

# Modelling PFAS transport in unsaturated soil using LEACHM and implications for source zone remediation

By

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> Thesis Submitted to Flinders University for the degree of

Master of Science (Groundwater Hydrology)

College of Science and Engineering 18 March 2024

AUGUST 21, 2023

# MODELLING PFAS TRANSPORT IN UNSATURATED SOIL USING LEACHM AND IMPLICATIONS FOR SOURCE ZONE REMEDIATION

Master of Science (Groundwater Hydrology) Thesis

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#### DECLARATION

I certify that this thesis does not incorporate without acknowledgment any material previously submitted for a degree or diploma in any university; and that to the best of my knowledge and belief it does not contain any material previously published or written by another person except where due reference is made in the text.

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Colin McKay 21 August 2023

## STATEMENT OF PROJECT INTELLECTUAL PROPERTY

This thesis represents a novel body of scientific work that has been prepared by the candidate under direction of their supervisors. The final dissertation presented herein is work of intellectual merit that has been produced by various inputs as follows:

| Funding acquired by:                   | N/A No funding provided                               |
|--|---|
| Project Concept by:                    | Ilka Wallis (supervisor)                              |
| Project Design by:                     | Candidate + Ilka Wallis and John Hutson (supervisors) |
| Laboratory work assisted by:           | N/A No laboratory work                                |
| Modelling program assistance:          | John Hutson (supervisor)                              |
| Field work done by:                    | N/A No field work                                     |
| Data processed by:                     | Candidate   |
| Thesis written by:                     | Candidate   |
| Illustrations, photography by:         | Candidate   |
| Input to shaping thesis drafts by:     | Ilka Wallis and John Hutson (supervisors)             |
| Inputs to methodology:                 | Ilka Wallis and John Hutson (supervisors)             |
| Intellectual discussions into results: | Ilka Wallis and John Hutson (supervisors)             |

The candidate acknowledges that authorship of any resulting publication from this work will be guided by advice from the supervisors and may include other authors who contributed intellectual merit in the project.

When Mkay

Colin McKay (Candidate) 21 August 2023

### FORMAT OF THESIS

This thesis has been written as a journal article and is therefore adhering to a short format following (Journal of Contaminant Hydrology) instructions to authors (see Appendix F). However, for readability the tables and figures are inserted within the text.

The table of contents (follow page) presents appendices that have been included.

#### PERSONAL ACKNOWLEDGEMENTS

This project is part of a Masters in Groundwater Hydrology I am conducting with the College of Science and Engineering at Flinders University. I would like to express my sincere appreciation and thanks to Associate Professor Ilka Wallis and Dr John Hutson for their support and encouragement as my supervisors.

Thank you Ilka for your inspiration for the project, recognising my need for a project that accommodated distance education and allowed for part-time research. Thank you also for your immense patience advice and direction throughout.

Thank you John for your invaluable support in setting me up with a copy of LEACHM and patiently helping me understand how to get the most out of it. It is a wonderful program you have created. Thank you also for your suggestions, guidance and troubleshooting advice.

It has been a pleasure working with you and learning from you both!

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#### SUPPLEMENTARY MATERIAL

Appendix A: Climatic Graphs

Appendix B: Calculations of Selected Model Parameters

- Appendix C: Water Balance Data and Graphs
- Appendix D: Untreated Flux Graphs, by Rainfall and Soil Type
- Appendix E: Treatment response graphs

#### **Appendix F: Vertical Section Time Series Pictorials**

## Appendix G: Journal of Contaminant Hydrology - Instructions to Authors

# Modelling PFAS transport in unsaturated soil using LEACHM – implications for source zone remediation.

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- 10 Highlights
- Soil type, rainfall and sorption affect transport of PFAS in unsaturated soil
- Stabilisation of PFAS is effective at reducing flux for material that is treated.
- PFAS residues beneath stabilised soil can sustain ongoing vertical PFAS flux.
- Capping may be more effective than limited stabilisation.
- Combined cap and stabilisation has limited additional benefit for vertical flux.
- 16 Abstract

The properties of per- and polyfluoroalkyl substances (PFAS), and environmental factors affecting their fate and transport in groundwater have been extensively researched. However, relatively few studies have focused on migration of PFAS in the unsaturated zone. Understanding of PFAS

behaviour in the vadose zone is particularly important for remediation planning at source sites, as

21 the bulk of PFAS mass is often present in shallow horizons.

This study was conducted to assess the relative effects of climate and soil type on the vertical migration of PFAS compounds PFOS, PFHxS and PFOA across a range of published soil sorption coefficient (Kd) values. The study also simulated PFAS flux following application of various management options to assess their efficacy over time compared with a "do nothing" approach.

The model outputs reinforce that soil type, rainfall and sorption properties govern the PFAS flux beneath source zones. Both stabilisation and capping, as remediation technologies, were effective at reducing the rate of vertical flux to groundwater. PFAS stabilisation was found to be very effective in reducing flux, for the material that was treated. However, any residual PFAS within the affected soil profile below the treated material was shown to continue to sustain vertical PFAS mass transfer into the future. The relative effectiveness of partial profile stabilisation appeared to decrease with increasing sand content, further exacerbated in soils of high sorption capacity.

In contrast, clay capping, controlling infiltration, was effective from the surface to depth, and in

34 sandier soils, was more effective than partial profile stabilisation alone. Generally, a combination of

- 35 partial profile stabilisation and clay capping was of little additional benefit compared to providing
- the cap alone.

#### 37 Key Words

Per- and polyfluoroalkyl substance (PFAS), numerical modelling, unsaturated zone, source zone
 remediation, stabilisation, capping

#### 40 1 Introduction

#### 41 **1.1 Background**

Per- and polyfluoroalkyl substances (PFAS) are chemicals that have variable length carbon chains
that are partially or completely fluorinated. PFAS chemicals are an anthropogenic group of
chemicals that have been used on a global scale in a wide range of industries (Glüge et al., 2020).
They are to be found in a wide range of household products and were a crucial component of
firefighting foams that were used globally as well as throughout Australia at defence sites,
commercial airports, fire stations, and oil terminals since the mid 1970s (Australian Government

48 Department of Health, 2019, Banwell et al., 2021).

49 PFAS includes an extremely broad range of chemicals (Buck et al., 2011) that includes

50 perfluoroalkyl acids (PFAAs) such as perfluoroalkyl carboxylic acids (PFCAs) and perfluoroalkane

51 sulfonic acids PFSAs) among others, a wide range of PFAA precursors (such as fluorotelomer

52 based substances), and countless other fluropolymers. Three of the most widely studied PFAS

53 compounds include perfluorooctane sulfonate (PFOS) and perfluorohexane sulfonic acid (PFHxS),

54 which are both PFSAs, and perfluorooctanoic acid (PFOA), a PFCA (Wang et al., 2017). These are

55 commonly found within legacy firefighting foams at both commercial and defence facilities.

56 PFAS compounds exhibit some unique physical and chemical properties that are generally

<sup>57</sup> unmatched by non-fluorinated products (Buck et al., 2012, Krafft and Riess, 2015). The fluorinated

58 hydrocarbon chain provides extreme hydrophilic properties making these compounds excellent

<sup>59</sup> water repellent surface treatments (Hill et al., 2017). The combination of the hydrophobic tail and

60 the polar functional groups make them excellent surfactants and foaming agents that together with

61 the fluorinated features result in PFAS compounds being extremely stable and chemically inert, and

62 able to perform under high temperatures – ideal for firefighting applications (Buck et al., 2012).

63 PFAS, however, have also been used in a myriad of other commercial and industrial processes and

64 products including in cosmetics, textiles, paints, printing and photography, fabrication of bearings

and seals, cables, electronics and biomedical uses (Prevedouros et al., 2006, Krafft and Riess, 2007,

66 Heads of EPAs Australia and New Zealand, 2020).

67 Since the late 1990s PFAS as a class of chemicals, and various individual component chemicals,

have increasingly gained global public attention due to concerns regarding their potential toxicity

and known persistence in the environment (Ahrens and Bundschuh, 2014, Wang et al., 2017). In

2009, the Stockholm Convention on persistent organic pollutants (POPs) listed PFOS in Annex B

(meaning parties must take measures to restrict production and use of the chemical), and in 2019

and 2022 PFOA and PFHxS were respectively also added to Annex A (meaning parties must take

measures to eliminate production and use of the chemical) (United Nations Secretariat of the

74 Stockholm Convention, 2022)

As a result of their relatively low sorption, high solubility, and extreme persistence (Prevedouros et

al., 2006, Buck et al., 2012) PFAS plumes in surface water and groundwater emanating from fire

training facilities can be extensive and affect communities many hundreds of metres or even

- kilometres from source zones (Krrman et al., 2011, Filipovic et al., 2015, Bräunig et al., 2017,
- AECOM, 2017, Coffey, 2018a, Coffey, 2018b, WSP, 2018). In addition, PFAS compounds with
- 80 long perfluoroalkyl chains have been found to bioaccumulate and biomagnify in animals and
- 81 humans (Martin et al., 2013, Ahrens and Bundschuh, 2014, Krafft and Riess, 2015).

