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Per- and poly-fluoroalkyl substances (PFAS): Current status and research needs



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ABSTRACT

An expert workshop focusing on per- and poly-fluoroalkyl substances (PFAS) was held in Adelaide, South Australia, Australia in September 2019 following the 8th International Contaminated Site Remediation Conference – CleanUp 2019. The workshop was organised by the Cooperative Research Centre for Contamination and Remediation of the Environment (CRC CARE) and was chaired by Professor Ravi Naidu, CEO and Managing Director of CRC CARE and Director of the Global Centre for Environmental Remediation at the University of Newcastle, NSW. The purpose of the workshop, which was attended by more than 50 experts in the field of contaminated land assessment and management, was to discuss the current state of play and research needs relating to PFAS contaminated sites. This paper provides a summary of the discussions and conclusions and lists actions

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and needs that the expert group identified as critical for pursuing successful PFAS management and remedy approaches.

This paper is intended to capture the shared information, comments, and current thinking related to PFAS challenges and research needs as identified by the group of expert participants; the write up is not intended to be a complete dissertation on the science and work that has been carried out. With a fast-evolving subject and increased government and public attention on PFAS presence in the environment, the group was convened with the objective of providing value in contributing to solutions to the PFAS challenges that are faced both in Australia and internationally. The text contained herein provides references to observations and methods that the experts drew on in their discussions and in support of their commentary; documentation of the original references was not provided, and the reader should consult the scientific literature if further information and confirmation of observations is required. Following a brief on the background to PFAS challenges, the paper focusses on research gaps identified by experts with focus on Australian soils and groundwater including climatic patterns, an overview of PFAS research in Australia with emphasis on:

- Regulatory
- Analytical considerations
- Ecological and Human Health Risks
- Fate and Transport
- Remediation and Risk Management.

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1. Introduction

1.1. Background to the PFAS challenge

Per- and poly-fluoroalkyl substances (PFAS) are man-made chemical compounds that have been used in industry and consumer products worldwide since the 1950's (Wang et al., 2014b). PFAS are extremely stable substances used in wide range of industrial and consumer applications as surfactants or surface protectors, and are globally distributed in the environment (Taniyasu et al., 2003; Prevedouros et al., 2006). Buck et al. provided a structural definition of PFAS as "the highly fluorinated aliphatic substances that contain 1 or more C atoms on which all the H substituents... have been replaced by F atoms, in such a manner that they contain the perfluoroalkyl moiety $C_nF_{2n+1}-$ " (where n is equal to or greater than 1, i.e. the structure must contain at least one CF_3- group) (Buck et al., 2011).

Along with their hydrophobic and oleophobic properties, their stability gives them very useful properties for multiple industrial uses, including in firefighting foams (known as 'aqueous film-forming foams', or AFFFs), non-stick cookware, food packaging, insecticides, and waterproof and fire-resistant fabric (Prevedouros et al., 2006; Buck et al., 2011; Moody and Field, 2000; Kärrman et al., 2011; Simon et al., 2019). However, this stability also means that they do not break down easily, and therefore accumulate in the environment. As a result, they are so widespread that almost every person in the world has been potentially exposed to PFAS and has traces of these chemicals in their blood (Taniyasu et al., 2003; Moody and Field, 2000; Kärrman et al., 2011; Schultz et al., 2004; Washington et al., 2009). Some PFAS can enter ecosystems and move up through food chains, accumulating in animal and human tissue, including the liver and kidney (Yang et al., 2019), particularly for long-chain perfluoroalkyl acids (PFAAs) (Wang et al., 2014b; Seow, 2013; Wang et al., 2014c,a, 2015b; Chropeňová et al., 2016).

Due to the persistent and bio-accumulative nature of perfluorooctanesulfonic acid (PFOS), PFOS and its salts and perfluorooctane sulfonyl fluoride (PFOSF) were added to the persistent organic pollutants (POPs) list at the Stockholm Convention on Persistent Organic Pollutants in 2009. In addition, C8–C14 perfluoroalkyl carboxylates (PFCAs) and perfluorohexane sulfonate (PFHxS), and their sodium and ammonium salts were included in the candidate list of Substances of Very High Concern (SVHC) in the EU.

While the manufacture of AFFFs containing long-chain PFAAs is now prohibited, it is still legal to use existing stockpiles in Australia. However, both Queensland and South Australia have recently moved to ban their use, including precursors, altogether.

The past use of AFFFs continues to represent a significant legacy issue. AFFF have been widely used by fire protection personnel, particularly where petroleum hydrocarbons have been stored and used, such as at airfields and in the petroleum industry (Kärrman et al., 2011; Houtz et al., 2013; Weiner et al., 2013; Hetzer et al., 2014; Houtz et al., 2016). Large quantities of AFFF have been released directly to the environment during regular training of firefighters and testing of fire protection systems. Such releases have occurred at many thousands of sites, particularly at airports, air force bases and other military sites, at metropolitan and country fire service training sites, and in the petroleum industry at refineries and oil terminals.

The PFAS constituents in the foam are very persistent and mobile and can enter into and migrate in soil and groundwater, eventually moving beyond the source of release, and in some cases for several kilometres (Simon et al., 2019; Milley et al., 2018; Wittenberg, 2018). At some of these sites, various PFAS have been found at elevated levels in surface water and groundwater well outside the facility boundaries (Hu et al., 2016). Given the stability of fluoro-alkyl substances in normal environmental conditions, once introduced in the environment, these substances can remain in environmental matrices for many years. Concern for PFAS exposure has increased since it has been found that some PFAS accumulate and can persist in human blood for many years (USEPA, 2016a,b). In communities where land and groundwater have been contaminated, there has been widespread concern, litigation, and property prices have fallen (Australia, P.o., 2018).

While there is no direct evidence linking PFAS exposure to human deaths, there are now numerous studies suggesting that PFAS exposure may be connected to a number of human health effects (USEPA, 2016a,b). For example, some PFAS may act as “endocrine disruptors”, and may have multiple other toxic effects such as hepatotoxicity, immunotoxicity, reproductive and developmental toxicity and tumorigenic effects. Some PFAS have also been linked to neurodevelopmental toxicants at various levels. A number of studies reveal a possible relationship between exposure to certain PFAS and neurodevelopmental disorders in humans. Additional data were obtained from an Ohio survey, where a positive association between “Attention deficit hyperactivity disorder” (ADHD) and the level of PFHxS and PFOS in serum samples was found in children (Stein and Savitz, 2011). It is estimated that increase of 1.5 ± 0.7 mg/dL and 0.4 ± 0.2 mg/dL of total cholesterol can occur for unit PFOA (perfluorooctanoic acid) and PFOS increase (ng/mL), respectively (Dong et al., 2019). Some studies have shown a significant correlation between PFOA levels in maternal blood and adverse pregnancy outcomes. Such findings have led to significant anxiety amongst people that are potentially exposed to PFAS. Due to the considerable uncertainty that has arisen through these various studies, and because measurable concentrations of several PFAS including PFHxS, PFOA, PFOS, and PFNA (perfluorononanoic acid) are routinely detected in human blood, health agencies have taken a precautionary approach in seeking to limit exposure, and have been setting very low acceptable levels of exposure (USEPA, 2016a,b).

It is recognised that PFAS are likely to be present at a large number of sites in Australia and, where present, can pose a significant concern to site owners, regulators, and communities. The concern increases with sites where PFAS contamination is found to migrate off site and contaminate groundwater or surface water. Where contamination is found or suspected, regulatory agencies can be expected to require a process of site assessment, risk characterisation and remediation, depending on the severity of the problem. As contaminants, PFAS presents particular difficulties (Simon et al., 2019) because there are large numbers of ill-defined chemical species that can be present for long periods. Some of the more commonly encountered PFAS are persistent, mobile, and potentially toxic. The toxicological criteria have been assigned for only a few of the PFAS, and there is limited scientific evidence to develop informed policy and regulation. In addition, analytical methods are practically limited to quantifying only a subset of the PFAS constituents that may be present (Kärman et al., 2011; Schultz et al., 2004; Washington et al., 2014a). The stability and widely distributed nature of the PFAS in the environment limited the comprehensive understanding of presence of PFAS (Codling et al., 2014; Dauchy et al., 2017). Furthermore, the difficulty in understanding the variety of factors that affect the mobility and presence of particular PFAS, makes control and remediation difficult (Hetzer et al., 2014; Hu et al., 2016; Place and Field, 2012; EPA, 2014; Anderson et al., 2016).

