

*The impact of increased flooding occurrence on the mobility of potentially toxic elements in floodplain soil – a review*

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2 The impact of increased flooding occurrence on the  
3 mobility of potentially toxic elements in floodplain soil  
4 – A review

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28 Abstract

29 The frequency and duration of flooding events are increasing due to land-use changes increasing run-  
30 off of precipitation, and climate change causing more intense rainfall events. Floodplain soils situated  
31 downstream of urban or industrial catchments, which were traditionally considered a sink of  
32 potentially toxic elements (PTEs) arriving from the river reach, may now become a source of legacy  
33 pollution to the surrounding environment if PTEs are mobilised by unprecedented flooding events.

34 When a soil floods, the mobility of PTEs can increase or decrease due to the net effect of five key  
35 processes; (i) the soil redox potential decreases which can directly alter the speciation, and hence  
36 mobility, of redox sensitive PTEs (e.g. Cr, As), (ii) pH increases which usually decreases the mobility of  
37 metal cations (e.g.  $\text{Cd}^{2+}$ ,  $\text{Cu}^{2+}$ ,  $\text{Ni}^{2+}$ ,  $\text{Pb}^{2+}$ ,  $\text{Zn}^{2+}$ ), (iii) dissolved organic matter (DOM) increases, which  
38 chelates and mobilises PTEs, (iv) Fe and Mn hydroxides undergo reductive dissolution, releasing  
39 adsorbed and co-precipitated PTEs, and (v) sulphate is reduced and PTEs are immobilised due to  
40 precipitation of metal sulphides. These factors may be independent mechanisms, but they interact  
41 with one another to affect the mobility of PTEs, meaning the effect of flooding on PTE mobility is not  
42 easy to predict. Many of the processes involved in mobilising PTEs are microbially mediated,  
43 temperature dependent and the kinetics are poorly understood.

44 Soil mineralogy and texture are properties that change spatially and will affect how the mobility of  
45 PTEs in a specific soil may be impacted by flooding. As a result, knowledge based on one river  
46 catchment may not be particularly useful for predicting the impacts of flooding at another site. This  
47 review provides a critical discussion of the mechanisms controlling the mobility of PTEs in floodplain  
48 soils. It summarises current understanding, identifies limitations to existing knowledge, and highlights  
49 requirements for further research.

50

51 **Key words;** floodplain soil, flooding, climate change, potentially toxic elements, contamination,  
52 mobility

53

54 1. Introduction

55         Flooding is a major event that currently affects an estimated 20 to 300 million people per year,  
56 and accounts for around 40% of natural disasters occurring worldwide, threatening both social  
57 security and sustainable development (Euripidou and Murray, 2004; Hirabayashi and Kanae, 2009).  
58 Alterations to land use and land cover are having widespread implications for catchment  
59 characteristics; with soil sealing and impermeable surfaces increasing surface run-off, as well as a  
60 reduction of natural buffering environments such as forests and wetlands, meaning there is less  
61 capacity to accommodate flood waters in the same river reach (Dadson et al., 2017; Kundzewicz et al.,  
62 2014). There is growing evidence, from climate models, that short-term extreme weather events (e.g.  
63 high-frequency rainstorms, heat waves and wind storms) are likely to become increasingly frequent  
64 in many parts of the world, threatening the long-term functioning of the terrestrial system (Harvey et  
65 al., 2019; Kharin et al., 2007; Madsen et al., 2014; Pendergrass, 2018; Stagl et al., 2014). It is likely that  
66 populations will experience warmer and drier summers, and an increase in the intensity of heavy  
67 rainfall, contributing to more frequent pluvial, fluvial, groundwater or coastal flooding, and resulting  
68 in the occasional inundation of land that has rarely been flooded in the past (Barber et al., 2017;  
69 Kundzewicz et al., 2014; Schaller et al., 2016). The likelihood of flooding is also determined by  
70 antecedent soil moisture conditions. The proportion of soil pore space that is filled with water at any  
71 given time is largely dependent on local hydrological processes and stores including; infiltration,  
72 surface and sub-surface runoff (when rainfall intensity exceeds infiltration capacity), redistribution  
73 and drainage to/from groundwater, evaporation, and transpiration (Stagl et al., 2014).

74         Soil contamination is among the most serious threats to soil resources globally (Nriagu et al., 2007;  
75 Srivastava et al., 2017; Tóth et al., 2016b). Since many commercial, industrial, residential and  
76 agricultural developments have historically been situated adjacent to rivers; they contribute to the  
77 contamination of river sediments, and these sediments are often deposited onto the floodplain soils  
78 downstream by overbanking river water during a flooding event (Arnell et al., 2015; Nshimiyimana et

79 al., 2014; Zhao et al., 1999). Here we use the term PTEs, also referred to in the literature as ‘trace  
80 elements’ or ‘heavy metals’, to encompass all metals, metalloids, non-metals and other inorganic  
81 elements in the soil–plant–animal system, of which their mobility and potential toxicity to that system  
82 and/or humans is largely dependent upon their concentration, bioavailability and chemical form  
83 (Hooda, 2010; Rodgers et al., 2015). The term "mobility" is a concept that has been frequently used  
84 to estimate the risk of contamination from the soil to the surrounding environment by PTEs  
85 (Domergue and Vedy, 1992). Here we define mobile PTEs as those elements that are dissolved in soil  
86 porewater or associated with colloids and thus capable of leaching from the soil profile, or being taken  
87 up into plants or soil organisms. The mobility and subsequent fate of PTEs in periodically (occasionally)  
88 flooded soils (such as floodplain soils) are imperfectly understood. The legacy of historic  
89 contamination and continuing increases in emissions from urban activities pose a serious  
90 environmental threat globally (de Souza Machado et al., 2016; Srivastava et al., 2017). Human actions  
91 to mitigate and adapt to the impacts of climate change may influence the fate of contaminants, with  
92 climate change itself also potentially affecting the toxicity of the contaminants within the environment  
93 (Stahl et al., 2013).

94 The aim of this literature review is to provide an understanding of the factors involved in the  
95 mobility of PTEs in soil by pulling together interdisciplinary knowledge in this area. The review will first  
96 consider in more detail the expected changes to global rainfall patterns, the implications of these  
97 changes for flooding, and the role that floodplains play during inundation, as well as the changes they  
98 undergo. The review will then showcase how PTEs have entered the floodplain soil and how flooding  
99 influences soil biogeochemical processes which, in turn, influence PTEs mobility, using examples from  
100 the literature. Finally, this knowledge is used to identify gaps that will help to make recommendations  
101 for future research into the effects of flooding on the mobility and fate of PTEs.