82 A substantial body of research has been conducted in the area of PFAS properties and their

- environmental fate and transport (Ahrens and Bundschuh, 2014, Du et al., 2014, Hamid et al., 2018,
- Li et al., 2018). However relatively few studies have focused on the vadose (or unsaturated) zone of
- soils, and how the environment of this zone affects PFAS migration to groundwater. This
- 86 unsaturated zone, in the vicinity of source sites, is particularly important for remediation planning
- as it is where the bulk of PFAS mass is often found (Høisæter et al., 2019, Adamson et al., 2020,
- 88 Wallis et al., 2022). This residual PFAS at source zones provides an ongoing risk to the
- 89 environment as it sustains groundwater plumes and surface water pollution (Dauchy et al., 2019).
- 90 In the Australian context, Defence, Air Services Australia, state fire and rescue services and oil
- 91 terminal operators have in recent years been moving from investigation phases at their PFAS
- 92 affected sites, to remediation programs. Due to the often widespread nature of impacts, and
- relatively low concentrations in plumes, remediation focus has generally been on management of
- source zones such as fire training grounds and fire station sites rather than remediation of diffuse
- downgradient plumes (Air Services Australia, 2023b, Australian Government Department of
- 96 Defence, 2018, Australian Government Department of Defence, 2019a, Australian Government
- 97 Department of Defence, 2019b).
- When evaluating remediation options, the environmental, economic, and social sustainability of the available options should be considered. Part of this process is to evaluate the benefits and effects of
- 100 the available technologies (National Environment Protection Council, 2013). Remediation of PFAS
- in soils is a relatively new and evolving challenge (Grimison et al., 2020). There are many emerging
- technologies, but proven treatment options are currently limited in capacity and demonstrated
- ability to meet chosen regulatory targets (Interstate Technology and Regulatory Council (ITRC),
- 104 2020). In fact, there is no clear leading technology to date.
- 105 The Australian Government's National Environment Protection (Assessment of Site Contamination
- 106 Measure (NEPM), 1999 (as amended 2013) provides a national framework for assessment of
- 107 contamination to ensure sound environmental management (National Environment Protection
- 108 Council, 2013). Whilst the NEPM hierarchy of remediation options places onsite or offsite
- 109 destruction above onsite management, the reality is that destructive technologies are at present quite
- 110 costly to implement with few proven at field scale (Grimison et al., 2020, Heads of EPAs Australia
- and New Zealand, 2020). Consequently, onsite management to reduce risk is often considered as
- part of a strategy for interim PFAS management. Onsite management often includes stabilisation
- (the practice of adding an amendment to the soil to reduce its leachability), such as through mixing
- the soil with activated carbon (McDonough et al., 2022, Niarchos et al., 2023, Navarro et al., 2023)
  or traditional hydraulic containment using capping technologies (Newell et al., 2022).
- 116 Stabilisation can be undertaken as an in-situ process by injection of activated carbon slurry, but in
- shallow source zones is commonly achieved by excavation of the soil, mixing with the amendment,
- and then placing the stabilised soil back into the excavation. Capping, in its simplest form involves
- importation of a low permeability clay that is spread and compacted over the waste to reduce

- infiltration. The surface is generally graded to provide a slight slope to aid in shedding of surfacewater.
- 122 This study was conducted to assess the relative effects of various physical and chemical parameters
- 123 on the vertical migration of PFOS, PFHxS and PFOA from a fictitious, but representative source
- area under a range of climatic conditions and soil types. PFAS mass flux over time was evaluated
- and compared over a range of common management technologies, including full and partial soil
- stabilisation, clay capping as well as a "do nothing" approach, relying on PFAS concentrations to
- dilute and disperse over time. The relative efficacy of these technologies in reducing PFAS flux
- 128 over the foreseeable future was assessed across the wide range of Australian climates and soil types,
- 129 covering effected site conditions from the tropical north to the relatively dry, Mediterranean climate130 in the south.

#### 131 **1.2** Objectives

- 132 The aim was to assess and compare flux of PFOS, PFHxS and PFOA draining from a 1.5 m soil
- profile under a range of climatic conditions and soil types, and assuming a range of sorptioncoefficients for each of these compounds.
- 135 The purpose was to better understand the PFAS mass flux from a simulated source zone over time,
- across a range of plausible environmental conditions and to assess the relative importance of
- 137 various environmental features to the leaching behaviour of PFAS.
- 138 A secondary objective was to simulate the effects of implementing various source zone remediation
- techniques on the flux, forward predicted over 30 years, to assess the relative efficacy of these
- 140 technologies in reducing PFAS flux over the foreseeable future.

#### 141 2 Materials and Method

- 142 2.1 Study site and numerical model simulator
- 143 This study comprised a theoretical leaching study, based on modelling of unsaturated flow using an
- updated version of LEACHP, the pesticide version of LEACHM (Leaching Estimation and
   Chemistry Model) (Hutson, 2003) unsaturated zone numerical model.
- 146 LEACHM is a one-dimensional, finite difference model to simulate water flow and solute transport
- in the vadose zone. The model allows for use of actual meteorological records. Solutes are
- transported by advection, dispersion and diffusion and the model considers solute retardation
- 149 through sorption onto soil particles and organic matter.
- 150 The code has been benchmarked against similar industry accepted models, such as HYDRUS and
- 151 GLEAMS (Nolan et al., 2005, Dann et al., 2006, Sarmah et al., 2006, Ogorzalek et al., 2008), and
- 152 has been widely used to simulate fate and transport of contaminants in the unsaturated zone (Webb
- 153 et al., 2008, Asada et al., 2013, Nasri et al., 2015), including PFAS (Wallis et al., 2022).
- 154 The model was designed to simulate land management at a plausible source zone, such as a fire
- training ground at a defence site, in which PFAS as an aqueous foam was deposited as a weekly
- event over approximately two decades (1981 to 1999), followed by a period of dormancy (while site
- assessment may have been conducted but no actual remediation performed, 2000 to 2019). Finally,
- the project seeks to assess the effects of various treatment technologies over a forward predicting 30
- 159 years period from 2021 to 2050. The model phases are summarised in Table 1.

#### 160 Table 1: Modelled timescale and land management

| Dates     | Model<br>duration (yrs) | Model Phase  |
|-----------|-------------------------|--|
| 1979-1980 | 2                       | Spin-up period to obtain moisture equilibrium within soil profile. No PFAS application and/or management scenario.   |
| 1981-1999 | 19                      | Wet application of PFAS (3M lightwater at a 3% formulation) to the soil surface, once per week, to simulate either a fire training exercise or a testing of pumps. |
| 2000-2020 | 21                      | Cessation of PFAS application; site dormant –Represents a period of investigation and remediation planning.  |
| 2021-2050 | 30                      | Remediation applied and maintained for 30 years. Repeat of weather record from 1989-2018 for this prediction period.   |

#### 161 2.2 Model setup

162 A summary of model inputs is provided in Table 2 with further discussion on the selection of the

- 163 key variables being tested (climate, soil types, and sorption coefficients) following.
- 164 Table 2: Adopted model constants and input selections

| Input parameter Units Modelled va               |                  |            | Source/Assumptions   |
|---|------------------|------------|--|
| Model setup                                     |                  |            |  |
| Profile depth                                   | mm               | 1,500      | Plausible soil profile depth to which remediation may be expected to be applied.   |
| Vertical discretisation                         | mm               | 20         | Modelled assumption  |
| Water flow model                                | -                | -          | Richards (1931) equation   |
| Lower boundary                                  | -                | -          | Unit gradient drainage   |
| Vegetation                                      | -                | -          | Absent   |
| Weather   | -                | Variable   | Section 2.3  |
| Soil Features                                   |                  |            |  |
| Humus, organic carbon                           | %                | 0.5        | Realistic model assumption, representative for the Australian context.   |
| Particle size distribution                      | %                | Variable   | Section 2.3.2  |
| Bulk Density                                    | t/m <sup>3</sup> | 1.4 to 1.8 | Linear increase with depth from surface to 1.5 m depth, reflecting compaction with depth.                                      |
| Hydraulic properties and water retention curves | -                | Variable   | Model based on pedotransfer functions in<br>Minasny and McBratney (2000)   |
| Initial matric potential                        | kPa              | Variable   | Model run for two years (1989-80) prior to<br>PFAS irrigation commencing to allow for<br>equilibration with natural conditions |

| Input parameter                            | Units             | Modelled value           | Source/Assumptions   |
|--|-------------------|--------------------------|--|
| Slope                                      | %                 | 0 to 1                   | <ul><li>0% assumed in baseline models and<br/>uncapped remediation model scenarios.</li><li>1% applied to capping remediation scenarios<br/>to simulate an engineered cap.</li></ul> |
| Chemical features                          |                   |                          |  |
| Molecular weight:<br>PFOS<br>PFHxS<br>PFOA | AMU               | 500.1<br>400.1<br>414.1  | As reported in Table 4-1 of Interstate<br>Technology and Regulatory Council (ITRC)<br>(2020).  |
| Solubility:<br>PFOS<br>PFHxS<br>PFOA       | mg/L              | 570<br>1,400<br>3,400    | As reported in Du et al. (2014)  |
| Sorption (Koc):<br>PFOS<br>PFHxS<br>PFOA   | L/kg              | Variable                 | Section 2.3.3  |
| PFAS application                           |                   |                          |  |
| Duration                                   | L/m <sup>2</sup>  | Weekly from<br>1981-1999 | Section 2.3.4  |
| Application rate                           | L/m <sup>2</sup>  | 6                        | Section 2.3.4  |
| Concentrations:<br>PFOS<br>PFHxS<br>PFOA   | mg/L              | 300<br>30<br>3           | Based on data in Backe et al. (2013). See<br>Section 2.3.4.  |
| Mass per week:<br>PFOS<br>PFHxS<br>PFOA    | mg/m <sup>2</sup> | 1,800<br>180<br>18       | Concentration × Rate   |

#### 165 2.3 Model variants

A total of 144 base model variants for the full model duration (1979 to 2050) were run, covering all potential combinations of four different soil types, four different climate records and low, medium to high sorption potentials, for three PFAS compounds.