This situation has given rise to community concern and litigation, with class action lawsuits (Bntham, 2020; Zillman, 2020; Studdert et al., 2019) now being mounted by affected communities at many locations in Australia.

1.2. Australian soils and ground water

Australia's climatic patterns, geology, soil and ground water conditions vary considerably from south to north, and also to some extent, from west to east. Variations in soil types are the result of continuous processes of movement and erosion over millions of years, resulting in a variety of landscapes across Australia. As shown in the soils map of Australia (Fig. 1), there are 15 major soil types with their coverage varying from 0.1% (organic soils) to 26% (Tenosols). There is considerable geographic variation in the organic matter (OM) content of soils, with slightly higher OM content in soils from Southern Australia relative to strongly weathered soils from Northern Territory. In addition, the salt content of Australian soils also varies, with West, East and Southern Australia showing a fairly extensive distribution of salt affected soils (sodosols) whereas Northern Australia is dominated by Kandosol, Ferrosol and Tenosol with these soils being relatively rich in oxidic material. Given these differences in physico-chemical and mineralogical properties of Australian soils, the fate and behaviour of PFAS in the soils are expected to vary considerably and as a consequence the risk profile to receptors is likely to vary as well. Hence there is a need for differing strategies for the management of remediation of these soils depending on geographical location of contaminated sites (Washington et al., 2009; Houtz et al., 2013; Washington et al., 2014b; Söregård et al., 2019).

As with soils, the composition of ground water also varies considerably depending on geographical location of the sites, with ground water in Mid and Southern Australia showing elevated salinity compared to that in Northern Australia. Under these conditions the mobility and dispersion of PFAS constituents could vary considerably – though there have been limited comparative studies conducted thus far to provide clear evidence of this. Discharges with varying salinities such as that from wastewater treatment plants (WWTPs) into rivers, estuarine and marine environments could alter surface water salinities. There has been some work on ‘salting out’ of some PFAS when discharging to estuarine and marine environs.

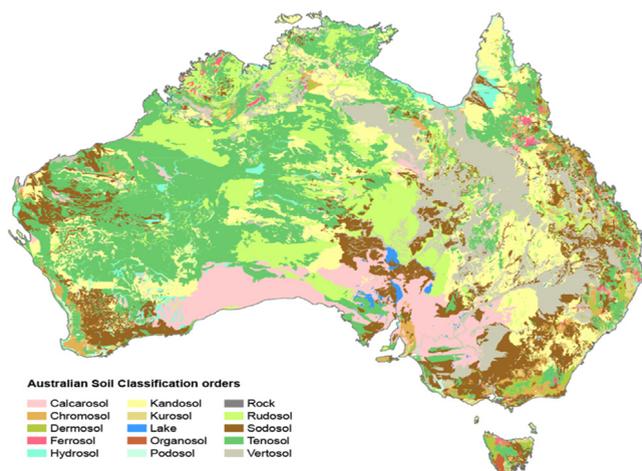


Fig. 1. Major soil types in Australia. Source: (Data from the Australian Soil Resource Information System and Conversion of the Atlas of Australian Soils to the Australian Soil Classification, <https://soe.environment.gov.au/theme/land/topic/2016/soil-understanding>).

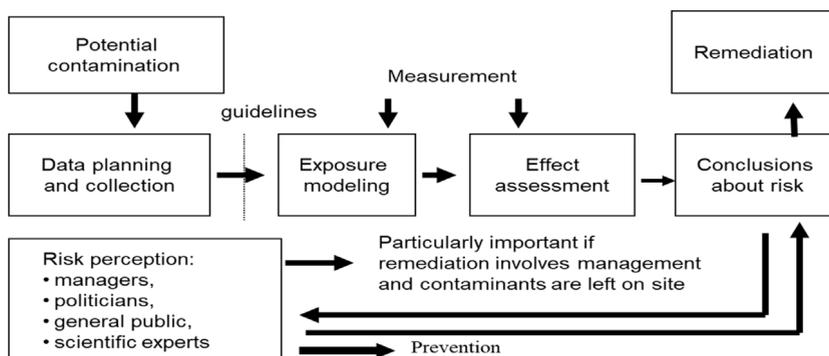


Fig. 2. Key components of contaminated site assessment leading to decision on either remediation or management of contaminated sites in Australia as part of risk-based management.

2. Overview of PFAS research in Australia

Most of Australia's initial research efforts in the area of PFAS have been driven by the Australian Department of Defence and by CRC CARE. The finding of PFAS on many Australian Defence bases led to a significant investment by the Department of Defence towards the assessment, risk characterisation and remediation of PFAS contamination. However, these initiatives were significantly constrained by the lack of scientific data underpinning the site assessment and risk characterisation, and the lack of proven methods of PFAS remediation (Seow, 2013; USEPA, 2016a; EPA, 2014). In addition to the large funds directed towards site assessment, the Department of Defence also initiated a significant PFAS research program via the Australian Research Council's PFAS special research initiative, particularly aimed at understanding the applicability of various remediation technologies. There has been relatively little research effort directed towards understanding the science underpinning regulations, analytical techniques, toxicological studies, and risk characterisation, although information relevant to some of these matters has been assembled through the Defence site assessment program.

It is apparent from Fig. 2 that any decision on the level of risk and the requirements for remediation (or risk management with the contaminant remaining in place) requires (a) site assessment for contaminant delineation, involving standard analytical procedures for the identification of contaminants, and (b) exposure modelling and risk assessment, which is underpinned by an understanding of the toxicity of the contaminants.

While information on all stages of site characterisation and remediation is generally available for conventional contaminants such as metal species/ions and petroleum hydrocarbons, PFAS is constrained on all fronts. That is, there

are significant limitations related to: (1) identification and quantification of all of the PFAS present; (2) their toxicology (both as single compounds as well as mixtures of the compounds present); (3) bioavailability and trophic transfer by plants and animals and the bioaccumulation and biomagnification. Consequently, these limitations will constrain the development of feasible technologies for remediation, and policy underpinned by sound science for determining end-points for remediation.

Given these challenges, CRC CARE organised PFAS experts' symposium with a view to determining key research gaps with particular emphasis on Australian conditions. The team of experts included regulatory representatives from Australia, US, Canada, and Finland and practitioners currently engaged with PFAS assessment and remediation. The symposium took into consideration key components of Fig. 2 and addressed the following themes:

- Regulatory
- Analytical considerations
- Ecological and Human Health Risks
- Fate and Transport
- Remediation and Risk Management.

The outcomes of the symposium are summarised in the following sections.

3. Regulatory challenges to PFAS and the Australian response

3.1. Australian government action

In August 2017, the Heads of EPAs Australia and New Zealand (HEPA) released a consultation draft of the PFAS National Environmental Management Plan (NEMP). The NEMP fits within an overarching management framework for PFAS that was developed by the State, Territory and Commonwealth governments. The NEMP provides a nationally consistent approach to environmental regulation of PFAS and was first published in February 2018 and updated in May 2020. In 2017 the Environmental Health Standing Committee (enHealth) of the Australian Health Protection Principal Committee published human health toxicological criteria for PFOS, PFOA and PFHxS, and the NEMP has adopted these criteria (Milley et al., 2018).