102

103 1.1. Expected changes to global rainfall patterns and implications for flooding

104 Anthropogenic (human) activities including intensified land use; urbanisation, forestry,  
105 cultivation, and fossil energy use have increased atmospheric greenhouse gas concentrations which  
106 are driving changes in climate and leading to increases in rainfall intensity and surface run-off that are  
107 associated with increased flood risk (Bronstert, 2003; Chang and Franczyk, 2008; Kharin et al., 2007;  
108 Kundzewicz et al., 2014; Wheater and Evans, 2009). Mean global temperatures have risen by 1.1 °C  
109 since the end of the 19<sup>th</sup> century; the “Paris Climate Agreement” seeks to contain global mean  
110 temperatures well below 2°C and, ambitiously, below 1.5°C (Alfieri et al., 2017; Bronstert, 2003;  
111 Huddart et al., 2020; Mullan et al., 2019). The Intergovernmental Panel on Climate Change (IPCC) has  
112 predicted that under the A1B (medium) emissions scenario, temperatures will increase between 1.1  
113 and 6.4 °C by the year 2100, leading to an increase in atmospheric water holding capacity and therefore  
114 variations to seasonal rainfall (Arnell et al., 2015; Bell et al., 2012; Chan et al., 2014; Clemente et al.,  
115 2008; González-Alcaraz and van Gestel, 2015; Jenkins et al., 2009). It has been argued that we will  
116 experience an intensification of short-duration heavy rainfall events rather than a uniform increase in  
117 the daily average rainfall (Chan et al., 2014; Hirabayashi et al., 2008; Kharin et al., 2007; Kundzewicz  
118 et al., 2014).

119 An IPCC Special Report (SREX) on climate extremes (IPCC, 2012) assessed it is *likely* there have  
120 been statistically significant increases in the number of heavy precipitation events in more regions  
121 than significant decreases, with strong regional and sub-regional variation. The observed changes to  
122 precipitation extremes have been found to be far less spatially coherent or statistically significant  
123 compared with changes found in temperature extremes (Kundzewicz et al., 2014). Projected scenarios  
124 with 4°C warming showed more than 70% of the global population will face increased flood risk (Alfieri  
125 et al., 2017). Increases in flood frequency are expected in; Europe, America, Southeast Asia, eastern  
126 Africa, and Peninsular India. Populations in regions such as Bangladesh, Mumbai and Thailand are  
127 potentially at higher risk from flooding due to predicted increases in rainfall, coupled with changes in  
128 land use (e.g. irrigation schemes and construction of dams), and increasing population size requiring

129 rapid expansion of urban areas (Hirabayashi et al., 2013; Kundzewicz et al., 2014; Tockner et al., 2010).  
130 However, climate projections tend to have relatively low levels of model confidence, particularly for  
131 the prediction of fluvial floods because there is still relatively limited evidence and the causes of  
132 regional changes to flood occurrence are complex (Hirabayashi et al., 2013).

133 Flooding tends to be heterogeneous as it is affected not only by variability of the climatological  
134 and hydrological systems but also by land-use and the effect is has on the storage capacity of the  
135 receiving catchment (storage and drainage basin conditions). Changes to the characteristics of  
136 precipitation (the frequency, intensity and timing of rainfall) will have decisive implications for flood  
137 risk (Bronstert, 2003; Hirabayashi and Kanae, 2009; Kundzewicz et al., 2014). However, pre-existing  
138 high river levels and groundwater levels, as well as saturated soils are equally important to establish  
139 the capacity of the receiving catchment to cope with further rainfall (Maggioni and Massari, 2018;  
140 Wilby et al., 2008). The extent of flooding in a particular catchment will depend largely on the  
141 topography (variation in elevation), along with vegetation type, proportion of land used for cultivation  
142 and the extent of urbanised areas positioned upstream (Arnell et al., 2015; Bell et al., 2012; Bronstert,  
143 2003; Chang and Franczyk, 2008; Kundzewicz et al., 2014; Qiao et al., 2019). Urbanisation is a global  
144 issue; with more than half the world's population now living in cities, the process of urbanisation is  
145 leading to greater human occupation of floodplains, often with inadequate drainage planning  
146 (Kundzewicz et al., 2014; Pathirana et al., 2014).

147 The probability of flooding occurring in a particular region is often related to regional processes  
148 like El Niño Southern Oscillation (ENSO) cycle and the North Atlantic Oscillation (NAO) that, in turn,  
149 cause global impacts. The intensity (frequency and amplitude) of both ENSO and NAO are influenced  
150 by other modes of variability, for example; Pacific Decadal Oscillation (PDO) and Interdecadal Pacific  
151 Oscillation (IPO) which cause opposite atmospheric and sea surface temperatures and can therefore  
152 determine the magnitude of floods (Grimm and Tedeschi, 2009; Johnson et al., 2020). ENSO is a rapid  
153 warming of the sea surface temperature (by 1–5 °C) of the equatorial Pacific over the duration of a



154 few weeks, resulting in extreme rainfall and increased cyclone activity in some regions, and risk of  
155 drought and forest fires in others (Berz et al., 2001; Grimm and Tedeschi, 2009; Karl and Trenberth,  
156 2003; Kundzewicz et al., 2014; Tedeschi and Collins, 2016). Periods of extreme rainfall and subsequent  
157 flooding have been found to correlate with ENSO events in North and South America as well as in  
158 Africa (Berz et al., 2001; Brönnimann, 2007; Kundzewicz et al., 2014). NAO is an atmospheric pattern  
159 that affects the severity of winter temperatures and precipitation over Europe and eastern North  
160 America (Karl and Trenberth, 2003). Intense rainfall is a common cause of river basin flooding;  
161 however, in high latitude regions it is changes in temperatures altering the timing of seasonal  
162 snowmelt and causing glacier retreat that commonly causes flooding, for example in north-eastern  
163 Europe, Central and South America, and in polar regions such as the Russian Arctic (Blöschl et al., 2017;  
164 Hirabayashi et al., 2008; Kharin et al., 2007; Kundzewicz et al., 2014; Stagl et al., 2014). Rising global  
165 sea-level (11-16cm in the 20<sup>th</sup> century and a further 0.5m predicted this century) will certainly increase  
166 risk of flooding caused by tidal processes, with current estimates that 630 million people live on land  
167 below projected annual flood levels for 2100 (Kulp and Strauss, 2019). While there is uncertainty  
168 regarding the effect that future climate change will have on river levels (Prudhomme and Davies,  
169 2009), changes made to land-use, and land cover, for example by urbanisation, will drive changes in  
170 the local climate (at the kilometre scale) influencing the hydrometeorological regime and resulting in  
171 more flooding (Foley et al., 2005; Hirabayashi and Kanae, 2009). Pathirana et al. (2014), using a 3D  
172 atmospheric model coupled with a land surface model (WRF-ARW) in southern India, found that in  
173 three out of four simulated cases there was a significant increase in local extreme rainfall when  
174 urbanisation in the area increased. This work was conducted in southern India, however the model  
175 could be applied and validated to other regions to establish whether this correlation is found globally.

176

## 177 1.2. The role of floodplains during floods

178 Floodplains are by definition dynamic environments subjected to fluctuations between flooding  
179 and drying (Vijver et al., 2007). They are distinctive landscape features, often on low-lying ground, and

180 characterised by a high spatio-temporal heterogeneity (Schulz-Zunkel et al., 2015; Stuart and  
181 Lapworth, 2011; Tockner et al., 2010; Tockner and Stanford, 2002). Periodic overbank inundation from  
182 the adjacent watercourse, overland flow, subsurface flow, and changes to the groundwater levels  
183 result in a constantly changing water balance and degree of floodplain saturation (Stuart and  
184 Lapworth, 2011; Tockner and Stanford, 2002). Floodplain topography and variations in elevation are  
185 usually slight but have an important effect on the degree of soil saturation across the floodplain,  
186 depending on the overall water balance from surface and sub-surface run-off (Arnell et al., 2015;  
187 Kundzewicz et al., 2014; Qiao et al., 2019).