- 169 Subsequently, for each of these 144 models four soil remediation treatments were modelled (as
- discussed in Section 2.4 for the period 2020 to 2050). Thus, including the untreated control plus
  four treatments, a total 720 model variants were run.
- 172 2.3.1 Model variant: Climatic conditions
- 173 A primary driver for PFAS vertical migration is likely to be the climate as this dictates the amount
- of drainage as well as evapotranspiration acting upon the soil profile throughout the year. Four

- weather records were selected which cover a wide range of Australian climates; from the wet
- tropics in the north to low rainfall climates in the South:
- Darwin NT representing the wet tropics, with a very high annual rainfall (average of 1,790 mm) and very strong summer dominant rainfall distribution;
- Williamtown NSW a temperate coastal climate, with high annual rainfall (average of 1,100 mm) typically falling in a uniform distribution across the year;
- Gatton QLD with a summer dominant, moderate rainfall (average of 710 mm); and
- Edinburgh SA representing a climate with winter dominant, relatively low rainfall (420 mm).
- 183 Climate files were prepared using daily weather observations provided on the Queensland
- 184 Government's SILO database of Australian climate data for the above weather stations (Queensland
- 185 Government, 2022). In each case the model utilised daily records for rainfall (mm),
- temperature (°C) and potential evapotranspiration (mm).
- 187 For the forward simulation between 2021 and 2050, the weather datasets from 1989 to 2018 were
- repeated. This timeframe was selected so that weather datasets for leap years would align.





dominated rainfall areas (Edinburgh). The annual average rainfall for the selected four weather stations are also shown
 (based on data for the modelled period 1979 to 2020).

#### 193 2.3.2 Model variant: Soil types

194 It was surmised that soil type was likely to have a significant influence on PFAS migration in the

unsaturated zone, primarily through physically controlling the movement of water. Therefore, four

- soil types were compared, covering a range of soil permeabilities. The model was based on a 1.5 m
- deep soil profile. The four soil textures modelled had varying clay and silt (fines) contents ranging
- 198 from 75% fines (60% clay and 15% silt) to 5% fines (2.5% clay and 2.5% silt). The selected
- 199 textures are presented on a texture triangle in Figure 2, and referred to in this paper as Clay, Clay

Loam, Loamy Sand, and Sand. In each case the organic carbon content was kept constant at 0.5%, which, although low, was considered representative in the Australian context, particularly averaged

across a 1.5 m model depth profile. It is recognised that the humus in soil is rarely a constant and

- typically decreases with depth. However, as part of this study was to compare the effects of various
- Kd values, and the model calculated Kd as the product of the Koc and fraction of organic carbon, it
- 205 was necessary to apply a constant organic carbon parameter.



- Figure 2: Summary of modelled soil textures (National Committee on Soil and Terrain, 2009).
- 207 2.3.3 Model variant: Applied sorption coefficients (Kd)

The soil-water partitioning coefficient (Kd) in groundwater modelling is frequently used to account for retardation of a solutes transport with respect to water. Retardation occurs primarily through the action of sorption and desorption from the solid phase. However, the mechanisms controlling PFAS

- sorption are complex (e.g. Higgins and Luthy, 2006, Li et al., 2018, Brusseau, 2018).
- 212 Factors affecting PFAS sorption have been shown to include PFAS compound functional groups
- and carbon chain lengths (Prevedouros et al., 2006, Milinovic et al., 2015), electrostatic interaction
- 214 (Higgins and Luthy, 2006, Du et al., 2014), hydrophobic interactions with the fluoridated
- hydrocarbon chain (Deng et al., 2012, Du et al., 2014), and organic carbon content of the soil
- (Milinovic et al., 2015, Gallen et al., 2017). In addition, pH effects (Benskin et al., 2012, Wang and
- 217 Shih, 2011, Gallen et al., 2017) and salinity (Xiao et al., 2011, Kim et al., 2015) have also been
- shown to be significant factors affecting sorption and desorption. In addition, sorption to the air-
- 219 water interface has also been shown to be an important factor for organics in the unsaturated zone
- (Kim et al., 1998), and specifically for PFAS, given the unique surface properties of these
- compounds (Brusseau, 2018, Brusseau et al., 2019).
- 222 This study sought to model likely behaviours of PFAS under a range of climatic and soil conditions,
- rather than to simulate PFAS migration at a specific site. Therefore, rather than seeking to account
- for individual factors affecting sorption as outlined above, though simplistic, application of linear
- 225 equilibrium sorption coefficients (Kd) was considered an appropriately representative
- approximation, accounting for these potentially interacting mechanisms that could control the
- 227 retardation of PFAS in the unsaturated zone. As the objective was to compare plausible behaviour

- under a range of typical soil types and climates assumed Kds within a range of measured Kds for
- 229 PFOS, PFHxS and PFOA was considered a satisfactory approximation of sorption capacities.
- 230 LEACHM calculates the Kd based on the product of the organic carbon-water partitioning
- coefficient ( $K_{oc}$ ) and the fraction of organic carbon ( $f_{oc}$ ). Therefore a range of  $K_{oc}$  values were
- selected from within the wide range of published values presented by the Interstate Technology and
- Regulatory Council (ITRC) (2020). Rather than modelling the extremes of the published range,
- three K<sub>oc</sub> values were selected, that for this study have been designated "Low", "Medium" and
- 235 "High" from within the published range as discussed in the supplementary material (Appendix B.1).
- A summary of the modelled Kd values (derived from the product of Koc and foc) is presented in
- 237 Table 3.
- Table 3: Summary of modelled Log Kd values. Values are based on published data from the Interstate Technology and
   Regulatory Council (ITRC) (2020).

| Analyte | Low   | Medium | High |
|---------|-------|--------|------|
| PFOS    | 0.16  | 0.69   | 1.21 |
| PFHxS   | 0.04  | 0.57   | 1.11 |
| PFOA    | -0.12 | 0.40   | 0.91 |

#### 240 2.3.4 Simulated PFAS application

241 PFAS was simulated to be applied to the soil surface as part of fire training or equipment testing 242 procedures, and based on representative historic application examples at airports or Australian defence sites. It was assumed that the AFFF product that was applied was similar to 3M lightwater, 243 applied in a 3% formation. The modelled concentration of PFOS, PFHxS and PFOA in the irrigated 244 foam was thereby based on Backe et al. (2013). The reported average concentration of these 245 246 compounds in 3M AFFF from 1989 to 2001 was rounded, and then the values multiplied by 0.03 to simulate 3M lightwater at a 3% formulation. These resulting application concentrations are 247 presented in Table 2. 248

The modelled frequency of application was once per week for the period 1981 to 2000, to simulate either a fire training exercise or a testing of pumps. The assumption of volume applied during these weekly events was 6 mm (6  $L/m^2$ ). See supplementary material, Appendix B.2, for the calculations underlying these assumptions.

- 253 2.4 Modelling PFAS source zone treatments
- 254 Remediation of PFAS source sites in Australia typically involves a combination of technologies.
- 255 Generally, due to cost implications, thermal desorption is reserved for the most heavily impacted
- soil, while soil with low to moderate contamination levels is typically managed onsite through
- either stabilisation, encapsulation or capping, or a combination of stabilisation and capping.
- In order to evaluate the efficacy of various onsite management options, the following modelvariants were run:
- a) Do nothing approach (control). This scenario simulates PFAS flux over time when relying only
   on natural dispersion and dilution to reduce concentrations below source zones

- b) Partial soil profile stabilisation (upper 1m): simulated through an increase of the Kd within the
  source zone area by 2 orders of magnitude. This was achieved by increasing the organic carbon
  by 2 orders of magnitude (to 5%) for the portion of the profile that was stabilised.
- c) Full profile stabilisation (whole 1.5m): as scenario b), however, Kd is increased by 2 orders of
   magnitude over the entire simulated soil profile;
- d) Clay capping of soil profile: addition of a 20 cm heavy clay cap under a 1% soil slope;
- e) Clay capping and partial profile stabilisation: combination of scenarios (b) and (d).
- 269 These scenarios are graphically depicted in Figure 3.



Model outputs: Annual PFAS mass drained over time, and total mass drained vs control

270 Figure 3: Modelled treatment scenarios

#### **3** Results and discussion

#### 272 **3.1** Water balance and transport

The study is focused on the transport of PFAS compounds; however, this transport is only through migration as a dissolved phase in percolating water. Therefore, this section is provided as a summary of the water distribution under the various soil and climate scenarios.

Figure 4 presents the average of various water balance elements (rainfall, runoff, infiltration, evaporation, and deep drainage) for the Clay Loam soil type under each modelled climate. Graphs for other soil types are available in the supplementary material (Appendix C), and show relatively similar features, although with increasing infiltration and drainage and decreasing runoff, as clay content decreases.

The very high and high rainfall climates are more prone to experiencing runoff, but in general the bulk of rainfall was seen to infiltrate, and then depending on the climate, is either evaporated from the profile or drains through the profile. The tropical climate shows significant water movement through the profile during the wet season but minimal water fluxes during the dry season. In contrast the uniform rainfall climate shows consistent monthly fluxes of water throughout the year, with a slightly higher evaporation occurring in the summer months.



■ Rainfall ■ Runoff ■ Infiltration ■ Evaporation ■ Drainage

Figure 4: Average of water balance elements by month for Clay Loam soil type, under the range of modelled climates. The tropical climate shows significant water movement during the wet season, including runoff and drainage but minimal water fluxes during the dry season. In contrast the uniform rainfall climate shows consistent monthly fluxes of water throughout the year. The lower rainfall climates, both summer and winter dominant patterns, show low levels of deep drainage compared with the higher rainfall models, with the majority of drainage corresponding to, but lagging slightly behind, the season when highest rainfall occurs.