The NEMP deals with a broader range of topics than those considered by CRC CARE, including waste disposal and the storage and transport of contaminated material. In parallel with this, CRC CARE prepared a guide for practitioners, which includes considerable detail on the assessment, remediation and management of PFAS contaminated sites. The CRC CARE guidance document has drawn on the screening criteria from the NEMP and is compatible with and supports the NEMP (CRCCARE, 2017b,a). The toxicological criteria incorporated in the NEMP (and hence the criteria for drinking water (70 ppt for PFOS+PFHxS), which depends on the matrix, regulation agency, and PFAS targets) are generally regarded as conservative, reflecting the uncertainty as to whether exposure to PFAS will give rise to significant health effects (Milley et al., 2018). The 70 ppt combined PFOA and PFOS concentration in drinking water is broadly consistent with the USEPA's Lifetime Health Advisories for PFOA and PFOS published on May 15, 2016 (USEPA, 2016a,b).

The NEMP also included screening criteria for protection of aquatic ecosystems; for many receiving waters the screening level for PFOS is 0.23 ng/L; this is less than the level of analytical quantification (unless certain sample preparations are carried out, such as pre-extraction and pre-concentration), and creates difficulty in determining the level of risk (Houtz et al., 2016; Wittenberg, 2018; Hu et al., 2016). The NEMP also includes guidance on tissue concentrations in biota to protect ecological and human receptors, but as there are large differences in measured bioaccumulation factors (BAF) across environments, species and PFAS, there are not reliable BAF to apply in a predictive sense to derive alternate environmentally protective concentrations.

3.2. Clean up goals and end points for soil and water remediation

Since its establishment in 2005, CRC CARE has worked in partnership with regulators and owners of potentially contaminated sites to develop guidance documents for assessment, management and remediation of contaminated sites. A key consideration in developing these documents was establishing what constitutes the end point for remediation, focusing initially on petroleum hydrocarbons. For sites with a significant remediation challenge that prevents groundwater restoration to Australian state clean up goals, CRC CARE in collaboration with regulators and end-users developed a guidance document on technical impracticability (TI) and Clean Up to the Extent Practicable (CUTEP) determination (CRC CARE 2010, Technical Report No. 18) with the decisions essentially based on an inability to treat, remove, or contain contaminants and long timeframes to reach clean up goals. However, in the absence of data on toxicity, analytical methods for the identification and quantitation of trace levels of active PFAS in groundwater, and remedial technologies, the application of TI and CUTEP for the management of PFAS contaminated groundwater is difficult. Guidance on TI and CUTEP for PFAS needs further consideration by Australian regulatory agencies – especially understanding what would be the key factors on which TI could be based (Milley et al., 2018).

3.3. Key regulatory gaps

It was recognised that much of the information on Environmental Investigation and Human Health threshold values included in the NEMP are based on guidance developed in other countries, and have many underlying assumptions.

Key research gaps and where work is needed are

- **Detection of PFAS:** There is a lack of data on previous or existing uses of PFAS in certain industries and products on the market, ambient levels, and the potential for unrealised emission/exposure.
- **Development of guidelines:** There is a lack of scientifically based environmental and human health investigation levels for many (most) PFAS, including the PFAAs and their precursors, and a lack of understanding of the reliability of guideline/investigation levels that have been published, whether these are likely to overestimate or underestimate the potential for significant effects, and the contribution of exposure through other pathways. There is excessive reliance and reference to very conservative drinking water guideline values being adopted in some of the regions of the US and European nations.
Also there is lack of guidance and harmonised policy on PFAS in landfill leachates, biosolids and wastewater treatment plants, lack of guidance on risk-based approaches to managing PFAS contamination in soil and sediment. There is lack of data on food intake to body weight ratios applicable to endemic Australian animal species, which is necessary for deriving more reliable guidance values. The current values in the PFAS NEMP are based on North American species.
- **Toxicological effects of PFAS:** there is lack of data on the toxicity (human health and eco) and valid approaches for mixtures of PFAS compounds and PFAS that may not be able to be analysed via standard methods, and lack of resulting policy approach that can take into account uncertainty and the potential for these uncertainties without leading to excessive levels of precaution and conservatism.
- **Remediation targets:** there is lack of information on end points (clean up goals) for soil and groundwater remediation, and whether current regulatory requirements are unreasonable and unsustainable.

These critical gaps in the PFAS knowledge base means that there are serious limitations on the ability to reduce what might be thought to be unnecessary conservatism, and instead adopt more cost-effective, risk-based remediation solutions which have been very successful in providing a sustainable solution to managing other contaminants. Rather, the current state of knowledge lends itself to cautious and uncertain policy development (Simon et al., 2019).

4. Analytical considerations for PFAS identification and quantitation in environmental matrices

4.1. Background

Regulatory and community concern regarding the possible effects of PFAS exposure have led to a large increase in the need for accelerated analyses of PFAS in environmental matrices, including animal tissues. Research labs have been challenged with the development of analytical techniques that can measure PFAS in a variety of matrices (e.g., surface waters, sea waters, landfill leachate, biota) that are seen as posing risks to receptors, including environment and human health (Chropeňová et al., 2016; Milley et al., 2018; USEPA, 2016a,b; Björnsdotter et al., 2019).

The key to successful analyses of mixtures of organic constituents in test material is effective separation, concentration and detection. For this, significant expertise in column chemistry, separation/concentration of the analytes and the availability of analytical instruments with low detection limits is required. Progress on these fronts has largely been made using liquid chromatography/dual mass spectrometry (LC/MS/MS) (USEPA, 2016a,b; EPA, 2014; Shoemaker et al., 2009; Coggan et al., 2019c). In the absence of this instrumentation, the ability to detect and quantify to the ultra-low (ppt) screening values set by the regulatory agencies is extremely difficult.

While the USEPA has taken the lead with the development of standard methods that quantify the presence of up to 23 PFAS constituents in water (USEPA Method 537 Version 1.1 and Method 537.1), there is a need for standard methods for identifying and measuring the presence of the other PFAS that are present, and for PFAS in matrices other than water. As of early 2020, USEPA is currently working on the development of additional methods for expanded PFAS analytical capabilities in terms of more analytes and more matrices (USEPA, 2020).

The considerable expenditure is now being directed to the assessment of PFAS contaminated sites, which has been a strong driver for commercial laboratories to develop new analytical techniques for the separation and analysis of PFAS constituents in a range of matrices including water, wastewater, soil, biosolids, tissues, blood etc. A key consideration for the commercial laboratories has been assuring quality control, with problems in the lack of internal standards, the lack of isotopically labelled standards, and limited access to sensitive instruments to detect low/trace concentrations. The use of isotope dilution for quantitation is superior to the internal standard method that is prescribed, because it accounts for matrix effects, allows lower reporting limits, and provides greater accuracy (van Leeuwen et al., 2009; Point et al., 2019). This is most useful for the analyses of PFAS in water and wastewater samples that may be turbid or may have elevated levels of salt that can interfere with the accuracy and precision of the other methods. In the USA, much progress on this front has been made with funding from the Strategic Environmental Research and Development program (SERDP) and USEPA, with support from instrument companies such as Agilent, Waters, Thermo and Sciex etc. A recent publication by

Agilent (Coggan et al., 2019a), for example, includes a single analytical method for the determination of 53 legacy and emerging per- and polyfluoroalkyl substances (PFAS) in aqueous matrices.

Other considerations include sampling and sample preparation for which readers are directed to numerous publications in the literature (Al Amin et al., 2020; Valsecchi et al., 2013; Jahnke and Berger, 2009; Munoz et al., 2019; Berger et al., 2011).

4.2. Measurement challenges – some important considerations

At the workshop, analysts identified a number of key measurement challenges that need to be taken into consideration while analysing for key constituents of PFAS (Rodowa et al., 2020; Shoemaker, 2009; D7979-19, A., 2019). These include:

- Field samplers must avoid contact with Teflon™ and non-stick food wrappers. Field blanks are recommended for water sampling projects.
- Laboratory method blanks must be processed regularly to show all materials used in the analysis, and the entire analytical process, is below the Limit of Reporting (LOR) for PFAS.
- Eluents labelled as high purity and other laboratory products and supplies are all potential sources of PFAS and thus need to be checked prior to use.
- Care must be taken when carrying out TCLP testing. Tumble (mix) in HDPE bottles (no glass). Sampling path must be free of Teflon™ and buffered leach solution must be tested for PFAS prior to using.
- Soil labelled compound recoveries are generally between 80 and 120%. Internal Standard (ISTD) correction yields satisfactory recoveries.
- Fluorotelomers (4:2 FtS (fluorotelomer sulfonic acid), 6:2 FtS and 8:2 FtS) suffer from high recoveries from matrix enhancement.
- 8:2 di PAP (fluorotelomer phosphate diester) – variable recoveries for some water matrices.