188 There are various sources and pathways of water that can lead to the inundation of a floodplain,  
189 including lateral overflow of rivers or lakes, rising groundwater, upland sources, and direct  
190 precipitation. Several different factors and water sources normally contribute to a flooding event, thus  
191 making flooding a complex phenomenon to study (Junk et al., 1989; Tockner and Stanford, 2002).  
192 Fluvial flooding tends to occur when excessive rain falls over an extended period of time, leading to a  
193 river exceeding its capacity, or because of heavy snow that subsequently melts and, via surface run-  
194 off, rapidly fills the river channels when infiltration is low because of frozen soils below the snow layer  
195 (Blöschl et al., 2017).

196 River flow regimes are affected by the increased rainfall and this also has the potential to affect  
197 erosion and generate additional sediment loads and particulate organic matter (POM) for deposition  
198 within river channels, lakes and estuaries (Arnell et al., 2015; Le Gall et al., 2018; Rinklebe and Du  
199 Laing, 2011). Intense rainfall over a short timescale (usually less than six hours i.e. “flash floods”) can  
200 also cause rivers to overbank leading to an intense, high velocity torrent of water that moves through  
201 river beds, disturbing river sediments and potentially bringing more PTEs contamination with the  
202 flood water, greatly influencing the contaminated status of the floodplain (Blöschl et al., 2017;  
203 Maggioni and Massari, 2018). The water inundating the floodplain contains dissolved matter (i.e. free  
204 ions, inorganic and organic complexes and uncharged molecules) as well as particulate matter (i.e.

205 large organic and inorganic polymers, oxides, clay minerals and organic matter) (Kirk, 2004). The  
206 sediment loads travel at different rates due to their particle size, which reflects the texture of the river  
207 bed and bank (Malmon et al., 2004). Approximately 90% of PTEs load has been associated with  
208 sediment particles, with dissolved PTEs playing a comparatively minor role in pollutant transfer to  
209 floodplains (Ciszewski and Grygar, 2016). There have been many fluvial geomorphology studies  
210 showing how erosion and sedimentation have been influenced by climatic variability in the past (e.g.  
211 Lewin and Macklin, 2010; Macklin and Rumsby, 2007; Mullan et al., 2019), indicating that rivers are  
212 sensitive to climatic change (Arnell et al., 2015). Fluvial flooding is receiving increased scientific and  
213 political interest because of the potential impact that climate change may have on this type of  
214 flooding, with climate model projections showing an increased flood risk at a global scale  
215 (Pappenberger et al., 2012; Wilby et al., 2008).

216 In floodplains that are underlain by permeable deposits, increased rainfall causes groundwater to  
217 rise (leading to groundwater flooding), which can result from direct rainfall recharge, when the soil  
218 water storage potential is exceeded, as well as flow into the floodplain sediments from rivers with  
219 high water levels, and from areas inundated with fluvial flooding. However, good hydraulic connection  
220 between river and aquifer means that the aquifer can drain quickly as fluvial flood waters recess.  
221 Groundwater flooding in these settings is relatively short-lived compared with other groundwater  
222 flood settings, for example in chalk catchments (MacDonald et al., 2012).

223 With increased frequency of rainfall events predicted, it has become widely recognised that  
224 the storage of floodwater on floodplains can help to reduce the magnitude of a flood downstream.  
225 Thus, floodplains are useful for flood risk management (Acreman et al., 2003; Vink and Meeussen,  
226 2007). As a result, floodplains may be deliberately managed to allow flooding to occur through  
227 engineered soakaways in order to protect an urban residential area (Lane, 2017; Wheater and Evans,  
228 2009). It is important to understand the potential implications of these types of management practices

229 on mobilisation of PTEs that may be associated with the sediments deposited on the floodplain during  
230 past flooding events.

### 231 1.3. Changes that floodplain soils undergo during and after inundation

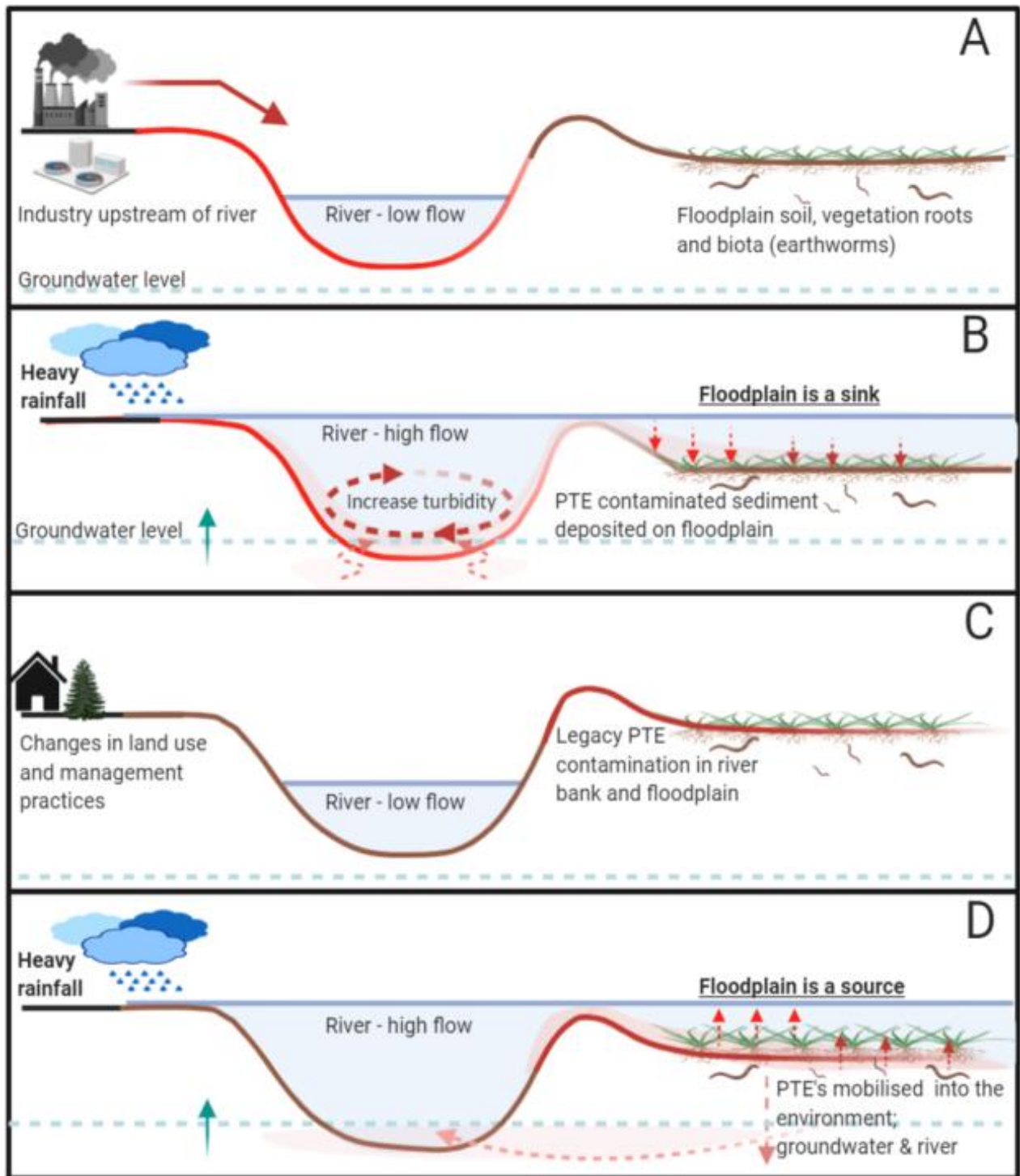
232 Extreme rainfall events leading to flooding have generally been found to alter soil physical and  
233 chemical properties and influence biological processes (Harvey et al., 2019). The fluctuations between  
234 inundation and subsequent drying, associated with periodically flooded soils, are major drivers of  
235 spatial and temporal differences in soil properties that affect the biogeochemical processes taking  
236 place in floodplain soils (Schulz-Zunkel et al., 2015; Tockner et al., 2010). These changes include; a  
237 decrease in redox potential ( $E_H$ ), which leads to, for example, reduction of iron (Fe) and manganese  
238 (Mn), which in turn can influence the soil pH (Rinklebe and Shaheen, 2017). Other processes affected  
239 include sulphur-cycling, changes to the presence of chelating agents such as dissolved organic carbon,  
240 mineralisation of POM and suppression of microbial activity (Poot et al., 2007; Puchalski, 2003; Schulz-  
241 Zunkel et al., 2015; Schulz-Zunkel and Krueger, 2009). Ibragimow, Walna, and Siepak (2013) showed,  
242 through analyses of fluvial samples before and after a flood, that the physicochemical properties (grain  
243 size,  $E_H$ , pH, POM, and calcium carbonate contents) as well as the total and available concentration of  
244 PTEs had changed. Harvey et al. (2019) found that after UK floods receded in the winter of 2013-14  
245 there was a decrease in the soil bulk density, pH and available P. The flood was found to have had a  
246 negative effect on the overlying vegetation and caused a shift in the microbial community structure.