290 The lower rainfall climates, including both summer and winter dominant, show significantly lower

levels of drainage compared with the higher rainfall models. The majority of drainage corresponds

- to, but lags slightly behind, the season when highest rainfall occurs. The evaporation presented is
- actual evaporation from the soil (not pan evaporation), and as such, for the winter dominant rainfall
- the highest rates of modelled evaporation actually occur during the winter months as that in when the surface and upper portion of the profile is wet.

#### 296 3.2 PFAS transport in the unsaturated zone

297 The following sections discuss effects that each modelled variables of sorption capacity (Kd),

climate and soil type have on the rate of leaching of PFOS, PFHxS and PFOA. These effects are

discussed in terms of the untreated (control) models.

#### 300 3.2.1 Effect of sediment sorption properties (Kd)

301 For the purposes of demonstrating the effect of the Kd on PFAS mass flux Figure 5 has been

302 included and presents the annual PFOS mass flux of a sandy clay loam under high rainfall

303 conditions. As can be seen sorption capacity (Kd) is a sensitive parameter in the model. In all

models, for each of the PFAS compounds, the lower Kd values resulted in more rapid expression of

305 PFAS at the base of the modelled 1.5 m soil column, with a more rapid rise in mass flux until PFAS

application ceased and then a relatively rapid decline in the flux as the PFAS is leached from the system.



308 Figure 5: Comparing PFOS leaching over time, for a sandy clay loam, under High rainfall conditions

309 Where the sorption capacity of the soil is high (high Kd models) the rate of contaminant migration

is significantly retarded resulting in delayed expression of the PFAS at the base of the soil column.

In the case of the example provided in Figure 5, by several decades. The flux then rises slowly but

- in the majority of modelled scenarios had not reached a maximum by the models end in 2050, and
- for some soil types (particularly clays) had not reached the base of the profile in this timescale.
- In practical terms this finding shows the importance of understanding the sorption capacity of soils

at a site-specific level. The Kd will be a site-specific feature, likely resulting from a range of soil

- properties; such as organic carbon, pH, salinity and minerology (Higgins and Luthy, 2006, Hamid et al., 2018). If the Kd on a site is inherently high then it could be expected that the rate of PFAS
- al., 2018). If the Kd on a site is inherently high then it could be expected that the rate of PFAS
  migration to groundwater will be slowed, leading to plumes with relatively low maximums but of
- extended duration. However, if the Kd is low, such as in a soil depleted of organic carbon, then it
- could be expected that the PFAS will migrate with little retardation resulting in a high concentration
- 321 load at the peak but a relatively short plume duration as the PFAS is more rapidly flushed from the 322 system.
- For the purposes of demonstrating the effects of rainfall and soil type on the flux relationships the models based on moderate Kd are presented. Models based on the high Kd and low Kd follow similar curves but are either more compressed, with higher peaks, or are more extended with flatter curves respectively.

#### 327 3.2.2 Effect of climatic conditions

328 The impact of climatic conditions on PFOS migration through the unsaturated zone is shown in

Figure 6 and illustrates the profound impact of rainfall on rate of migration across all soil types.

330 Leaching behaviour for PFHxS and PFOA follows similar patterns, and have been provided along

331 with the corresponding graphs for low and high Kd, in the supplementary material (Appendix D).



Figure 6: PFOS mass flux [mg/m<sup>2</sup>] at the base of the soil profile (1.5m) over time is shown for four different Australian climates and soil types. A median distribution coefficient for PFOS was assumed for all model scenarios (see Table 3).

- 334 For this example, under "very high" rainfall conditions across most soil types assessed, the PFAS
- compounds were observed at the base of the unsaturated zone (1.5. metres depth) within five years
- of AFFF application commencing. Once PFAS begins to express at the base of the unsaturated zone
- the annual mass flux rapidly rises, peaking at the time that the PFAS use ceases before a relatively
- rapid tailing off of annual fluxes as PFAS is leached beyond the modelled 1.5 m soil profile. The
- exception to this are heavy clay soils which display a much slower expression of the PFAS at the
- base of the soil profile, delayed by approximately 10 years compared to the sandier soils and not
- peaking until 30 years post PFAS applications ceasing.
- 342 Under the "high" rainfall (Williamtown, NSW) and "moderate" rainfall (Gatton, Qld) respectively,
- the models showed an elongating and flattening of the annual mass flux curves. In the case of the
- high rainfall, although the PFAS was appearing at the base of the profile within a decade of PFAS
- being applied, the mass flux peak appeared to take several decades to pass.
- 346 Under low rainfall conditions (Edinburgh, SA) the migration of PFAS is minimal and the simulated 347 mass flux at the base of the soil profile is still rising by the end of the model timeframe.
- These findings may have a bearing on regulator and site owner's remediation planning. In high rainfall environments the planning timeframe should be shorter than in environments where rainfall
- is lower so that any remedial action can take place before the worst of the flux has occurred. Thus,
- in high rainfall areas, management responses may need to be implemented more rapidly and involve
- a more robust approach compared to the response required in lower rainfall environments.
- 353 3.2.3 Effects of soil type
- Figure 6 is also helpful for understanding the response by soil type. Generally, particle size distributions have an effect on the hydraulic conductivity of a soil and therefore, unsurprisingly, increased clay content resulted in models with increasing delay in the expression of PFOS at the base of the profile, and an extending of the timeframe for the leaching process to occur.
- Soils with clay contents up to 35% (sand, sandy loam and loamy clay), however, display a
  somewhat similar response, while the result for the clay soil (with 60% clay) was markedly delayed.
  Overall, model results demonstrate that the effect of soil type on PFAS migration and retention is
  much less pronounced than the impact of climatic conditions, except for heavily clay-dominated
- 361 much less pron
  - Again, these findings are of relevance to regulators and owners of sites with PFAS source zones. Lighter textured (sandier) soils will more rapidly allow migration of PFAS to depth than source sites that are on clay soils. Without intervention, plumes arising from sites with sandy sands can be
  - expected to have relatively high mass flux peaks compared with sites on more clayey soils, but the
     plumes will be of shorter duration.
  - 368 3.3 Treatment efficacy
  - 369 3.3.1 PFAS flux responses to treatment
  - 370 The efficacy of various treatments under the range of modelled climates is graphically displayed for
- 371 PFOS (assuming a moderate Kd and sandy clay loam soil type) in Figure 7 for illustrative purposes.
- 372 Similar graphs for all other iterations of Kd and soil type are presented in the supplementary
- 373 material (Appendix E).

- All treatment options were effective at reducing PFAS leaching, as compared with the untreated
- 375 "control" models. The response to treatment was an immediate reduction in PFAS mass flux at the
- base of the modelled soil profile. The stabilisation of 1 m of soil (grey line) was generally the least
- effective treatment, however stabilisation of the full 1.5 m profile (blue line) was extremely
- effective resulting in a mass flux at least 1 to 2 orders of magnitude lower compared to other
   treatment scenarios.

380 This indicates that a stabilisation technology that is capable of increasing the Kd by 2 orders of 381 magnitude is extremely effective at reducing flux from the material that is stabilised. However, as demonstrated in the partial profile stabilisation scenarios (the 1 m of stabilised soil), any PFAS 382 remaining beneath the treated zone will continue to sustain ongoing leaching to groundwater. This 383 observation is similar to the findings of Mahinroosta and Senevirathna (2021), who also modelled a 384 stabilisation strategy and found that although the PFAS concentrations in groundwater reduced from 385 a "do nothing" approach they remained significant. Mahinroosta and Senevirathna (2021) surmised 386 this was due to residues in the ground beyond the remediation area. Whilst lateral residues beyond 387 the treated area would be expected to result in further leaching to groundwater, this current study 388 389 shows that residues beneath the treated zone are also a source of ongoing flux.

The two scenarios where a cap was applied (the orange and yellow lines) essentially overlap one 390 391 another in most of the modelled scenarios, showing that a cap is generally more effective than a partial profile stabilisation, and that the cap is ultimately controlling the leaching even when in 392 combination with stabilisation of part of the profile (at least in this model scenario). Of practical 393 note, where a cap is installed to provide vertical hydraulic control it will be effective from the 394 395 surface to the depth of the groundwater. Although in this study there was little benefit in stabilising and capping together, some sites where subsurface lateral flows are a feature (e.g. sites with 396 seasonally perched water) may benefit from that zone being stabilised to prevent shallow lateral 397 migration of PFAS with perched water. 398

In the case of the very high rainfall and high rainfall scenarios presented in Figure 7, the rate of annual PFAS mass flux was on the decline prior to the treatment being implemented, indicating that the majority of the mass had already leached beyond the modelled 1.5 m soil column. This was also frequently observed in the low Kd models (see Appendix E.1), particularly in lighter textured soils where in some cases the PFAS of interest had fully leached from the profile prior to the treatment phase commencing (from 2021 to 2050).



Figure 7: PFOS leaching from Sandy Clay Loam, under various treatment regimes, each graph showing efficacy under a different rainfall regime. Treatments generally showed initial improvements compared to the control (No treatment), with full profile stabilisation (Stabilise 1.5m) being most effective. Partial profile stabilisation (Stabilise 1 m) was less effective than capping, and a combination of cap and stabilisation showed little added benefit compared to cap alone.