4.3. Key analytical gaps seen from an Australian perspective

CRC CARE commenced PFAS research in 2005 with the focus being PFOS and PFOA in fire training waste water. However, it quickly became clear that there was a need for analytical techniques extending beyond these two compounds following the community concern and media publicity surrounding class actions from exposure to PFAS in the USA and the recognition of potential contamination from fire training activities associated with Defence sites. The focus of most PFAS site assessments is on potential risk to human health, and this requires an understanding of the relative importance of a variety of PFAS exposure pathways. Quantifying the full range of PFAS in the various matrices, at varying and often very low concentrations presents considerable analytical challenges (Chropeňová et al., 2016; Houtz et al., 2016; Hu et al., 2016; Anderson et al., 2016). Understanding the potential impact of very low PFAS concentrations on ecosystems is also becoming a concern for many sites, and this presents similar analytical challenges.

Commercial laboratories in Australia are developing analytical methods for environmental matrices such as soil, plants, blood and animal tissue (including aquatic species). However, unlike the USEPA, which places emphasis on standard methods tested and validated for all matrices, there is a significant gap in Australia from this perspective. Different commercial laboratories may adopt different methods for doing the same analysis, as long as the labs have NATA accreditation and participating in proficiency benchmarking. This helps in improving the method development while limitations exist as there is a lag period from method introduction through proficiency testing to method development. The lack of centrally funded resources to develop new and more sensitive analytical methods for the detection and quantification of emergent contaminants is a major challenge.

Key research gaps and where work is needed are:

- **New separation and analytical techniques for PFAS:** sensitive methods of analysis for soil, water, wastewater, food and biological matrices are required to measure the concentrations and speciation of PFAS and the bioavailability of the different contaminant species. Improved separation methods and sensitive bioassay techniques are needed.
- **New standard analytical methods for PFAS in waste matrices:** it is becoming clear that PFAS are not only associated with fire protection services, but they are ubiquitous and will be found in biosolids, trade waste, landfill leachate (Masoner et al., 2020; Hepburn et al., 2019) and process waste streams – in some cases ultra-trace levels will be of interest (Coggan et al., 2019b).
- **Methods for precursor analyses that are indicative of precursor transformations in the environment:** for some sites precursors can make up the majority of the PFAS that are present, and the potential for these precursors to transform to identifiable PFAS with known toxicity characteristics is a key matter in risk assessment and determining the need for remedial action (Eriksson et al., 2017).
- **PFAS dark matter:** the identification and quantification of PFAS 'dark matter' component and constituents that are not identified in standard methods; and linking this with an indicator of precursor transformation in the environment to inform site assessment (Kotthoff and Bücking, 2018).

- **Analytical methods that are fast, robust and cheap:** these methods are needed to identify the most important sources of PFAS on sites and where remedial effort should be focussed, and to determine the pathways of migration from source areas and the extent to which land and waters are contaminated and can pose a risk. In addition, methods to assess the level of contamination resulting in other matrices, including plants, biota, trade wastes, leachates, treated wastewater, recycled water, process waste streams etc. are required to determine the level of risk and the need for remedial or management action.
- **Improved methods that will allow fingerprinting and distinguishing the source of PFAS:** high resolution mass spectrometry might allow for a broader detection of species, targeted and non-targeted methods. Non-targeted analysis (NTA) as a relatively new technique could be used to identify contaminants of emerging concern (CEC) (e.g. various PFAS compounds) in relevant samples to provide mostly qualitative and some semi-quantitative results (Ulrich et al., 2019).
- **Rapid site assessment technologies and probes for real time monitoring:** the need is similar to that for developing analytical methods that are fast, robust and cheap. These can use advanced techniques; for example geophysics and on-site/on-line measurement methods integrated with conventional sampling and analysis to increase the speed and efficiency of PFAS field investigations, offering one pass delineation and continuous longer term monitoring.
- **Identifying and addressing major interferences on PFAS analyses:** understanding the potential for interferences is needed to avoid errors in characterising the extent of contamination; such interferences could be from ionic strength and ionic composition, etc. (Pan et al., 2020);
- **Comparative study of internal standard with isotope dilution method** with respect to sensitivity, reliability and accuracy of PFAS detection and quantification in different extracts.
- **Application of the Total Oxidisable Precursor (TOP) assay to soils:** the TOP assay is widely applied to soils to estimate the presence of PFAS precursors, but there is a lack of information confirming its applicability and reliability. It is unclear if precursors are efficiently extracted from the soil and hence validation is required (e.g. by conducting a fluorine mass balance). Its use in characterising risk is also unclear, as the assay does not necessarily indicate if (or what proportion of) the PFAS precursors will transform to persistent PFAS under ambient environmental conditions (Casson and Chiang, 2018).

5. Eco-toxicological and human health risk assessment

5.1. The challenge

The underlying principle of Australian environmental regulation is to ensure that land and water is fit for use, and that contamination is not present in a form that poses a risk to human health or the environment (Naidu, 2008). The requirement is stringent: there should be no adverse effects. It is a specific intention of regulation to avoid prescriptive remedies and unnecessary cost, and to allow solutions that are appropriate for future use. State and Territory legislation takes this approach, and this has been embodied in national guidance such as the NEPM and NEMP. The application of this approach requires an assessment of risk that contamination poses to human health and ecological systems, extending to a consideration of all potentially contaminated media, and requires an understanding of human toxicology, ecotoxicology, bioavailability, exposure pathways and the fate of contaminants in soil and water (Wang et al., 2015b; Milley et al., 2018).

Risk assessment follows well established methods; however, it is complex. In the case of PFAS the high degree of uncertainty in toxicological criteria and understanding the process of bioaccumulation, makes the overall assessment very complicated. Reaching decisions on the extent of action required in terms of a management response and remediation is difficult and often not well funded, which can lead to organisations not wanting to commit the considerable funds necessary for remediation.

Often the level of uncertainty in the potential for effects that the known PFAS can cause to human health and ecological systems will range over several orders of magnitude – with the uncertainty further increased and complicated by the presence of unidentified and unquantified PFAS with unknown toxicological effects. This uncertainty has resulted in conservative regulatory responses, which in turn have resulted in very large costs to organisations responsible for the contamination, with respect to assessment, remediation, and the potential for litigation by concerned communities. Overly-protective regulatory responses also run the risk of needlessly impacting natural resources and diverting societal attention from other potentially more-substantial matters. Striking the proper balance in addressing PFAS risks will require a concerted and targeted technical and policy effort to reduce and manage uncertainty.

This very high level of uncertainty points to the most serious problem relating to PFAS that our community, regulators and industry needs to solve.

5.2. Eco-toxicological and human health risk assessment gaps in the research

Taking some of the present or proposed policy/regulations into account (e.g. in the EU), the data gaps on PFAS toxicity and its assessment have resulted in very conservative approaches and environmental objectives that cannot be achieved, at least for the existing contamination. These have resulted in unreasonable and unsustainable remediation practices (Simon et al., 2019), which needs to be avoided. For example, Australia Department of Defence's PFAS investigation and

management program is 'possibly the largest programme of environmental investigations ever conducted in Australia'. 'At the time of the Government's submission, the total combined size of the investigation areas was approximately 1150 square kilometres, and Defence had spent in excess of \$130 million on the programme' (Australia, P.o., 2018).