247 Inundation during a flooding event can carry PTEs dissolved in rising groundwater and  
248 potentially contaminated suspended sediment from upstream overbanking water, depositing this  
249 onto the floodplain during a flood (Acreman et al., 2003; Bednářová et al., 2015; Du Laing et al., 2009;  
250 Gröngröft et al., 2005; Junk et al., 1989; Rudiš et al., 2009; Tockner and Stanford, 2002; Weber et al.,  
251 2009). Subsequently, this deposition of suspended riverine sediments/POM by flood water results in  
252 the floodplain topsoil becoming a sink for PTEs (Du Laing et al., 2009; Frohne et al., 2011;  
253 Nshimiyimana et al., 2014; Overesch et al., 2007; Rinklebe et al., 2007; Visser et al., 2012; Zhao and

254 Marriott, 2013). As a result, floodplain topsoil (uppermost 15cm) can often initially contain elevated  
255 concentrations of PTEs such as the metalloid; arsenic (As), and metals; chromium (Cr), copper (Cu),  
256 lead (Pb), and zinc (Zn), but later due to post-depositional reactions with organic matter/other organic  
257 components the PTEs concentrations will vary (Adekanmbi et al., 2020; Ciszewski and Grygar, 2016;  
258 Hurley et al., 2017; Izquierdo et al., 2013; Jiao et al., 2014; Kelly et al., 2020). When laboratory  
259 experiments are undertaken on samples gathered from floodplain site, soils are collected as single or  
260 composite samples, air or oven dried and then homogenised, resulting in a loss of soil stratigraphy  
261 and therefore the potential differences in PTEs concentration with depth may be unaccounted for  
262 (Ciszewski and Grygar, 2016). Zhao and Marriott, (2013) looked at PTEs concentrations along a vertical  
263 profile and found that there were peak values at varying depths; affected by translocation and  
264 duration of inundation. The process of breaking up of soil samples for laboratory experiments will  
265 make interpretation of PTEs levels difficult. Kelly et al. (2020) took intact soil cores to overcome this  
266 and more closely reflect natural samples, they too found the duration of inundation influenced the  
267 fate of PTEs.

268 The biological health of floodplain soils is important as they act as an interface between terrestrial  
269 and aquatic environments, therefore playing an important role in maintaining the environmental  
270 quality of surface waters (Izquierdo et al., 2013; Stuart and Lapworth, 2011). Artificial or constructed  
271 wetlands have been used for flood and pollution control; storing and filtering excess water to protect  
272 rivers from various kinds of runoff e.g. high nutrient loads from farm land (Blackwell and Pilgrim, 2011;  
273 Ellis et al., 2003; Rizzo et al., 2018). An example of this technology is demonstrated at the Rothamsted  
274 Research North Wyke experimental farm (Pulley and Collins, 2019). Even if river and groundwater  
275 water quality improves due to the implementation of more stringent environmental policy,  
276 contaminated floodplains remain as a legacy of historic upstream pollution (Bradley and Cox, 1990;  
277 Förstner, 2004; Kowalik et al., 2004). With increased frequency and duration of flooding, there is the  
278 possibility that changes to soil properties and biogeochemical processes will ultimately lead to the  
279 mobilisation of PTEs from floodplain soils (Ciszewski and Grygar, 2016; González-Alcaraz and van

280 Gestel, 2015). Therefore, historically contaminated floodplains may become a source of legacy  
281 pollution to the surrounding environment (Kelly et al., 2020; Pulchalski, 2003; Schulz-Zunkel and  
282 Krueger, 2009), as shown in Figure 1.



283 Figure 1: How floodplains may switch from being a sink of pollution to becoming a source of legacy  
 284 pollution: A) PTEs contaminated river sediment (red) due to industry in the catchment upstream, B)  
 285 heavy rainfall influences the receiving catchment (increased river flow and groundwater level), resulting  
 286 in flooding and the deposition of contaminated sediment onto the adjacent floodplain; dissolved  
 287 contaminants may also reach the floodplain surface via rising groundwater, C) Later, the river is  
 288 uncontaminated (brown) due to rising environmental quality standards, with legacy of PTEs  
 289 contamination (red) in the river bank and floodplain soil, D) heavy rainfall results in flooding of the  
 290 contaminated floodplain, mobilisation of the legacy PTEs by desorption and resuspended particulate  
 291 matter into the surrounding environment and thus making them potentially available for uptake by

292 *vegetation and soil organisms, as well as pollutant transfer leaching into the overlying flood water, the*  
293 *groundwater, and ultimately the river. Created with BioRender.com.*  
294