- 408 Where the leaching front had passed and the annual mass was in decline at the time of treatment,
- 409 the untreated control showed an ongoing decline in the mass flux over time, in some cases resulting
- 410 in lower annual fluxes by the model end than if remediation had occurred (e.g. in the case of very
- 411 high rainfall scenario in Figure 7). This illustrates that where non-destructive remediation
- technologies are applied (capping or stabilisation), and there is no biodegradation occurring (as was
- modelled) then in the long term the treatment will essentially delay the leaching of the PFAS from
- the environment. Thus, treatment will result in plumes remaining in the environment for a longer
- 415 period of time than if a "do nothing" approach was adopted, albeit with relatively lower maximum
- 416 fluxes compared with pre-treatment levels.
- 417 3.3.2 Observations from pictorial representations of PFAS mass flux
- The effects of the modelled treatments are also shown pictorially in Figure 8. These diagrams show
- 419 relative water content and PFOS concentration in the soil profile (vertical axis) over the modelled
- period. The diagram presents these attributes for PFOS in an environment with moderate sorption
   potential (Kd) applied to a sand soil profile under low rainfall conditions. Diagrams for other PFAS
- 422 chemicals, for each modelled Kd, soil type and rainfall regime are available in the supplementary
- 423 material (Appendix F).
- Figure 8 shows that without treatment the PFOS applied at the surface will migrate vertically over
- time to the base of the profile (as discussed above). The diagrams show visually the effects of the various treatments applied in the year 2021.
- 427 The "Stabilise (1 m)" graphic shows PFOS in the treated upper metre becoming significantly
- retarded resulting in a shadow of relatively low PFOS concentration beneath the stabilised zone.
- However, PFOS at depths greater than 1 m continues to leach through the base of the modelled soilprofile as has been discussed earlier.
- The clay capping scenarios are shown with the additional profile thickness introduced in 2021. The
- 432 clay cap retains a higher moisture content than the sand profile and so can be seen as a darker blue.
- The cap results in lower moisture content in the sand beneath the cap (lighter blue compared with
- the control), resulting in a slowing of the downward PFOS migration. The "cap only" scenario,
  however, shows a degree of back diffusion or wicking of PFAS into the cap over time, so that
- 436 within a few years a low presence of PFAS can be observed throughout the entire cap (light pink).
- 437 Wicking would likely be a slow process driven by matrix potential gradients in the climate driven
- 438 wetting and drying cycles of the cap. A similar observation of PFAS wicking was made by Thai et
- 439 al. (2022) in their experiments on surface water fluxes from concrete. They surmised that sustained
- 440 PFAS in runoff over repeated simulations may be explained by vertical wicking through the
- 441 concrete over time.
- In contrast, in the modelled example of a cap overlying stabilised material, very little back diffusion
- 443 or wicking was observed. Pictorially (in Figure 8) it is indiscernible, but in reality, for this example,
- 444 wicking in the cap overlying stabilised material was detectable but at a maximum of 2.5 % of the
- 445 concentration in the unstabilised scenario. This is likely due to the high Kd of the PFAS in the
- stabilised material significantly retarding the migration of PFAS into the matrix of the overlying
- 447 clay.



Cap only



#### Stabilise (1m)



#### Cap and Stabilise



Figure 8: Pictorial demonstration of the modelled leaching process and the effects of various treatments. These graphics show PFOS concentration in a sand profile under the low

rainfall scenario. The "No treatment" model shows PFOS slowly leaching through the profile. The "Stabilise (1m)" graphic shows PFOS in the treated upper metre becoming entrapped resulting in a shadow of relatively low PFOS beneath the stabilised zone. However, PFOS below 1m continues to leach through the base of the soil profile. The clay capping scenarios

451 are shown with the additional profile thickness introduced in 2020. The cap results in lower moisture content in the sand beneath the cap, slowing the downward PFOS migration rate.

452 Of note, the "cap only" scenario shows a degree of back diffusion or wicking of PFAS into the cap, so that within a few years a low presence of PFAS can be observed at the surface of

453 the cap. In contrast where a cap is placed over stabilised material no back diffusion is observed.

- This observation has implications for PFAS source zone management. If upward diffusion or
- 455 wicking of PFAS occurs, it may contribute to ongoing surface water flux from source sites. Whilst
- 456 quantification of the vertical wicking through the cap, and subsequent runoff concentration
- 457 increases is beyond the scope of this study, remediators should be mindful that this could occur, and
- 458 may need to consider mitigation. This may comprise a capillary break layer engineered into the cap,
- a guard layer of activated carbon, or stabilisation of the upper layer of the PFAS impacted zone
- 460 prior to placement of the cap.

#### 461 **3.3.3** Treatment efficacy as a percent of untreated flux

The total PFOS, PFHxS and PFOA leached over the 30 year modelled treatment period, for each treatment scenario, was also compared as a percent of the totals leached from the untreated control models. This allowed assessment of the relative effectiveness of the treatment options over the modelled period as summarised in Figure 9.

- 466 The matrix of box plots show that stabilisation of the full profile (the blue bars) is extremely
- effective, generally resulting in the total PFAS leached being less than 10% of the untreated control
  over a 30 year period. The models of complete profile stabilisation, across all rainfall regimes, were
  more effective than any other treatment. However, in practical terms it is rarely feasible to treat all
  PFAS in a source zone. It is likely some residue will remain at depth.
- When comparing all other treatment scenarios, the models for the clay soil profiles showed the
- 472 greatest similarity in the percent leached compared to the control as shown by generally similar
- 473 position and size of boxplots between treatments. Results show that for the 1 m partial profile
- stabilisation (grey columns), sandy soils generally performed worse than the cap alone (orange),
  and this difference was more pronounced where higher Kds were applied (i.e. soils with a higher
- sorption capacity). The only exception to this was a marginally better response by stabilisation of
- 477 clayey soils than capping of those soils. As the clay content of the soil decreases, the capped
- 478 scenarios become increasingly effective and the partial stabilisation approach increasingly less
- 479 effective.
- 480 The combination of cap and stabilisation (yellow columns) was generally very similar to the cap
- alone and usually not markedly better. It was only under the low Kd models that there was any
- 482 notable improvement in the combination compared to the cap alone, and that only under the higher
- 483 leaching environments (high to very high rainfall).



Stabilise 1.5 m Stabilise 1 m Cap Cap + Stabilise 1 m

Figure 9: Summary of PFAS leaching as a percent of untreated control, by soil type, under various management strategies. The bars in each case account for the spread of results across the range of modelled rainfall regimes. Full profile stabilisation was most effective, but may not be practical. Partial stabilisation (stabilise 1m) was generally less effective than capping or capping with stabilisation. This difference became more apparent in models of sandier soils and those with higher Kds.

#### 487 4 Conclusions and implications for PFAS source zone management

- This study reinforces that soil type, rainfall and sorption capacity are all important factors in the transport of PFAS compounds within the unsaturated zone.
- 490 The sorption capacity (Kd) of the unsaturated zone significantly influences PFAS migration and
- 491 leaching. Soils with high sorption capacity showed relatively slow migration, leading to delayed
- 492 PFAS drainage to groundwater, but extended plume duration. Conversely, PFAS migration through
- 493 soils with low sorption capacity was relatively rapid, resulting in high peak PFAS loads but of
- 494 relatively short duration. The Kd is a site-specific feature influenced by various soil properties.
- 495 Therefore, understanding the sorption capacity of soils at a site-specific level is crucial.
- Rainfall significantly impacts PFAS migration. Under high rainfall, with the exception of heavy
   clay soils, PFAS compounds reached the base of the unsaturated zone within five years, with a rapid
- flux increase and decline. However, heavy clay soils show slower PFAS expression at the base of
- the modelled 1.5 m soil column, delayed by about 10 years. Under more moderate rainfall regimes,
- 500 curves of annual PFAS mass flux are more elongated and flattened, while low rainfall conditions
- show minimal PFAS migration. These findings are vital for remediation planning, indicating the
- 502 need for more rapid and robust responses in high rainfall areas.
- 503 Model results demonstrate that the effect of soil type on PFAS migration and retention, though 504 important, were less pronounced than the impact of climatic conditions, except for heavily clay-505 dominated soils.
- 506 In terms of various treatment options based on capping and stabilisation technologies:
- All treatment options assessed were effective at reducing the rate of vertical PFAS mass flux.
- Contaminant stabilisation is very effective in reducing mass flux for the material that is treated.
   However, residual PFAS below the treated material will continue to sustain ongoing vertical
   fluxes unless hydraulic controls are also implemented.
- The relative effectiveness of partial profile stabilisation, compared to capping, appears to
   decrease with increasing sand content, and this is exacerbated in scenarios with higher sorption
   capacity compared to those with lower sorption capacity.
- In contrast clay capping, controlling infiltration is effective from the surface down and in lighter textured soils is likely to be more effective at controlling drainage from the profile than partial profile stabilisation alone. Generally, a combination of partial profile stabilisation and clay cap was of little additional benefit, with respect to PFAS drainage, compared to providing the cap alone.
- A potential concern for simple capping solutions, however, is that if high levels of PFAS are in contact with the underside of the cap the compounds may wick into the cap eventually being observed to be present throughout the cap. This does not appear to occur in scenarios where
   stabilisation and capping are combined.
- 523 This study provides valuable insights into the retention and migration of PFAS in the unsaturated 524 zone beneath AFFF source sites, under different remedial treatment approaches. The findings may 525 reduce uncertainties around the effectiveness of various in-situ soil treatment options and may 526 support remedial project managers and practitioners evaluating site management approaches.