Key research gaps and where work is needed are:

- **Comprehensive information on the fate and effects of residual PFAS compounds in the environment:** The composition and chemical and physical form of contamination is often complex, and current practice is to utilise total concentrations of contaminants in risk analyses. Total concentrations, especially of contaminants that can occur in different matrices, can be of limited relevance, and require that conservative assumptions be made regarding the fate and transport and potential for adverse effects to result from the contamination. The matter is complicated by PFAS having both polar and non-polar characteristics, and there is a wide range of soil chemistries and mineralogies, and groundwater composition, in Australia (Brusseau et al., 2019; Brusseau, 2018; Hunter Anderson et al., 2019).
- **An ability to assess the fraction of PFAS that is bioavailable or bioaccessible:** the risk posed by PFAS to animals/humans or the environment is dependent on the fraction that is bioavailable or bioaccessible. Standard operating procedures are needed to assess PFAS bioavailability in soils and other environments. Leaching tests often show that some soluble PFAS can be leached, suggesting that bioavailability is high. However, PFAS tends to remain present in surface soils, and the leaching potential for PFAS in the environment needs to be confirmed, along with the "unknown PFAS precursors". Mahinroosta and Senevirathna (2020)
- **Agreed methods of risk assessment that can take into account uncertainty and avoid unsustainable conservatism:** risk characterisation requires toxicological data for both aquatic and terrestrial organisms, animals and humans, and for the extent of and effects of bioaccumulation and biomagnification to be taken into account. While procedures are available for aquatic organisms and plants, ethical considerations limit this for animals and humans. Animal models and pharmacokinetic models are applied, but have large uncertainties and the potential to result in highly conservative management strategies.
- **Agreed methods of assessing the toxicity and risk posed to humans and ecological systems by mixtures of PFAS:** evaluation of the toxicity and human health risks posed by PFAS is a major challenge given the many different PFAS compounds that can be present. Methods based on single contaminant toxicities and groups of PFAS, grouped on carbon chain length and the comparative potency of mixtures are being considered. The single contaminant approach is being adopted at present. There is insufficient information for assessing groups of PFAS, and very little information on the toxicity of mixtures including additive and synergistic effects of combinations of PFAS, and the risk posed by short-chain PFAS, fluorotelomers and precursors, both with respect to human health and ecological receptors. As a result of these limitations, there is no clear approach to adopt simpler methods such as treating PFAS as several classes with relative potency schemes, use of surrogates, New approach methodologies (NAMs) or other approaches to infer toxicity/fate based on data from representative compounds are needed.
- **An expanded toxicological database for key PFAS:** the current toxicological database is limited to just a few PFAS; more information is required to properly inform ecological and human health risk assessments. Data that relates to locally-relevant taxa are needed to inform Australian risk assessments, and the setting of water quality guidelines.
- **Exposure pathway assessment:** Australian environmental regulations require assessment of risk to a variety of receptors, but there is insufficient information to complete these assessments currently. Key issues are: phyto-accumulation into food crops (including commercial food and shorter chain PFAS); uptake from soil of PFAS (particularly >C6 perfluoroalkyl sulfonates (PFASs) and >C8 perfluoroalkyl carboxylates (PFACs) into cattle (beef and dairy), sheep and poultry, animal plant food and stock water to ensure compliance with FSANZ (Food Standards Australia New Zealand) trigger points for mammalian meat, offal and milk, inform dietary intakes, and provide for risk harmonisation (ecological and human health).
- **Risk posed to Australian native species:** collation of information is required on food intake rates and feeding preferences of native Australian species likely to have a high food intake to body weight ratio to aid refinement of the guidance values for secondary exposure.
- **Potential impact of PFAS on the transmission of zoonotic diseases to humans:** Previous work has shown that PFAS act as immunosuppressants in birds (Peden-Adams et al., 2009) and detectable levels of PFAS were identified in northern cardinals, a known reservoir host species for West Nile virus (Russell et al., 2019). There is an increasing risk of transmission of certain zoonotic diseases to humans if the avian host is immunosuppressed by PFAS. A similar concern was identified in the Ross River virus in Australia as both marsupials and birds can act as reservoir hosts for the mosquito-borne Ross River virus. Future research can include understanding how PFAS exposure might effect these reservoir hosts, as well as quantifying the potential impact of immunosuppressed reservoir hosts on the risk of human infection.

6. Fate and transport

6.1. PFAS fate and transport processes

The fate and transport of contaminants in the vadose zone and groundwater is an integral part of the conceptual site model (CSM) that identifies the potential contaminant linkages underpinning each stage of a site investigation,

and helps inform the risk assessment process and the requirements for remediation and management. Following entry into the environment, contaminants undergo a series of physico-chemical interactions that include transport within the unsaturated and saturated zones, distribution over the various environmental compartments and potential transformation into other chemicals (Wang et al., 2014b; Prevedouros et al., 2006; Kärrman et al., 2011; Wang et al., 2014c, 2015b; Place and Field, 2012; Lim et al., 2011).

The transport of chemicals can occur within air, water or in soil and mass transfer can occur between compartments or phases (i.e., solid, liquid, gas). Such processes can also include chemical and biological reactions. While it has been commonly believed that only the soil liquid and gaseous phases were mobile and could facilitate the transport of chemicals through the unsaturated zone (Brusseau et al., 2019), it is now accepted that mobile organic and inorganic colloids may facilitate chemical transport. However, the magnitude and significance of colloidal transport processes for PFAS are yet to be determined.

For conventional contaminants, including the petroleum hydrocarbons and chlorinated solvents, modelling fate and transport, and characterising the hydrogeological properties that influence transport, is a well-studied area. However, modelling the fate and transport of PFAS in porous soil systems, especially in field-scale systems, is a far-from-developed science, complicated by the large number of PFAS involved (many of which cannot be identified), and the potential for transformation, and their affinity for phase interfaces.

While the concentrations of PFAS in a receiving environment (such as receiving waters) determine risk, it is the mass flux that flows from source areas that is most important, as it will determine the PFAS concentrations that the receptors are exposed to and the rate of change over time of these concentrations. Mass balances show that the mass in the source area is generally far higher than in the receiving environment, and it is understanding and controlling the mass flux from each source area that is particularly important. In Australia it is also being found that it is often the mass transport in surface water (rainfall runoff) rather than in groundwater that can be important. Recently, research suggests that water reuse and biosolid applications can also contribute to the overall environmental loading of PFAS. That is, it is not just source zones like AFFF training areas/airports etc. Since the chemicals are very persistent, they can cycle through and then accumulate in crops/dairy etc.

Several factors can be noted: the mass flows of PFAS generally depend on the concentrations in the source area, the equilibrium relationship between adsorbed and dissolved phase PFAS, and the rate of the equilibration processes. In the case of PFAS, this is complicated by the fact that some PFAS are cationic and zwitterionic and thus sorb strongly to negatively charged soil particles, and there can also be co-contaminants that will affect the availability of binding sites and hence increase the rate of transport (e.g. from the unsaturated into the saturated zone) (Seow, 2013; Gao and Chorover, 2012; Du et al., 2014; McKenzie et al., 2015). Recent work suggests that the retention behaviour of PFAAs is determined by their strong surface activity, and the solid-water distribution coefficient (K_d) may not be a good predictor of retention in the unsaturated zone, with additional retardation occurring in the unsaturated zone through adsorption at air-water interfaces, partitioning to trapped non-aqueous phase liquids (NAPL) (i.e. hydrocarbon fuels), and adsorption at NAPL-water interfaces. Conceptual models are being proposed.