## 295 2.Impact of flooding on the mobility of potentially toxic elements in floodplain soil

### 296 2.1 PTEs in floodplain soil

297           Several PTEs are also essential nutrients that are required in low concentrations for healthy  
298 functioning and reproduction of microorganisms, plants, and animals, although may become toxic in  
299 high concentrations, these include; Cu, Cobalt (Co), Nickel (Ni), Vanadium (V), Zn, chlorine (Cl), Mn,  
300 Fe, boron (B), and molybdenum (Mo) (Adamo et al., 2014; Hooda, 2010; Wyszowska et al., 2013).  
301 Other PTEs are non-essential and can cause toxicity even when they are found at low concentrations,  
302 these include; As, Pb,) and mercury (Hg); (Adamo et al., 2014; Nriagu et al., 2007; Wuana et al., 2011;  
303 Wyszowska et al., 2013). Cadmium (Cd) is generally considered a non-essential element to soil  
304 organisms, but it has been found to be beneficial to some microalgae (Xu et al., 2008) Chromium can  
305 be considered a micronutrient but its toxicity depends on its valence state (i.e. Cr (VI) is the more  
306 mobile and toxic form compared with Cr (III)). Redox potential therefore not only affects the mobility  
307 of PTEs, but also their toxicity (Lee et al., 2005; Shahid et al., 2017). The consequences of PTEs  
308 contamination of soils are rarely observed with immediate effect, rather they tend to cause delayed  
309 adverse ecological changes, due to the fact that PTEs are persistent in the environment for long  
310 periods, non-biodegradable and can only be bio-transformed through complex physico-chemical and  
311 biological processes (Chrzan, 2016; Czech et al., 2014; Hooda, 2010). PTEs cause adverse ecological  
312 effects on plants and organisms such as impacting their activity, growth rate/yield, metabolism and  
313 reproduction, causing symptoms of physiological stress and potentially death. The extent of the  
314 adverse effect depends on the exposure route (ingestion, dermal absorption or uptake of pore water)  
315 and time, resistance (related to residence time of the PTEs in the environment) and detoxification  
316 mechanisms of the plant or animal (Alloway, 2013; Eggleton and Thomas, 2004; Ehlers and Loibner,  
317 2006; Hooda, 2010; Pan et al., 2018; Shahid et al., 2017; Winger et al., 1998). Leaching of PTEs from



318 the floodplain soil into the groundwater or river will also cause adverse effects to aquatic organism in  
319 these environments (Zia et al., 2018).

320 PTEs are either present naturally in the floodplain soil from the underlying or upstream  
321 geology and subsequent geogenic processes (e.g. weathering of parent material, emissions from  
322 volcanoes, forest fires) or introduced by anthropogenic sources, including solid and dissolved inputs  
323 from; aerial deposition, transport emissions, industrial, municipal and diffuse runoff from agricultural  
324 practices landfills and sewage treatment facilities (Alloway, 1995; Álvarez-Ayuso et al., 2012). PTEs can  
325 be adsorbed to colloidal suspended particulate material, transported in the river water and  
326 accumulate in the floodplain soil during inundation (Du Laing et al., 2009; Frohne et al., 2011;  
327 Peijnenburg et al., 2007; Rinklebe et al., 2007). PTEs have been found to be primarily associated with  
328 fine-grained clay or silt minerals and can reside in the floodplain for longer when compared with river  
329 sediments, as they are less likely to be susceptible to erosion (Lučić et al., 2019; Malmon et al., 2002).  
330 Contamination of the floodplain soil may result from a point source such as a sewage treatment  
331 facility, or from diffuse sources that have no specific point of discharge (e.g., agricultural applications).  
332 Impacts of diffuse pollution are difficult to predict as they can be affected by weather systems,  
333 meaning soils far from the source may be affected (Gregory et al., 2015; Neal et al., 1996). The  
334 anticipated changes to intense rainfall may result in increased delivery of diffuse pollution to rivers  
335 and groundwater (Arnell et al., 2015; Foulds et al., 2014), particularly as contaminated floodplain soils  
336 may become a diffuse source of pollution themselves during a flooding event (Schulz-Zunkel and  
337 Krueger, 2009).

## 338 2. 2. Influence of flooding on PTEs mobility

339 During a flooding event, biogeochemical processes occur in the floodplain soil at the oxic-  
340 anoxic interface and in the anoxic layers. The kinetics of these processes are of great importance  
341 because the location of the oxic-anoxic interface is subject to change due to fluctuating water table  
342 levels (Du Laing et al., 2009; Puchalski, 2003). In their review of trace metal behaviour in floodplain

343 sediments, Du Laing et al. (2009) state that the spatial occurrence of processes affecting metal mobility  
344 and availability is largely determined by the topography of the floodplain. Remobilisation of PTEs from  
345 sediments into the overlying water column during a flooding event depends on the flood regime; the  
346 frequency of these intense floods which flush or remobilise contaminated material as well as the  
347 duration or alternation of flood with dry spells (Arnell et al., 2015; Foulds et al., 2014; González-Alcaraz  
348 and van Gestel, 2015). Whilst research has suggested that the longer the flood duration, the greater  
349 the metal mobility (Shaheen et al., 2014a, 2014b), Stafford et al. (2018) suggest that even short  
350 periods of soil saturation can have an influence the solubility of PTEs.

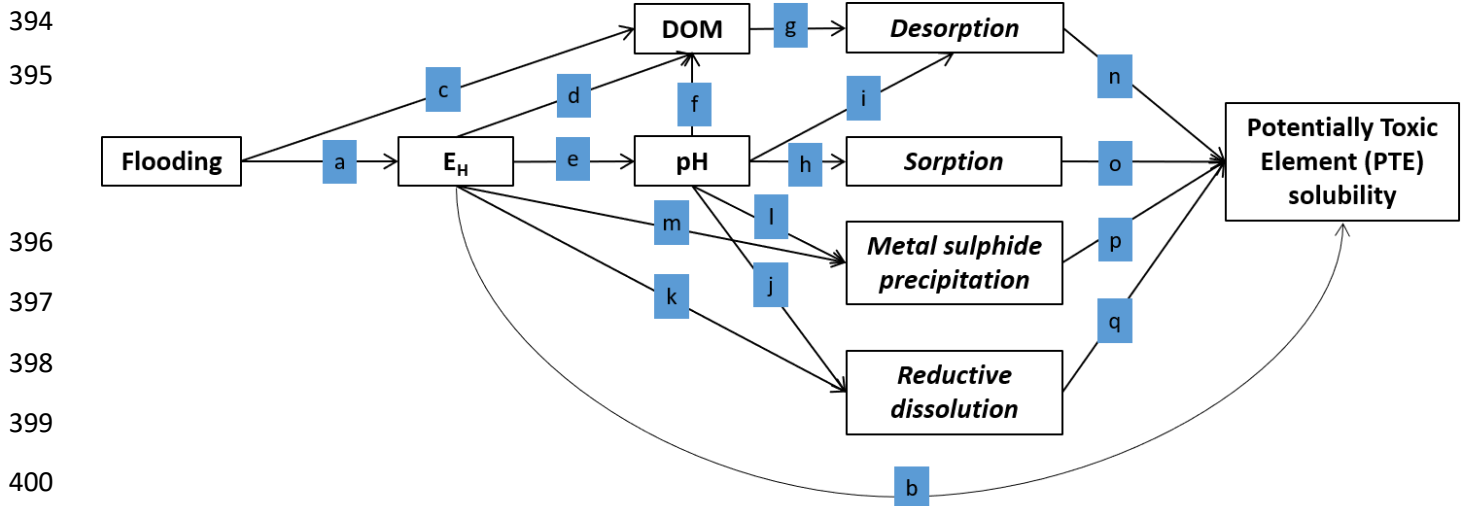
351           There are conflicting results in the literature regarding the effect of flooding on the mobility  
352 of PTEs, expressed by changes in PTEs concentration (increase or decrease), in floodplain soils (Table  
353 S1). This may largely be the result of different site-specific conditions (e.g. soil pH, texture, mineralogy)  
354 or different laboratory set-ups (e.g. submerging soils in deionised water, or the use of inert gas to  
355 simulate the anoxic conditions of a flood), illustrating the complexity of the processes involved in  
356 mediating PTEs mobility in floodplain soils (Abgottspon et al., 2015; Du Laing et al., 2007; Frohne et  
357 al., 2011; Schulz-Zunkel et al., 2015). Many of the considerations in the literature are founded on  
358 research of soils or sediments in microcosm experiments, which often involves homogenising the soil  
359 samples, resulting in loss of natural soil structure, loss of roots and biota, short-exposure time to flood  
360 conditions, and the control of variable factors such as temperature and soil water conditions (Frohne  
361 et al., 2011; Rinklebe et al., 2010). Redox conditions are often simulated and controlled through  
362 additions of O<sub>2</sub>, to increase E<sub>H</sub>, and N<sub>2</sub>, to lower E<sub>H</sub> (Frohne et al., 2014, 2011; Schulz-Zunkel et al.,  
363 2015; Shaheen et al., 2016; Shaheen and Rinklebe, 2017). These differences make extrapolation of  
364 these laboratory-based findings to field situations difficult (Hooda, 2010).