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#### Acknowledgements 528

529 We thank Flinders University for their support of this research and particularly feedback from the staff in the College of Science and Engineering. 530

#### Formatting of funding sources 531

532 This research did not receive any specific grant from funding agencies in the public, commercial, or

533 not-for-profit sectors.

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Climate averages - Gatton QLD (summer dominant, moderate rainfall).





#### Appendix B Calculations of Selected Model Parameters

#### B.1 Selection of Koc Values

The intention was not to model the extremes of published Koc values. Published ranges were taken from Interstate Technology and Regulatory Council (ITRC) (2020) referenced sources. Within those ranges any perceived outliers were removed and then the upper and lower extents were trimmed by adding approximately 0.3 to the lower Log Koc and subtracting approximately 0.3 from the upper extent of selected published Log Koc values.

The medium value for modelling was taken as the mid point between the upper and lower selected Koc values as shown in the following table. The modelled Kd was then the Koc  $\times$  the fraction of organic carbon (0.005):

| Parameter                                | PFOS |        |      | PFHxS |        |      | PFOA  |        |      |
|--|------|--------|------|-------|--------|------|-------|--------|------|
|  | Low  | Medium | High | Low   | Medium | High | Low   | Medium | High |
| Log K <sub>OC</sub> (Raw)                | 2.17 | -      | 3.80 | 2.05  | -      | 3.70 | 1.89  | -      | 3.50 |
| Log K <sub>OC</sub> (Selected)           | 2.46 | 2.99   | 3.51 | 2.34  | 2.88   | 3.41 | 2.18  | 2.70   | 3.21 |
| Log Kd (Modelled)<br>(Koc × foc of 0.005 | 0.16 | 0.69   | 1.21 | 0.04  | 0.57   | 1.11 | -0.12 | 0.40   | 0.91 |

Table B.1: Calculation of sorption coefficients (Kd) used in the models.

#### B.2 Calculation of irrigation rate

Background Assumptions:

- Fire fighting equipment tested every week and after any service (Colville and McCarron, 2003)
  - Therefore, assume a weekly irrigation cycle
  - Assuming three trucks per fire station, 2 hoses or a cannon per truck
- Assume it takes 1 to 5 minutes per nozzle to test
  - Therefore 2 minutes  $\times 6 = 12$  minutes of spraying per week.

#### Calculation:

- Regulated requirement for capacity of fire fighting equipment in Australia depends on the size of the airfield (Air Services Australia, 2023a).
  - A Category 8 airfield needs to be able to discharge 7,200 L/min.
  - Assuming the total of all vehicles pump that amount and it takes 2 minutes per piece of equipment to test.
  - $\circ$  Equates to a total of 14,200 L discharge per week = round to 15 KL
  - Assuming the test area is 50 m by 50 m = 250 m<sup>2</sup> = 6 L per m<sup>2</sup> = 6 mm irrigation per week.

# Appendix C Water Balance Data and Graphs

#### C.1 Clay Models

| Figure C.1   | Water Balance Elements – Clay, Tropical, Very High Rainfall            | C-2 |
|--------------|--|-----|
| Figure C.1   | Water Balance Elements – Clay, Uniform, High Rainfall                  | C-2 |
| Figure C.1   | Water Balance Elements - Clay, Moderate, Summer Dominant Rainfall      | C-3 |
| Figure C.1   | Water Balance Elements - Clay, Low, Winter Dominant Rainfall           | C-3 |
| C.1 Clay Loa | m Models   |     |
| Figure C.1   | Water Balance Elements – Clay Loam, Tropical, Very High Rainfall       | C-4 |
| Figure C.1   | Water Balance Elements – Clay Loam, Uniform, High Rainfall             | C-4 |
| Figure C.1   | Water Balance Elements - Clay Loam, Moderate, Summer Dominant Rainfall | C-5 |
| Figure C.1   | Water Balance Elements - Clay Loam, Low, Winter Dominant Rainfall      | C-5 |
| C.1 Loamy Sa | and Models   |     |
| Figure C.3   | Water Balance Elements – Loamy Sand, Tropical, Very High Rainfall      | C-6 |
| Figure C.3   | Water Balance Elements – Loamy Sand, Uniform, High Rainfall            | C-6 |
| Figure C.3   | Water Balance Elements – Loamy Sand, Moderate Summer Dominant Rainfall | C-7 |
| Figure C.3   | Water Balance Elements - Loamy Sand, Low, Winter Dominant Rainfall     | C-7 |
| C.1 Sand Mo  | dels   |     |
| Figure C.3   | Water Balance Elements – Sand, Tropical, Very High Rainfall            | C-8 |
| Figure C.3   | Water Balance Elements – Sand, Uniform, High Rainfall                  | C-8 |
| Figure C.3   | Water Balance Elements – Sand, Moderate, Summer Dominant Rainfall      | C-9 |
| Figure C.3   | Water Balance Elements - Sand, Low, Winter Dominant Rainfall           | C-9 |
### C.1 Clay Models



| Month | Rainfall | Runoff | Infiltration | Evaporation | Drainage |
|-------|----------|--------|--------------|-------------|----------|
| Units | mm       | mm     | mm           | mm          | mm       |
| Jan   | 468.47   | 285.94 | 179.68       | 111.51      | 55.62    |
| Feb   | 388.94   | 233.26 | 158.87       | 101.66      | 62.43    |
| Mar   | 320.70   | 173.09 | 150.90       | 110.79      | 59.33    |
| Apr   | 105.94   | 43.15  | 64.02        | 70.73       | 25.71    |
| May   | 23.03    | 5.52   | 17.67        | 33.54       | 6.71     |
| Jun   | 1.90     | 0.72   | 1.18         | 17.04       | 2.78     |
| Jul   | 0.88     | 0.00   | 0.88         | 13.59       | 1.59     |
| Aug   | 3.22     | 0.98   | 2.40         | 12.88       | 1.01     |
| Sep   | 14.97    | 2.33   | 12.78        | 18.96       | 0.70     |
| Oct   | 66.29    | 15.38  | 51.37        | 46.45       | 0.66     |
| Nov   | 141.64   | 43.34  | 99.83        | 81.30       | 1.26     |
| Dec   | 271.22   | 133.48 | 139.74       | 97.03       | 13.51    |





| Units                    | mm                               | mm  | mm                               | mm  | mm                           |
|--------------------------|----------------------------------|---|----------------------------------|---|------------------------------|
| Jan                      | 75.88                            | 20.16                                     | 55.69                            | 55.30                                     | 6.20                         |
| Feb                      | 119.77                           | 49.91                                     | 71.11                            | 57.98                                     | 7.98                         |
| Mar                      | 112.35                           | 39.68                                     | 74.22                            | 62.73                                     | 12.49                        |
| Apr                      | 116.05                           | 44.18                                     | 73.77                            | 56.44                                     | 17.68                        |
| May                      | 115.40                           | 43.71                                     | 73.94                            | 47.96                                     | 25.17                        |
| Jun                      | 128.78                           | 56.47                                     | 74.60                            | 43.15                                     | 31.62                        |
| Jul                      | 70.41                            | 18.97                                     | 53.41                            | 44.36                                     | 22.26                        |
| Aug                      | 54.91                            | 17.12                                     | 38.85                            | 44.74                                     | 14.79                        |
| Sep                      | 56.78                            | 12.45                                     | 45.12                            | 47.97                                     | 7.81                         |
| Oct                      | 66.68                            | 12.38                                     | 55.25                            | 55.08                                     | 6.34                         |
| Nov                      | 80.69                            | 18.21                                     | 63.71                            | 60.12                                     | 6.56                         |
| Dec                      | 65.96                            | 11.66                                     | 55.29                            | 59.85                                     | 6.72                         |
| Sep<br>Oct<br>Nov<br>Dec | 56.78<br>66.68<br>80.69<br>65.96 | 17.12<br>12.45<br>12.38<br>18.21<br>11.66 | 45.12<br>55.25<br>63.71<br>55.29 | 47.97<br>47.97<br>55.08<br>60.12<br>59.85 | 7.81<br>6.34<br>6.56<br>6.72 |

Figure C.2 Water Balance Elements – Clay, Uniform, High Rainfall



| Month | Rainfall | Runoff | Infiltration | Evaporation | Drainage |
|-------|----------|--------|--------------|-------------|----------|
| Units | mm       | mm     | mm           | mm          | mm       |
| Jan   | 101.71   | 33.07  | 68.48        | 65.16       | 7.19     |
| Feb   | 97.41    | 33.92  | 64.30        | 61.14       | 5.84     |
| Mar   | 65.43    | 16.38  | 49.76        | 53.11       | 7.70     |
| Apr   | 45.80    | 13.45  | 33.35        | 39.71       | 6.68     |
| May   | 55.18    | 21.40  | 34.70        | 36.66       | 5.57     |
| Jun   | 31.07    | 5.84   | 25.80        | 31.63       | 4.05     |
| Jul   | 25.84    | 3.51   | 22.81        | 30.18       | 2.58     |
| Aug   | 21.75    | 1.23   | 20.82        | 28.35       | 1.98     |
| Sep   | 28.56    | 2.37   | 26.65        | 34.43       | 1.44     |
| Oct   | 60.12    | 9.98   | 51.01        | 48.70       | 1.65     |
| Nov   | 82.77    | 22.72  | 61.12        | 57.60       | 2.38     |
| Dec   | 97.33    | 22.23  | 76.31        | 70.80       | 4.71     |