6.2. The influence of co-contaminants on fate and transport

The complex interactions of PFAS with soils, sediments and groundwater limits the application of currently available predictive models. Although persistent in groundwater and surface waters, some limited studies indicate that key active ingredients may partition to organic matter rich sediments and soil particles due to their propensity to sorb to natural organic matter, and at appropriate pH conditions, to soil mineral surfaces (Du et al., 2014). The adsorption/accumulation of PFAS at the air-water interface influences the retention of PFAS (e.g. PFOA) in the unsaturated zone, and this could be further complicated by the presence of co-contaminants and PFAS mixtures. Additionally, non-aqueous phase liquid (NAPLs) and other non-fluorinated AFFF surfactants could affect PFAS sorption (McKenzie et al., 2016; Guelfo and Higgins, 2013). This particularly applies in the case of long-chain species, although the process of PFAS partitioning at the liquid-liquid interface (aqueous and NAPL) is not well understood. Thus, there is potential for several processes to affect PFAS retention and retardation in the subsurface, including: solid-phase adsorption, adsorption at the air-water interface, partitioning to the soil atmosphere, adsorption at the NAPL-water interface, and partitioning to NAPL.

The fate and transport of PFAS are further complicated by the large number of PFAS that are present in the complex AFFF formulations and by the differences between linear and branched forms (isomers) of some PFAS (Du et al., 2014; Wang et al., 2017; D'Agostino and Mabury, 2014). The simultaneous presence of these varying compounds is likely to strongly influence the fate and transport behaviour of PFAS. The propensity of individual fractions to partition onto soil differs hence there is separation of compounds, particularly in dissolved plumes. However, such critical information is significantly lacking in the literature. This is further complicated by the presence of non-PFAS co-contaminants in soil and groundwater, such as heavy metal(loid)s and petroleum hydrocarbons. In addition, much of the research conducted to date used freshly spiked soils which may not reflect long-term field contaminated sites. A greatly improved understanding of the underlying physico-chemical processes that control the overall fate and transport behaviours in the surface, including source zone depletion, is critical for the formulation of conceptual site models and for risk assessment modelling.

6.3. Fate and transport of PFAS – research gaps

As discussed above, unlike conventional contaminants, the fundamental information needed for determining the transport, fate and short- and long-term toxicity of most PFAS is limited. This is further challenged by the large number of PFAS constituents that can be present, each with a different molecular structure (including branched vs. straight chain isomers). The lack of understanding of the physical–chemical properties of each of the constituent compounds and their biogeochemical interactions, including competing interactions for the binding sites, precludes establishing reliable models that will predict the transport and fate of PFAS, and how PFAS composition will change with transport distance and contaminant age.

Key research gaps and where work is needed are:

- **Understanding the release and transport of PFAS to surface water vs. groundwater in source areas.** The mass flux in surface water that occurs from source areas is a critical matter for many sites, both with respect to determining the risk posed to receptors, and what can be an effective approach to minimising the mass flux and hence form a major component of the remedial strategy. Despite PFAS surfactants being relatively soluble, a surprising amount can be retained in surface soils. There is a need to understand the fundamental factors that determine the rate of release of various PFAS from contaminated soil, concrete and sediments into rainfall runoff and overlying water, vs. that which leaches into groundwater. The rate of migration can depend on the soil water content and porosity, the degree to which the water contacts the soil, and the kinetics of sorption and rate of release across the soil–water boundary. Complicating factors that need to be understood include the range of PFAS constituents including precursors, the effect of the age of the contaminants on adsorption and release and the potential for transformations to occur, the effect of their ionic charge (zwitterionic and cationic) and co-contaminants on release and transport, the effect of soil properties including salinity, pH and organic matter, and the relative rate of transport and possible transformations (e.g., abiotic reactions) of precursors. Australian-specific information for leachability from soils and understanding the tests that are most relevant to determine leaching, are of interest. Also of importance is the rate of depletion of PFAS that occurs in surface soils and in the source area more generally through these various processes, and the role that this plays in affecting the release into surface water.
- **Understanding transport in surface water.** After PFAS have been released and is transported in surface water runoff, an understanding is needed of how the PFAS is transported in the water, whether in the form of dissolved phase or attached to soil particles, and how this relates to the concentrations observed in sediments along drainage lines and creeks. Conversely, an understanding is needed as to the significance of PFAS measured in sediments along drainage lines, and how this may be related to and provide estimates of PFAS mass flux that occur in time varying and ephemeral drainage lines.
- **Understanding transport in groundwater.** After PFAS have leached into rainwater (or firewater), it can migrate vertically through the vadose zone and then flow with the groundwater. A better understanding is required of the factors determining the rate of leaching and transport of the various PFAS in the sub-surface, including the effect of soil geochemical properties, salinity, organic content, and co-contaminants. Other factors can include the use of freshly spiked soils to characterise transport behaviour, which may not reflect the characteristics of historic contamination. The rate of source zone depletion and reduction in groundwater concentrations over time can also be important in determining the acceptability of allowing residual contamination to remain in place.
- **Understanding the transport and dispersal processes that occur in urban areas.** It is important to understand the interplay between airborne transport, surface flow, and groundwater migration. The combination of these multiple transport mechanisms, along with the presence of numerous dispersed sources in the urban setting (small industrial sources, spills, historical fire-fighting, waste discharge, septic tanks etc.), poses a challenge for regulators and contaminated land consultants in interpreting contamination sources and setting realistic remediation targets. This will allow a better understanding of the nature and origins of urban ambient background PFAS, and will also be of value to inform ways to improve management/mitigations of urban PFAS sources.
- **Monitoring data to allow understanding of off-site transport.** In the absence of good models for fate and transport, a body of monitoring data is needed of PFAS contaminated sites that will allow estimates of the rates of off-site transport of PFAS in surface waters and groundwater to be determined, together with related information that will allow the rates of release and kinetics to be determined for different inundation situations and soil types.
- **Information on the surface activity of PFAAs.** Information is needed on retardation and transport in the unsaturated zone and potential adsorption at air–water interfaces, partitioning to trapped non-aqueous phase liquids (such as hydrocarbon NAPL and chlorinated solvent DNAPL), and adsorption at NAPL–water interfaces (Guelfo and Higgins, 2013). Research is needed to better understand these additional retention processes in the unsaturated zone at the field scale and to incorporate these processes into fate and transport models.
- **Transport and fate models for WWTPs.** There is a need to better understand the fate of PFAS in WWTPs, including the relative amounts partitioned to biosolids vs. that in the effluent, and the transformations that occur within the WWTP. Information is also required on the concentrations of PFAS that are typically present in the influent to WWTPs (i.e. the amount contributed by the urban catchment and any trade waste), the specific PFAS present, the concentrations and risk posed by PFAS in treated effluent that is reused (e.g. irrigation and stock), in WWTP outfall discharges to receiving waters, and in the reuse and disposal of biosolids and biosolids leachate (Coggan et al., 2019b; Gallen et al., 2018; Hamid and Li, 2016).

- **Developing more reliable conceptual site models for PFAS contaminated sites.** Developing a conceptual site model (CSM) that is able to explain the various fate and transport processes and the variation of PFAS over distance and time is critical to determining the risk that PFAS will pose to human or ecological receptors, and in developing an appropriate management response. At present conceptual and mathematical models are simplistic and mainly focus on just one or a few PFAS, and solid-phase sorption in the saturated zone as the sole source of retention. Better models are required, including numerical models that can be used to describe the fate and transport of various PFAS and mixtures of PFAS in the subsurface in variably saturated source zones and in the presence of co-contaminants. Parameterising such a model would require detailed laboratory experiments to determine or estimate key distribution variables between the solution phase and the solid matrix (Hodgkins et al., 2019).

7. Remediation and risk management

7.1. Challenges in the remediation and management of PFAS contamination

Like sites contaminated by more conventional contaminants, remediation of PFAS contaminated sites can require treatment or management of both the impacted soil and ground water (Washington et al., 2009; Liu et al., 2012; Vecitis et al., 2009). Often the strategy for PFAS contaminated sites will be to delineate the source area that is important in contributing to off-site contamination and the risk posed to receptors, and to treat or contain this source area. If the off-site risk is such that it requires an urgent response, then interception of PFAS closer to the receptors (such as at the site boundary) may be required or, in the case of groundwater, point of use treatment or provision of alternative water supplies.