365           A key factor in determining the fate and transport of PTEs is their chemical form which, in  
366 combination with environmental factors, can influence their mobility in the soil. The chemical form of  
367 an element is often referred to as its “speciation”, “oxidation state”, or “valence” (Rodgers et al., 2015;

368 Wuana et al., 2011). There are important redox sensitive PTEs for which the oxidation state has a large  
369 influence on solubility and mobility. For example, Cr(VI) is more mobile than Cr(III), but As(V) is less  
370 mobile than As(III) (Frohne et al., 2015; Rinklebe et al., 2016; Schulz-Zunkel et al., 2015; Shaheen et  
371 al., 2014b; Yang et al., 2015). Speciation of PTEs within the environment has a distinct influence upon  
372 their behaviour; specifically, reactivity, toxicity, mobility and bioavailability within the floodplain (Du  
373 Laing et al., 2009; Gambrell, 1994; Hooda, 2010; Rodgers et al., 2015). This understanding is important  
374 for predicting the environmental impact of contaminated soils, although we are only beginning to  
375 converge on consensus on how bioavailability or speciation soil tests can help with risk-assessments,  
376 while this is slowly introduced into legislation (Cipullo et al., 2018; Naidu et al., 2015, 2008; Ng et al.,  
377 2015).

### 378 2. 3. Changes to soil physical and chemical properties that influences PTEs mobility

379 Potentially toxic elements present in soils are often adsorbed to or protected within  
380 aggregates that are stabilised by organic matter. During a flooding event, these particles may be  
381 leached through the soil profile, or suspended in flood waters where they may be redistributed across  
382 floodplain soils, or be carried downstream by the river, potentially contributing to river pollution of  
383 the contamination of downstream floodplains. The solubility and therefore mobility of PTEs from the  
384 soils to the surrounding environment depends largely on the intrinsic soil physical and chemical  
385 properties (texture, availability of soil particulate surfaces and dissolved organic matter, salinity and  
386 the presence of Fe/Mn oxides, carbonates, phosphates and sulphides) and a range of variables that  
387 are directly affected by periodic inundation of the floodplain, including; soil pH, redox potential ( $E_H$ ),  
388 dissolved organic carbon (DOC) and the valance of individual PTEs (Adewuyi and Osobamiro, 2016;  
389 Dawson et al., 2010; Du Laing et al., 2009; Frohne et al., 2015; González-Alcaraz and van Gestel, 2015;  
390 Lee et al., 2005; Puchalski, 2003; Rinklebe and Du Laing, 2011; Schulz-Zunkel and Krueger, 2009;  
391 Shaheen et al., 2016; Shaheen and Rinklebe, 2014; Steinnes, 2013). A conceptual model (Figure 2) has  
392 been produced based on our literature review (Table S1) as a way of visualising the various factors  
393 and processes influencing the solubility of PTEs in a floodplain soil.



401 **Figure 2: Conceptual model depicting the key processes influencing the solubility of Potentially Toxic**  
 402 **Elements (PTEs) after a soil becomes flooded.**  
 403 *a) Oxygen is rapidly consumed by microbial and root respiration, decreasing the redox potential ( $E_H$ ).*  
 404 *b) Decreasing  $E_H$  can lead to redox sensitive elements (e.g. As and Cr) changing valence state, directly*  
 405 *affecting solubility. c) Greater soil moisture brings dissolved organic matter (DOM) into solution. d)*  
 406 *Reducing conditions (lower  $E_H$ ) leads to the release of more DOM. e) Lower  $E_H$  results in the reduction*  
 407 *of Fe and Mn, consuming protons ( $H^+$ ) and increasing pH. f) an increase in pH often results in the release*  
 408 *of more DOM. g) DOM acts as a chelating agent, forming soluble organo-metal complexes with PTEs*  
 409 *desorbed from soil surfaces. h) as pH increases metal cations (e.g. Cu, Pb, Zn) are adsorbed on pH-*  
 410 *dependent adsorption sites of particulate matter. i) as pH increases, anions and oxy-anions (e.g. As)*  
 411 *are desorbed from pH-dependent adsorption sites. j) Dissolution of reducible Fe and Mn oxides is*  
 412 *facilitated by increasing pH. k) Microbial reduction of Mn and Fe oxides increases their solubility and*  
 413 *can cause reductive dissolution of co-precipitated PTEs. l) An increase in pH facilitates the precipitation*  
 414 *of insoluble metal sulphides. m) Microbial reduction of sulphate results in the precipitation of metal*  
 415 *sulphides. n) Release of adsorbed PTEs from soil surfaces increases PTEs solubility. o) Immobilisation*  
 416 *of PTEs through adsorption processes reduces PTEs solubility. p) Reductive dissolution of PTEs*  
 417 *associated with Fe and Mn oxides increases PTE solubility. q) Precipitation of PTEs as metal sulphides*  
 418 *decreases PTEs solubility.*

420 Soil physical, chemical and biological processes determine the mobility and redistribution of  
 421 PTEs (Hooda, 2010). These processes include; sorption, desorption, dissolution and precipitation  
 422 (Puchalski, 2003; Wijngaard et al., 2017). Subsequently, PTEs are redistributed into different  
 423 geochemical fractions, associated with other soluble species, released from the soil matrix into the  
 424 soil solution or porewater, and transferred through the ecosystem and food web to other terrestrial  
 425 or riparian areas downstream from the floodplain; thus potentially becoming a risk to human and  
 426 environmental health (Adamo et al., 2014; Adewuyi and Osobamiro, 2016; Baran and Tarnawski, 2015;

427 Dang et al., 2002; Du Laing et al., 2009; Rinklebe et al., 2016; Schulz-Zunkel et al., 2015; Shaheen et  
428 al., 2014a, 2014b; Sizmur et al., 2011). Sorption processes that control PTEs mobility and bioavailability  
429 in soil are affected by the soil pH, redox and their interactions with other ions and substances present  
430 in soil solution (Antoniadis et al., 2018; Frohne et al., 2011; Ostergren et al., 2000; Violante, 2013).