Figure C.3

Water Balance Elements - Clay, Moderate, Summer Dominant Rainfall



| Month | Rainfall | Runoff | Infiltration | Evaporation | Drainage |
|-------|----------|--------|--------------|-------------|----------|
| Units | mm       | mm     | mm           | mm          | mm       |
| Jan   | 20.46    | 3.05   | 17.30        | 26.85       | 1.64     |
| Feb   | 17.45    | 3.01   | 14.53        | 22.12       | 0.96     |
| Mar   | 23.17    | 3.82   | 19.61        | 26.95       | 0.85     |
| Apr   | 28.86    | 1.95   | 27.23        | 27.69       | 0.75     |
| May   | 43.10    | 2.18   | 41.79        | 35.15       | 0.95     |
| Jun   | 54.48    | 6.20   | 50.08        | 32.10       | 3.83     |
| Jul   | 52.28    | 3.94   | 50.45        | 36.59       | 11.62    |
| Aug   | 50.36    | 3.16   | 49.15        | 44.57       | 13.49    |
| Sep   | 46.39    | 3.62   | 43.95        | 48.31       | 10.27    |
| Oct   | 33.83    | 3.35   | 31.01        | 43.30       | 5.77     |
| Nov   | 24.37    | 1.17   | 23.49        | 34.76       | 2.85     |
| Dec   | 26.58    | 3.66   | 23.21        | 32.55       | 1.88     |

Figure C.4 Water Balance Elements – Clay, Low, Winter Dominant Rainfall

### C.2 Clay Loam Models



| Month | Rainfall | Runoff | Infiltration | Evaporation | Drainage |
|-------|----------|--------|--------------|-------------|----------|
| Units | mm       | mm     | mm           | mm          | mm       |
| Jan   | 468.47   | 88.42  | 374.06       | 115.35      | 249.42   |
| Feb   | 388.94   | 66.50  | 322.53       | 104.72      | 226.19   |
| Mar   | 320.70   | 50.25  | 270.75       | 114.68      | 188.52   |
| Apr   | 105.94   | 6.36   | 99.63        | 75.33       | 68.26    |
| May   | 23.03    | 0.26   | 22.77        | 36.23       | 11.76    |
| Jun   | 1.90     | 0.00   | 1.90         | 18.06       | 4.66     |
| Jul   | 0.88     | 0.00   | 0.88         | 13.90       | 2.39     |
| Aug   | 3.22     | 0.00   | 3.22         | 13.27       | 1.49     |
| Sep   | 14.97    | 0.00   | 14.97        | 19.85       | 1.18     |
| Oct   | 66.29    | 0.61   | 65.68        | 50.12       | 4.31     |
| Nov   | 141.64   | 1.16   | 140.49       | 90.26       | 20.03    |
| Dec   | 271.22   | 35.40  | 235.51       | 104.53      | 100.15   |

Figure C.5 Water Balance Elements – Clay Loam, Tropical, Very High Rainfall



| Units | mm     | mm   | mm     | mm    | mm    |
|-------|--------|------|--------|-------|-------|
| Jan   | 75.88  | 3.99 | 70.84  | 58.33 | 16.37 |
| Feb   | 119.77 | 8.29 | 111.48 | 61.89 | 37.34 |
| Mar   | 112.35 | 2.58 | 109.84 | 67.29 | 48.02 |
| Apr   | 116.05 | 2.74 | 113.31 | 59.52 | 58.05 |
| May   | 115.40 | 4.12 | 111.32 | 49.57 | 65.56 |
| Jun   | 128.78 | 5.89 | 122.90 | 43.76 | 81.13 |
| Jul   | 70.41  | 1.10 | 69.30  | 45.38 | 41.85 |
| Aug   | 54.91  | 3.03 | 51.95  | 46.35 | 29.12 |
| Sep   | 56.78  | 0.18 | 56.60  | 50.17 | 17.92 |
| Oct   | 66.68  | 0.02 | 66.66  | 57.67 | 16.20 |
| Nov   | 80.69  | 1.52 | 79.27  | 62.95 | 19.43 |
| Dec   | 65.96  | 0.35 | 65.62  | 63.57 | 14.97 |
|       |        |      |        |       |       |

| Figuro C 6 | Water Balance Elements Clay Learn Uniform High Painfall |
|------------|---|
|            | Vale Dalance Elements – Clav Luam, Unitum, num Raiman   |
|            |   |



Figure C.7 Water Balance Elements – Clay Loam, Moderate, Summer Dominant Rainfall



| Month | Rainfall | Runoff | Infiltration | Evaporation | Drainage |
|-------|----------|--------|--------------|-------------|----------|
| Units | mm       | mm     | mm           | mm          | mm       |
| Jan   | 20.46    | 0.00   | 20.18        | 28.60       | 3.16     |
| Feb   | 17.45    | 0.00   | 17.45        | 23.46       | 2.00     |
| Mar   | 23.17    | 0.06   | 23.11        | 28.61       | 2.86     |
| Apr   | 28.86    | 0.00   | 28.86        | 28.40       | 1.81     |
| May   | 43.10    | 0.00   | 43.10        | 35.74       | 2.33     |
| Jun   | 54.48    | 0.00   | 54.48        | 32.34       | 6.77     |
| Jul   | 52.28    | 0.00   | 52.28        | 36.79       | 16.69    |
| Aug   | 50.36    | 0.00   | 50.36        | 45.08       | 17.47    |
| Sep   | 46.39    | 0.00   | 46.39        | 49.31       | 13.72    |
| Oct   | 33.83    | 0.00   | 33.83        | 44.46       | 7.77     |
| Nov   | 24.37    | 0.00   | 24.37        | 35.62       | 4.86     |
| Dec   | 26.58    | 0.00   | 26.58        | 33.77       | 3.34     |

Figure C.8 Water Balance Elements – Clay Loam, Low, Winter Dominant Rainfall

### C.3 Loamy Sand Models



| month | Numan  | nunon | minutation | Lupplucion | Dramage |
|-------|--------|-------|------------|------------|---------|
| Units | mm     | mm    | mm         | mm         | mm      |
| Jan   | 468.47 | 0.00  | 462.05     | 114.95     | 337.33  |
| Feb   | 388.94 | 0.35  | 388.58     | 104.29     | 292.94  |
| Mar   | 320.70 | 0.00  | 320.70     | 114.10     | 239.29  |
| Apr   | 105.94 | 0.00  | 105.94     | 74.02      | 78.25   |
| May   | 23.03  | 0.00  | 23.03      | 35.03      | 15.20   |
| Jun   | 1.90   | 0.00  | 1.90       | 17.16      | 6.57    |
| Jul   | 0.88   | 0.00  | 0.88       | 13.27      | 3.67    |
| Aug   | 3.22   | 0.00  | 3.22       | 12.80      | 2.38    |
| Sep   | 14.97  | 0.00  | 14.97      | 19.56      | 1.77    |
| Oct   | 66.29  | 0.00  | 66.29      | 50.07      | 4.96    |
| Nov   | 141.64 | 0.00  | 141.64     | 90.23      | 18.45   |
| Dec   | 271.22 | 0.00  | 270.59     | 104.03     | 133.46  |





| Figure C.10 | Water Balance Elements - Loamy Sand, Uniform, High Rainfall |
|-------------|---|

66.68

80.69

65.96

56.87

62.14

62.56

16.99

21.56

16.70

0.00

0.00

0.00

Oct

Nov

Dec

66.68

80.69

65.96



Figure C.11 Water Balance Elements – Loamy Sand, Moderate Summer Dominant Rainfall



| month | naman | nunon | minutation | Liaporation | Dramage |
|-------|-------|-------|------------|-------------|---------|
| Units | mm    | mm    | mm         | mm          | mm      |
| Jan   | 20.46 | 0.00  | 20.18      | 28.01       | 4.20    |
| Feb   | 17.45 | 0.00  | 17.45      | 22.95       | 2.63    |
| Mar   | 23.17 | 0.00  | 23.17      | 28.17       | 3.45    |
| Apr   | 28.86 | 0.00  | 28.86      | 28.02       | 2.46    |
| May   | 43.10 | 0.00  | 43.10      | 35.48       | 2.72    |
| Jun   | 54.48 | 0.00  | 54.48      | 32.21       | 6.07    |
| Jul   | 52.28 | 0.00  | 52.28      | 36.64       | 15.61   |
| Aug   | 50.36 | 0.00  | 50.36      | 44.45       | 18.20   |
| Sep   | 46.39 | 0.00  | 46.39      | 48.35       | 14.61   |
| Oct   | 33.83 | 0.00  | 33.83      | 43.41       | 9.39    |
| Nov   | 24.37 | 0.00  | 24.37      | 34.82       | 6.02    |
| Dec   | 26.58 | 0.00  | 26.58      | 33.14       | 4.33    |
|       |       |       |            |             |         |

Figure C.12 Water Balance Elements – Loamy Sand, Low, Winter Dominant Rainfall

### C.4 Sand Models



| 1.90   | 0.00   | 1.90  | 14.92  | 11.38   |
|--------|--|---|--|---|
| 0.88   | 0.00   | 0.88  | 11.69  | 6.93  |
| 3.22   | 0.00   | 3.22  | 11.57  | 4.60  |
| 14.97  | 0.00   | 14.97   | 18.59  | 3.33  |
| 66.29  | 0.00   | 66.29   | 48.99  | 5.43  |
| 141.64 | 0.00   | 141.64  | 88.15  | 13.69   |
| 271.22 | 1.93   | 268.66  | 101.40   | 117.96  |
|        | 1.90<br>0.88<br>3.22<br>14.97<br>66.29<br>141.64<br>271.22 | 1.90 0.00   0.88 0.00   3.22 0.00   14.97 0.00   66.29 0.00   141.64 0.00   271.22 1.93 | 1.90 0.00 1.90   0.88 0.00 0.88   3.22 0.00 3.22   14.97 0.00 14.97   66.29 0.00 66.29   141.64 0.00 141.64   271.22 1.93 268.66 | 1.90 0.00 1.90 14.92   0.88 0.00 0.88 11.69   3.22 0.00 3.22 11.57   14.97 0.00 14.97 18.59   66.29 0.00 66.29 48.99   141.64 0.00 141.64 88.15   271.22 1.93 268.66 101.40 |

