualization. **Li Yijing:** Writing – original draft, Methodology, Formal analysis, Data curation. **Thai Phong:** Writing – review & editing, Supervision, Conceptualization. **Mueller Jochen:** Writing – review & editing, Supervision. **Barnes Craig:** Writing – review & editing. **Fiorenza Stephanie:** Writing – review & editing. **McDonough Jeff:** Writing – review & editing, Conceptualization. **Thapalia Anita:** Writing – review & editing. **Portman Scott:** Writing – review & editing, Conceptualization. **Vo Phong:** Writing – review & editing, Methodology.

Declaration of Competing Interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Phong Thai reports financial support was provided by ExxonMobil Environmental and Property Solutions Company.

Data availability

Data will be made available on request.

Acknowledgments

The Queensland Alliance for Environmental Health Sciences, The University of Queensland gratefully acknowledges the financial support of Queensland Health. The study is contract research for Arcadis and ExxonMobil Environmental and Property Solutions Company. Jochen Mueller is funded by an Australian Research Council (ARC) Laureate Fellowship (FL 200100028).

Environmental implication

There is a gap in our understanding of PFAS contamination in concrete and asphalt materials in critical infrastructure that have been exposed to extensive use of aqueous film forming foams (AFFF). This study will address this issue with a detail investigation of the distribution and release of PFAS from both concrete and asphalt cores collected from site affected by legacy use of AFFF. The findings will help design better and more cost-effective PFAS management in the affected sites.

Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.jhazmat.2024.133627.

References

[1] Moody, C.A., Field, J.A., 2000. Perfluorinated surfactants and the environmental implications of their use in fire-fighting foams. Environ Sci Technol 34 (18), 3864–3870.

[2] Baduel, C., Paxman, C.J., Mueller, J.F., 2015. Perfluoroalkyl substances in a firefighting training ground (FTG), distribution and potential future release. J Hazard Mater 296, 46–53.

[3] Srivastava, P., Williams, M., Du, J., Navarro, D., Kookana, R., Douglas, G., Bastow, T., Davis, G., Kirby, J.K., 2022. Method for extraction and analysis of per- and poly-fluoroalkyl substances in contaminated asphalt. Anal Methods 14 (17), 1678–1689.

[4] Thai, P.K., McDonough, J.T., Key, T.A., Thompson, J., Prasad, P., Porman, S., Mueller, J.F., 2022. Release of perfluoroalkyl substances from AFFF-impacted concrete in a firefighting training ground (FTG) under repeated rainfall simulations. J Hazard Mater Lett 3, 100050.

[5] Williams, M., Douglas, G., Du, J., Kirby, J., Kookana, R., Pengelly, J., Watson, G., Bowles, K., Davis, G., 2023. Quantification of the variability and penetration of per- and poly-fluoroalkyl substances through a concrete pad. Chemosphere 333, 138903.

[6] Vo, P.H.N., Key, T.A., Le, T.H., McDonough, J.T., Porman, S., Fiorenza, S., Nguyen, H.T.M., Dao, V.T.N., Mueller, J.F., Thai, P.K., 2023. Evaluation of sealants to mitigate the release of per- and polyfluoroalkyl substances (PFAS) from AFFF-impacted concrete: Characterization and forecasting. Water Res X 20, 100195.

[7] Douglas, G.B., Vanderzalm, J.L., Williams, M., Kirby, J.K., Kookana, R.S., Bastow, T. P., Bauer, M., Bowles, K.C., Skuse, D., Davis, G.B., 2023. PFAS contaminated asphalt and concrete - Knowledge gaps for future research and management. Sci Total Environ 887, 164025.

[8] Vo, P.H.N., Hamilton, B.R., Wepf, R.A., Key, T.A., Nguyen, T.M.H., Thai, P.K., Thomas, K., Mueller, J.F., 2023. Visualization of the Distribution of PFOS and PFHxS in Concrete by DESI MSI. Environ Sci Technol Lett 10 (5), 446–451.

[9] Yun, D., Kang, D., Cho, K.H., Baek, S.-S., Jeon, J., 2023. Characterization of micropollutants in urban stormwater using high-resolution monitoring and machine learning. Water Res 235, 119865.