431 Sorption processes are influenced by the changing conditions that flooding brings, particularly  
432 with regards to soil moisture content, temperature and redox potential. The mobility of PTEs in  
433 flooded soils is closely related to changes in redox potential which, in turn, is altered by flooding. This  
434 can have direct impacts on the mobility of redox sensitive PTEs (e.g. As and Cr). Inundation of soils  
435 with floodwater may indirectly affect PTEs mobility and speciation because it also influences, the  
436 population, community composition, and behaviour of invertebrates inhabiting the floodplain which,  
437 in turn, influence the mobility of PTEs through their burrowing and bioturbation behaviour. For  
438 example, earthworms are known to increase the mobility of PTEs due to passage through the  
439 earthworm gut (Sizmur et al., 2011; Sizmur and Richardson, 2020) and their populations are suppressed  
440 by flooding events (Plum and Filser, 2008; Kiss et al., in review). Bioturbation/bioirrigation behaviour  
441 by chironomid larvae has been found to increase oxygen uptake at the soil/sediment-water interface,  
442 promoting POM decomposition that results in the release of dissolved organic matter and subsequent  
443 release of PTEs (He et al., 2019). Furthermore, the reduction of Mn and Fe can cause reductive  
444 dissolution of co-precipitated PTEs, and an increase in pH facilitates the precipitation of PTEs as  
445 insoluble sulphides. The mobility of PTEs can therefore increase or decrease due to the net effect of  
446 these processes (Figure 2). Which process dominates will depend primarily on the mineralogy of the  
447 soil.

448 The following sub-sections will explain how key soil physical and chemical properties are  
449 affected by flooding and how this influences PTEs mobility, followed by a discussion on the role of soil  
450 organisms and plants in mediating PTEs mobility in floodplain soils. Attention will be given to how each  
451 of these factors influence each other to distinguish direct and indirect impacts on PTEs mobility.

452 *2.3.1 Soil texture and related properties*

453 Soil texture is a stable property that refers to the physical composition of mineral fragments;  
454 sand, silt and clay and varies due to differences in underlying or upstream geology. The texture and  
455 related clay mineralogy reflect the particle/pore size distribution and overall soil surface area  
456 (Amacher et al., 1986) which, in turn, affects the soils' water holding capacity (WHC); the maximum  
457 quantity of water a soil can potentially contain, also known as the field capacity (Stürck et al., 2014).  
458 Therefore, soil physical properties play a role in flood duration because they determine the soils'  
459 ability to receive (via infiltration) and drain water during a rainfall event (Rinklebe et al., 2007). Clayey  
460 soils are likely to be saturated for longer than freely draining sandy soils (Sherene, 2010). Soil hydraulic  
461 (water retention and hydraulic conductivity curve) as well as thermal properties (thermal conductivity  
462 and heat capacity) affect the hydrothermal regime of the soil. Together these properties determine  
463 the ease in which water, and dissolved PTEs, moves through the soil pore continuum, how much water  
464 can be stored in the pore volume, and how soil temperature varies with depth. These properties are  
465 strongly dependent on soil texture, pore size distribution and mineralogy (Hillel, 1998; Tack et al.,  
466 2006; Thomas et al., 2016). Soil temperature affects the flow of water through the soil due to changes  
467 in viscosity and hence affects infiltration calculations (Gao and Shao, 2015; Prunty and Bell, 2005), so  
468 this is often corrected for when reporting hydraulic conductivity data (Thomas et al., 2016).

469 PTEs must be in the soluble phase or associated with colloids to be transported through the  
470 soil. The soil properties will play a part in the movement of PTEs into and out of the soil solution. Clay  
471 minerals and organic matter compounds have a large number of binding sites, so act as adsorption  
472 surfaces for PTEs in soils. The type of clay mineral present (kaolinite, illite, montmorillonite etc.) will  
473 also affect the specific surface area (Meegoda and Martin, 2019; Tack et al., 2006). As a result, soils  
474 with high clay and silt (fine fractions) tend to retain higher amounts of PTEs, compared to course  
475 textured sandy soils (Sherene, 2010; Zhao et al., 1999).

476 2.3.2 *Organic matter*

477 Soil POM, along with the surfaces of clay particles and Fe and Al oxides, acts as a binding phase  
478 for PTEs due to the attraction of positively charged cations to negatively charged surfaces (Evans,  
479 1989). Thus, dissolved organic matter raises the cation exchange capacity (CEC) of a soil, and is thus  
480 considered to be an important factor controlling PTEs distribution and mobility in floodplain soils and  
481 sediments (Baran and Tarnawski, 2015; Bufflap and Allen, 1995; Du Laing et al., 2009; Ehlers and  
482 Loibner, 2006). The mechanisms that bind the PTEs with particulate and dissolved organic matter  
483 include adsorption, complexation and chelation (Alvim Ferraz and Lourenço, 2000; He et al., 2019;  
484 Selinus et al., 2005). Floodplains are subject to changing water table levels and occasional inundation  
485 that brings about associated changes in redox conditions. This can result microbially mediated soil  
486 POM degradation, either during prolonged periods of flooding or in the subsequent oxidising  
487 conditions when the flood recedes, which releases organically bound PTEs, such as As, Cu, Co, Cr, Ni,  
488 Pb, and Zn from the soil into the soil solution (Adewuyi and Osobamiro, 2016; Alvim Ferraz and  
489 Lourenço, 2000; Dang et al., 2002; Kalbitz and Wennrich, 1998; Koretsky et al., 2007; Rinklebe and Du  
490 Laing, 2011). Therefore, the extent to which flooding of soils results in the mobilisation of PTEs into  
491 solution is mediated by the proportion of the PTEs that are associated with soil POM, and the  
492 susceptibility of this organic matter to degradation (as a result of microbial activity (Fe(III) and Mn(IV)-  
493 reducing micro-organisms) under reducing conditions. The free ions that are then in solution are highly  
494 reactive with the solid phase and are thought to be a major determinant of bioavailability and causing  
495 the most significant biological effects (Bufflap and Allen, 1995; Dang et al., 2002; Dawson et al., 2010;  
496 Degryse et al., 2009; Lloyd, 2003).

497 2.3.3 *Salinity*

498 Salinity is proportional to the conductivity of a sample solution; which is a measure of its ability  
499 to conduct or carry electric current and depends on the presence of charged ion species (anions and  
500 cations) (Ander et al., 2016; de Souza Machado et al., 2018; De Vivo et al., 2008)). Increasing salinity  
501 in flood water is associated with an increase in major cations that compete with PTEs for sorption

502 sites. This competition promotes PTEs desorption from the floodplain soil in the absence of sulphides  
503 and hence increases total PTEs concentrations in the soil porewater (Rinklebe and Du Laing, 2011).  
504 The presence of Ca-salts releases more PTEs into the soil solution compared with Na-salts that are less  
505 competitive for sorption (Du Laing et al., 2009; Hahne and Kroontje, 1973).