Figure C.13 Water Balance Elements – Sand, Tropical, Very High Rainfall



| May 115.40 0.00 115.40 46.85 73.18   Jun 128.78 0.88 127.90 41.98 84.57   Jul 70.41 0.00 70.41 42.21 52.94   Aug 54.91 0.00 54.91 41.81 39.76   Sep 56.78 0.00 56.78 46.13 24.37   Oct 66.68 0.00 66.68 54.06 19.22   Nov 80.69 0.00 80.69 59.29 24.10   Dec 65.96 0.00 65.96 59.28 20.73 |  | Apr | 116.05 | 0.00 | 116.05 | 56.31 | 62.68 |
|---|--|-----|--------|------|--------|-------|-------|
| Jun 128.78 0.88 127.90 41.98 84.57   Jul 70.41 0.00 70.41 42.21 52.94   Aug 54.91 0.00 54.91 41.81 39.76   Sep 56.78 0.00 56.78 46.13 24.37   Oct 66.68 0.00 66.68 54.06 19.22   Nov 80.69 0.00 80.69 59.29 24.10   Dec 65.96 0.00 65.96 59.28 20.73                                      |  | May | 115.40 | 0.00 | 115.40 | 46.85 | 73.18 |
| Jul 70.41 0.00 70.41 42.21 52.94   Aug 54.91 0.00 54.91 41.81 39.76   Sep 56.78 0.00 56.78 46.13 24.37   Oct 66.68 0.00 66.68 54.06 19.22   Nov 80.69 0.00 80.69 59.29 24.10   Dec 65.96 0.00 65.96 59.28 20.73   |  | Jun | 128.78 | 0.88 | 127.90 | 41.98 | 84.57 |
| Aug 54.91 0.00 54.91 41.81 39.76   Sep 56.78 0.00 56.78 46.13 24.37   Oct 66.68 0.00 66.68 54.06 19.22   Nov 80.69 0.00 80.69 59.29 24.10   Dec 65.96 0.00 65.96 59.28 20.73  |  | Jul | 70.41  | 0.00 | 70.41  | 42.21 | 52.94 |
| Sep 56.78 0.00 56.78 46.13 24.37   Oct 66.68 0.00 66.68 54.06 19.22   Nov 80.69 0.00 80.69 59.29 24.10   Dec 65.96 0.00 65.96 59.28 20.73   |  | Aug | 54.91  | 0.00 | 54.91  | 41.81 | 39.76 |
| Oct 66.68 0.00 66.68 54.06 19.22   Nov 80.69 0.00 80.69 59.29 24.10   Dec 65.96 0.00 65.96 59.28 20.73  |  | Sep | 56.78  | 0.00 | 56.78  | 46.13 | 24.37 |
| Nov 80.69 0.00 80.69 59.29 24.10   Dec 65.96 0.00 65.96 59.28 20.73   |  | Oct | 66.68  | 0.00 | 66.68  | 54.06 | 19.22 |
| Dec 65.96 0.00 65.96 59.28 20.73  |  | Nov | 80.69  | 0.00 | 80.69  | 59.29 | 24.10 |
|   |  | Dec | 65.96  | 0.00 | 65.96  | 59.28 | 20.73 |

Figure C.14 Water Balance Elements – Sand, Uniform, High Rainfall



Figure C.15 Water Balance Elements – Sand, Moderate, Summer Dominant Rainfall

97.33

73.50

23.61

0.00

97.33

Dec



| Feb | 17.45 | 0.00 | 17.45 | 21.66 | 4.26  |
|-----|-------|------|-------|-------|-------|
| Mar | 23.17 | 0.00 | 23.17 | 26.92 | 4.77  |
| Apr | 28.86 | 0.00 | 28.86 | 27.03 | 3.97  |
| May | 43.10 | 0.00 | 43.10 | 34.65 | 3.79  |
| Jun | 54.48 | 0.00 | 54.48 | 31.81 | 5.56  |
| Jul | 52.28 | 0.00 | 52.28 | 36.03 | 13.95 |
| Aug | 50.36 | 0.00 | 50.36 | 42.65 | 19.84 |
| Sep | 46.39 | 0.00 | 46.39 | 45.65 | 16.68 |
| Oct | 33.83 | 0.00 | 33.83 | 40.66 | 13.05 |
| Nov | 24.37 | 0.00 | 24.37 | 32.79 | 8.88  |
| Dec | 26.58 | 0.00 | 26.58 | 31.48 | 6.78  |

Figure C.16 Water Balance Elements – Sand, Low, Winter Dominant Rainfall

# Appendix D Untreated Flux Graphs, by Rainfall and Soil Type

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| Figure D.9 | PFOA leaching over time, assuming high K <sub>d</sub>      | D-6 |



Figure D.1

PFOS leaching over time, assuming Low K<sub>d</sub>



Figure D.2 PFHxS leaching over time, assuming Low K<sub>d</sub>



Figure D.3 PFOA leaching over time, assuming Low K<sub>d</sub>



#### D.2 Moderate K<sub>d</sub> Models

Figure D.4 PFOS leaching over time, assuming moderate K<sub>d</sub>



Figure D.5 PFHxS leaching over time, assuming moderate K<sub>d</sub>



Figure D.6 PFOA leaching over time, assuming moderate K<sub>d</sub>



Figure D.7 PFOS leaching over time, assuming high K<sub>d</sub>



Figure D.8 PFHxS leaching over time, assuming high K<sub>d</sub>





PFOA leaching over time, assuming high  $K_d$ 

# Appendix E Treatment response graphs

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### E.1 Low K<sub>d</sub> Models























Figure E.6 Low K<sub>d</sub> PFOA leaching from Clay Loam profiles











Figure E.9 Low K<sub>d</sub> PFOA leaching from Loamy Sand profiles













#### E.2 Moderate Kd Models







Figure E.14 Moderate K<sub>d</sub> PFHxS leaching from Clay profiles



Figure E.15 Moderate K<sub>d</sub> PFOA leaching from Clay profiles



Figure E.16 Moderate K<sub>d</sub> PFOS leaching from Clay Loam profiles



Figure E.17 Moderate K<sub>d</sub> PFHxS leaching from Clay Loam profiles



Figure E.18 Moderate K<sub>d</sub> PFOA leaching from Clay Loam profiles



Figure E.19 Moderate K<sub>d</sub> PFOS leaching from Loamy Sand profiles



Figure E.20 Moderate K<sub>d</sub> PFHxS leaching from Loamy Sand profiles



Figure E.21 Moderate K<sub>d</sub> PFOA leaching from Loamy Sand profiles


Figure E.22 Moderate K<sub>d</sub> PFOS leaching from Sand profiles



Figure E.23 Moderate K<sub>d</sub> PFHxS leaching from Sand profiles



Figure E.24 Moderate K<sub>d</sub> PFOA leaching from Sand profiles

## E.3 High K<sub>d</sub> models

























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Figure E.32 High K<sub>d</sub> PFHxS leaching from Loamy Sand profiles

















| Appendix F Vertical Section Time Series Pictorials |  |      |
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# JOURNAL OF CONTAMINANT HYDROLOGY

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ISSN: 0169-7722

# DESCRIPTION

The *Journal of Contaminant Hydrology* is an international journal publishing scientific articles contributing to a broad understanding of contamination of water resources. Emphasis is placed on investigations of the physical, chemical, and biological processes influencing the behaviour and fate of organic and inorganic contaminants in the aqueous environment including ecological impacts. Water-based science, technology and management approaches that monitor, assess, control and mitigate contamination and its eco-environmental impacts at multiple scales are invited. Broad latitude is allowed in identifying contaminants of interest, and includes legacy and emerging pollutants, nutrients, nanoparticles, microorganisms (e.g., bacteria, viruses, and protozoa), microplastics, and various constituents associated with energy production (e.g., methane, carbon dioxide, and hydrogen sulfide).

The journal's scope embraces a wide range of topics that include: surface and subsurface hydrology as it relates to contamination; experimental and computational investigations of contaminant sorption, diffusion, biological and chemical transformation, volatilization and transport in the surface and subsurface; characterization of soil and sediment properties only as they influence contaminant behaviour; development and testing of mathematical models of contaminant behaviour; innovative techniques for restoration of contaminated sites; development of new tools or techniques for monitoring the extent of soil, sediment, and water contamination; development of mathematical models and system analysis techniques for understanding and managing surface and subsurface water resources systems including hyporheic zone processes; analyses of interactions between water-use activities and the environment; carbon sequestration and turnover; and water contamination issues associated with energy production.

#### Types of paper

There are some types of papers that are *not suitable* for publication in the journal, namely:

Environmental monitoring. We are pleased to see field data, but we do not publish reports of, for example, unusual observations in the field unless they are interpreted at a process level. Similarly, we do not act as a public repository for datasets unless they are interpreted.

Case studies. We will not publish case studies unless they provide insight into processes relevant to other sites or conditions. Thus, a paper based on a particular site must draw out principles, prove a conceptual model, or develop and test a method; these principles, models, or methods must have broader applicability than to a site of study.

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