While simple in concept, there are considerable challenges in identifying and gaining approval for practical and cost-effective remedial strategies. These challenges include:

- (a) Currently only a subset (<30) amongst the large number of possible PFAS (>5000) can be quantified. PFAS other than those quantified may be present or confirmed indirectly by use of the TOP Assay (or may become present in the future through transformation), and their contribution to risk and therefore the requirements for remediation or management of these other PFAS are uncertain. To assess and better manage risk requires significant investment into animal feeding studies especially for species with low depuration rates.
- (b) With respect to contaminated soil, the extreme stability of key PFAS (including PFOS and PFHxS) and the inability to destroy these PFAS in-situ in soil to achieve concentrations that would be deemed to be acceptable to remain in place. Excavation and ex-situ soil washing of excavated contaminated soil is a developing technology, but some residual PFAS will remain and this may limit re-use of the treated soil. In addition, treatment of the resulting washwater to remove PFAS is required. Excavation of soil and off-site disposal by landfill is possible but practically limited because of the limits placed on the acceptance of PFAS contaminated soils in landfills, with leaching potential being the most limiting characteristic. Containment of contaminated soil in-situ or in an on-site repository can be a practical and acceptable strategy, and is often being selected as the most practicable option. However, there is the concern that in the future exhumation and treatment of the contained material may become necessary, and this is an important consideration.
- (c) With respect to contaminated groundwater, in-situ treatment of groundwater remains generally impractical other than perhaps through the addition of immobilising agent (e.g. PlumeStop™), although some developing methods are being evaluated in field trials. There are well proven methods available for treatment of groundwater ex-situ; these generally involve separation (e.g. sorption by adsorbents such as granular activated carbon and ion exchange resins following necessary pretreatment).
- (d) The lack of guidance on end points for soil and ground water remediation, and for the reuse or disposal of contaminated soil and water.
- (e) The large areas over which PFAS may migrate in surface water and groundwater, and the inability to cost-effectively treat or control PFAS that are present off site in large areas at low concentrations in soil and waters.
- (f) The ubiquitous nature of PFAS, and uncertainty as to whether the PFAS that are observed off site originates from the site of interest or from other sources.

A recent research symposium held in USA recommended management of PFAS contaminated sites until technologies are developed that offer a practical and cost effect option for treating contaminated soil. This symposium also recognised the need for a combination of technologies to provide for an effective remediation and management strategy.

7.2. Approaches to PFAS remediation

Numerous remediation approaches for PFAS currently exist, mostly at laboratory scale, including sorption, immobilisation, stabilisation, filtration, coagulation, separation, chemical oxidation/reduction, thermal decomposition, UV/photocatalytic degradation, etc. (Vecitis et al., 2009). Early work conducted (2006 to 2009) by CRC CARE (unpublished) on bioremediation of PFAS contaminated soils was not successful. However, numerous studies aimed at bioremediation

have been funded by SERDP in US. All these remediation approaches can be generally categorised into two types, non-destruction (removal, such as sorption) and destruction (degradation, or mineralisation).

The sorption approach transfers PFAS from one matrix to another where the PFAS are present at much higher concentration, and in a much smaller volume (Yu et al., 2009; Wang et al., 2012). The smaller volume offers the advantages of a reduced cost for destruction (e.g. by incineration), which would not be possible in its original dispersed form. This is the approach adopted in water treatment where the PFAS is adsorbed on an adsorbent such as granular activated carbon (GAC) or ion exchange resin (IX). It is also applied in soil washing.

The second approach, degradation or destruction, has the advantage of destroying the PFAS, but because this requires high energy methods (such as incineration), it is costly and generally not practical other than where PFAS are in concentrated form (Du et al., 2014; Vecitis et al., 2009; Merino et al., 2016). While some degradation can be achieved by severe chemical conditions, this tends to be limited to the more complex incompletely fluorinated compounds, with these degrading to fully fluorinated carboxylates. When destruction is carried out, it is necessary to confirm that complete mineralisation has occurred through a mass balance of fluorine and analysis to determine the absence of fluorine-containing byproducts. Because analytical methods may not be able to identify and quantify these byproducts, it can be difficult to confirm that destruction is complete and unwanted byproducts have not been formed (Lei et al., 2020).

The following sections present an overview of some of the techniques currently being developed for soil and ground water; the outline is not complete and is intended to provide a general indication of methods that are being applied or are being considered. Some of these were not discussed in the workshop, and have been added to this summary.

7.3. Sorption: non destruction

7.3.1. Water

The sorption-based technology (e.g. GAC and IX) dominates the current remediation approaches in the field, and treatment of contaminated water (such as groundwater) can reduce the concentrations to less than trace levels of detection (Seow, 2013; Du et al., 2014). This approach has been used to treat groundwater that forms a drinking water supply at Australian Defence bases and contaminated surface water at industrial and airport sites. Important considerations in the application of adsorbents are: the efficiency of removal, the loading that is able to be achieved on the sorbent, and what needs to be done to destroy the adsorbed PFAS. In the case of IX, the IX may be able to be regenerated and the adsorbent reused, and the PFAS further concentrated, thus reducing its cost for destruction. However, regeneration can be difficult, involving a solvent (such as methanol), and it may be preferred to simply use resin (and GAC) on a once-use basis, with direct destruction when exhausted.

7.3.2. Soil

For PFAS-contaminated soil treatment, soil-washing is being developed to separate PFAS from the soil, and then from the wash water, resulting in the PFAS being in concentrated form (e.g. on an IX or GAC) where it can be more cost effectively destroyed. The cost effectiveness of soil washing is dependent on the fines content of the soil to be washed, with a high fines content (e.g. clay soils) potentially making separation more difficult and dewatering of the washed slurry more costly. The residual concentrations and form of the treated soil, and whether it is suitable for reuse or requires landfill disposal, are also important factors determining the applicability of this method.

7.3.3. Proprietary adsorbents – soil

Another option is to add an immobilising agent to the soil, which will greatly reduce the potential for PFAS to leach from the soil. Rembind™ and matCARE™ are two such immobilising agents. The objective of this type of treatment is to sufficiently reduce the leaching potential to allow disposal of the treated soil (e.g. by landfill). The cost of the immobilising agent and uncertainty as to the long-term stability of the immobilising agent and providing confidence that PFAS will not be released at some time in the future are important factors in the successful application of this technology.

The matCARE™ material is a novel adsorbent available in granular or powder form, formulated by surface modification of mineral palygorskite, for the specific adsorption of PFAS (Das et al., 2013). The natural clay base (molecular structure) has been modified to achieve a high removal efficiency and high loading affinity. matCARE™ can be used both for immobilisation of PFAS in soil, and as an adsorbent for water treatment, like GAC and IX. PFAS are irreversibly loaded to the adsorbent matrix but are not destroyed; if destruction is required then incineration or other high temperature method will be required.

Rembind™ is a similar product with a different composition; similar considerations apply.

7.4. Destruction – PFAS in water

7.4.1. Electrochemical destruction

Electrochemical destruction of PFAS in water is being trialled in laboratory and pilot scale work has been reported at conferences (Söregård et al., 2019; Li et al., 2011; Lin et al., 2013; Zhuo et al., 2016; Schaefer et al., 2017; Le et al., 2019; Schaefer et al., 2019). Destruction of PFAS in water is difficult to achieve, as some PFAS are very resistant to destruction. Long residence times can be required to achieve high levels of destruction treatment. Even though the primary PFAS

subject to regulation may be reduced to very low concentrations, intermediates or by-products can be generated if the treatment is not complete or the reaction products generate other compounds. These intermediates and by-products may be stable and more toxic and mobile than the PFAS subject to regulation. In general, achieving satisfactory levels of destruction is likely to be unachievable for field applications where reuse of the treated water or its discharge to the environment is proposed, which requires extremely low concentrations for acceptance. Electrochemical destruction may offer a method of reducing the concentration of PFAS in applications where there is a subsequent removal step (such as by IX, and it is the latter step that achieves the required endpoint), or as a step to reduce the concentration of PFAS in a concentrated wastewater stream, and it is not intended to seek to discharge the treated wastewater to the environment (e.g., if the wastewater is recycled). Whether applying an electrochemical destruction step offers advantages over a single separation/adsorption process is uncertain, and detailed testing will be required to determine effectiveness.

In general, achieving and validating the level of destruction can be difficult, and seeking to complete mass balances and account for fluorine before and after treatment can be uncertain in view of the potential for unidentified fluorine-containing compounds, including short chain fluorine compounds, to be present. Use of analytical methods such as TOF (total organic fluorine) may assist, but TOF itself is neither well established nor standardised, and may not have a limit of reporting low enough to meet remediation objectives.

Other complicating factors that need to be resolved include: whether the destruction efficiency will be lower or the kinetics will be slower, due to other substances present in the water to be treated (such as other unidentified PFAS, or other inorganics, organics, or biological matter); whether “saturation” (no more destruction occurs after a prolonged time, particularly when the PFAS concentrations have become very low); and whether the electrode is subject to fouling or poisoning.

7.4.2. Electrochemical advanced oxidation

The advanced oxidation process (AOP) has received attention in recent decades (Sirés et al., 2014; Petrie et al., 2015; Lin et al., 2018). It uses radicals, typically hydroxyl radicals ($\bullet\text{OH}$), to attack the organic contaminants and degrade them to fragments or inorganic ions/molecules, towards mineralisation (Fang et al., 2017).

However, the AOP requires chemicals or other inputs (such as UV, catalysis) to generate radicals. The Electrochemical Advanced Oxidation process (EAOP) can potentially overcome some of these limitations because the radicals can be generated *in-situ* by the process of electrochemical oxidation. The reduced consumption of chemicals in EAOP has potential to offer a more environmentally-friendly approach. In addition, the electrochemical process can directly oxidise the PFAS or generate other radicals beyond $\bullet\text{OH}$. However, it can be hampered by expensive electrode materials and surface fouling, although it has been highlighted as “promising” for PFAS destruction.

CRC CARE has developed and patented pfasCARETM to effectively destroy PFAS using a cheap electrode material and a combined technology to minimise fouling (Fang et al., 2018). Laboratory test work is encouraging, and further research is required to confirm that the process can be scaled-up and applied commercially.

7.4.3. Sono-chem

The basis of sono-chemistry is the super-sonication-generated micro-bubbles (cavitation and plasma), which can result in localised temperature of $> 5000\text{ }^\circ\text{C}$, at which PFAS can be effectively destroyed (Lei et al., 2020). In addition, the process can generate highly reactive radicals ($\bullet\text{OH}$). The combined processes can result in the complete mineralisation of PFAS to carbon dioxide and fluoride ions.

Challenges in the application of Sono-chem include confirming that short-chain PFAS are destroyed and do not escape through vaporisation, and how to scale up the method for field application, and to apply it for soil remediation (Vecitis et al., 2008).

7.5. Destruction – PFAS in soil

Destruction of PFAS in soil requires a very high energy method, such as is achieved at an elevated temperature. Thermal destruction usually refers to the combustion of PFAS at high temperature and with sufficient residence time ($> 1200\text{ }^\circ\text{C}$, $> 2\text{ s}$). An alternative is to apply thermal desorption at lower temperatures (perhaps $550\text{ }^\circ\text{C}$), with subsequent destruction of the evolved gases at high temperature ($1200\text{ }^\circ\text{C}$). Incineration is generally not considered to be acceptable in Australia, whereas thermal desorption has been applied widely for the treatment of contaminated soil.

There is potential to improve the destruction process by combining it with sorption, with recent research showing that destruction can occur at $< 500\text{ }^\circ\text{C}$, if the PFAS are adsorbed onto a matrix surface. This suggests that remediation may be able to be achieved by removing PFAS by sorption, followed by subsequent destruction at a reduced temperature. This is similar to the thermal regeneration and reactivation of activated carbon that has been used to treat PFAS contaminated water. Challenges in the application of this process is determining the conditions or adsorbent that will avoid the generation of gaseous intermediates such as Freon or HF (Wang et al., 2015a).

7.6. Bioremediation

Although bioremediation has proven to be an effective and sustainable green technology for several organic contaminants, in particular the petroleum hydrocarbons and several pesticides, this technology has so far proven to be very ineffective for the PFAS due to their strong C–F bonds. There have been several attempts by researchers to biodegrade PFOS and PFOA and isolate the bacteria able to breakdown the PFOS and PFOA. A few studies using activated sludge, soils pure and mixed bacteria cultures have shown partial degradation of PFOS, PFOA, PFHxA (perfluorohexanoic acid), PFBA (perfluorobutanoic acid) etc. (Liu and Avendano, 2013). So far, the literature shows biodegradation of PFAS is mostly limited to the molecules, or the regions within the molecules that are not fully fluorinated, such as fluorotelomer, compounds (Liu and Avendano, 2013; Ochoa-Herrera et al., 2016). However, the recent report on the substantial defluorination (60% of 100 mg/L) of PFOS and PFOA by pure and enriched novel bacterium *Acidomicrobium* sp. strain A6 capable of anaerobically oxidising ammonium under iron reducing conditions is a break-through in PFAS bioremediation research (Huang and Jaffe, 2019). This finding is a step in the right direction for PFAS bioremediation and shows some promise in the future for bioremediation of PFAS through anaerobic defluorination by bacteria.

7.7. Remediation: Challenges and research gaps

While several remediation techniques have been demonstrated in the laboratory, their extension to a commercially available full-scale remediation method poses a significant challenge. Other than thermal destruction at high temperature, soil treatment may result in the formation of ill-defined by-products with ill-defined toxicological criteria, and the possibility that such by-products will be more mobile and present an unacceptable risk. Treatment of groundwater is possible, but is challenged by the uncertainty of the PFAS present, uncertainty in the effectiveness of pump and treat options, uncertainty in the degree of treatment required to achieve an acceptable risk level, uncertainty regarding the form and mobility of residuals, and uncertainty regarding the ability to dispose of more concentrated PFAS wastes that result from treatment.

Key research gaps and where work is needed are:

- **Soil and groundwater: endpoints for remediation:** acceptable endpoints for remediation and/or reuse or disposal of soil and groundwater containing PFAS are needed but have yet to be developed. These are critical requirements for determining the requirements for remediation and management, and the methods that are able to be applied.
- **Soil and groundwater: guidance on Clean Up to the Extent Practicable, Technical Impracticability and what might accord with the principles of sustainable remediation.** while guidance and a body of practice have been developed for establishing whether it is impracticable to carry out further clean up for conventional contaminants, such guidance and a body of practice does not exist for sites with PFAS contaminated soil and groundwater. Critical considerations are what constitutes an acceptable level of risk for a remediated site, whether the risk level needs to be similar to that on which screening levels are based, or whether alternative risk measures such as are applied in the USA and the Netherlands, are applicable. This guidance needs to be developed.
- **Groundwater: pump and treat, and beyond pump and treat:** pump and ex-situ treatment is commonly being considered as a component of the remedial strategy for PFAS contaminated groundwater. Information is required on the efficacy of pump and treat for the purposes of hydraulic containment, or perhaps as a remedial strategy, what can be practicably achieved over what timeframe, and the influence of the site-specific source and hydrogeological parameters.
- **Information on remediation strategies and the results of the application of these strategies.** The results of the application of different remedial strategies and technologies needs to be made available, and the applicability and limitations of strategies and technologies are explained. Under the Australian audit system, such information can be expected to be published and available, but the relevant factors need to be drawn out and explained.
- **Information on new and developing technologies.** There is a continuing body of work being carried out on new technologies; however much of this is laboratory based and has not reached a stage where technologies can be applied commercially. It would greatly assist the industry if regular reviews of new technologies could be carried out and made available.
- **Bioremediation of PFAS.** Comment: The limited research on biodegradation of PFAS has shown partial transformation of PFAS with several unknown products. More in depth biodegradation studies on PFAS, in particular focusing on defluorination are required. Although there is some progress recently, it seems there is a long way to go prove at field-level.

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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