506 Changes in salinity may affect the soil physical properties and result in a destabilisation of the  
507 soil structure (Gregory et al., 2015). The salinity of the water causes a neutralisation of negatively  
508 charged clay particles, followed by flocculation (particles attaching together) which increases the  
509 deposition of sediments (along with the PTEs adsorbed to them) onto the floodplain. This process  
510 results in the floodplain becoming a sink for PTEs (Rinklebe and Du Laing, 2011). An extended flood  
511 duration, particularly when accompanied by low flow-rates (including stagnant water), results in  
512 sedimentation of fine grain sediment and organic matter that may have PTEs bound (Ciszewski and  
513 Grygar, 2016; Du Laing et al., 2009; Shaheen and Rinklebe, 2014).

#### 514 2.3.4 Redox potential ( $E_H$ )

515 Waterlogging of soils generally results in a reduction in oxygen availability due to rapid  
516 consumption of oxygen by soil microbial activity and root respiration (Du Laing et al., 2007; Rinklebe  
517 and Du Laing, 2011) and because the dissolution of oxygen through water is many times slower than  
518 through air (Alloway, 1995; Du Laing et al., 2009; Frohne et al., 2015; Schulz-Zunkel et al., 2015). The  
519 soil microbial community (e.g. bacterial species such as *Thiobacillus ferrooxidans*, *Thiobacillus*  
520 *thiooxidans* and *Leptospirillum ferrooxidans*) then uses alternative electron acceptors (such as nitrate,  
521 sulphate and Fe/Mn oxides), in anaerobic respiration, which results in a decrease in redox potential  
522 ( $E_H$ ) (Maluckov, 2017) as the floodplain soils change from oxic ( $[O_2] > 30 \mu\text{mol L}^{-1}$ ) to anoxic  
523 ( $[O_2] < 14 \mu\text{mol L}^{-1}$ ) conditions (Bellanger et al., 2004). Associated alkalinity generation drives increases  
524 in soil pH, a change which can be observed after a few days (Du Laing et al., 2007; Johnston et al.,  
525 2014; Karimian et al., 2017). Soil temperature has been found to dictate the rate and type of redox  
526 reactions; with soils at low temperatures (1 - 4 °C) requiring greater durations of saturation (20 days)



527 before the onset of reducing conditions were seen, whereas soils at higher temperatures (above 9 °C)  
528 only required 2 days of saturation (Vaughan et al., 2009).

529 Redox potential has important effects on the speciation of As, Cu and Cr, as well as N, S, Fe,  
530 Mn, because these elements can exist in soils in more than one oxidation state (Selinus et al., 2005)  
531 and solubility depends on oxidation state. Copper solubility decreases after reduction from Cu (II) to  
532 Cu (I) under anaerobic conditions and the presence of electron donors (Fe (II)) and bacteria. However,  
533 other PTEs such as Cd and Zn change valence state as a consequence of redox dependent pH changes,  
534 complexation with organic matter or precipitation with Fe and Mn (hydr)oxides or sulphides (Du Laing  
535 et al., 2009; Frohne et al., 2011).) Shaheen et al. (2014a) demonstrated that sufficient time is needed  
536 for transformations between valence states to take place. For example, the oxidation of Cr from Cr  
537 (III) to the highly mobile Cr (IV) form was found to be a slow process. This means that with shorter  
538 flooding duration and quicker cycling between oxic and anoxic conditions, Cr mobility may be difficult  
539 to predict.

540 The presence of variable charge minerals, such as Fe and Mn oxides, phosphates, carbonates  
541 and sulphides provide a reaction surface for sorption processes, allowing PTEs to bind and become  
542 immobilised (Antoniadis et al., 2018; De Jonge et al., 2012; Sipos et al., 2014; Violante, 2013). Reducing  
543 conditions change the oxidation state of Fe and Mn, increase their solubility and may have indirect  
544 effects (known as reductive dissolution) on the mobility of associated metal cations (e.g. As, Cd, Cu,  
545 Ni, Pb, and Zn), releasing them from the solid phase to pore waters, depending on flood duration  
546 (Abgottspon et al., 2015; Ciszewski and Grygar, 2016; Du Laing et al., 2009; Frohne et al., 2011;  
547 Karimian et al., 2017; Rinklebe and Du Laing, 2011; Schulz-Zunkel et al., 2015; Shaheen et al., 2016,  
548 2014b; Vaughan et al., 2009). Redox processes are a key factor for the reductive dissolution of Mn and  
549 Fe (hydr)oxides, these processes are often catalysed by microorganisms and result in the release of  
550 PTEs from the sediment (Du Laing et al., 2009; Frohne et al., 2011; Stafford et al., 2018; Yang et al.,  
551 2015). Relatively insoluble Fe(III) and Mn(IV) prevail under aerobic soil conditions providing sorption

552 surfaces for many metals, whereas under anaerobic conditions Mn(IV) and Fe(III) are reduced to more  
553 soluble forms (Mn(II) and Fe(II)) with consequential dissolution of Mn and Fe hydrous oxides, co-  
554 sorbed PTEs ions (e.g. As, Cd, Cr, Ni and Pb), are released into soil solution (Simmler et al., 2017;  
555 Stafford et al., 2018; Yang et al., 2015). After inundation, Fe and Mn may re-precipitate as oxides and  
556 can bind (by desorption or co-precipitation) the trace metals back into the solid state (Ciszewski and  
557 Grygar, 2016; Davranche et al., 2011; Du Laing et al., 2009).

558           Decreasing of  $E_H$  can initiate microbial sulphate reduction and this can reduce the mobility of  
559 some PTEs (e.g. As, Cd, Cu, Cr, Ni and Pb) through coprecipitation of metal cations with sulphides  
560 (Abgottspon et al., 2015; Borch et al., 2010; Weber et al., 2009), although many of these minerals are  
561 metastable and so prone to change (Karimian et al., 2018). Yang et al., (2015) put mixed sediment  
562 samples into a laboratory culture tanks and found that microbially induced release of sulphur with  
563 subsequent As precipitation was more important for controlling As adsorption/desorption than  
564 reductive dissolution of Fe/Mn oxides. As the flood recedes, the floodplain soils undergo drying and  
565 aeration that change the conditions from anoxic back to oxic. The now oxic environment causes  
566 sulphides to be oxidised, which then releases PTEs back into the pore waters (Abgottspon et al., 2015;  
567 Du Laing et al., 2007; Frohne et al., 2011). In addition to this, when exposed to oxygen and water,  
568 sulphides are oxidised to sulphates which leads to the formation of sulphuric acid thereby causing a  
569 decrease in pH and release of the PTEs (Emerson et al., 2017; Forstner and Wattman, 1981). Frohne  
570 et al. (2011) suggested that the mobility of Cd, Cu, Mn, Ni and Zn under oxidising conditions could be  
571 attributed to dissolution of sulphides and the resulting release of those metals. The extent to which  
572 the mineralogy of a floodplain soil is dominated by Fe/Mn oxides or sulphates may dictate whether  
573 PTEs are mobilised or immobilised during inundation, and the extent to which this phenomenon is  
574 reversed after floodwater recedes.

575 2.3.5 Soil pH

576 pH is a measure of the hydrogen ion concentration and can also be referred to as the degree  
577 of acidity or alkalinity. The soil pH is affected by flooding because of a well-established correlation  
578 between soil pH and changing redox conditions; as a soil becomes flooded, this creates reducing  
579 conditions where ( $H^+$  ions) are consumed (for example due to reduction of Fe and Mn oxides) and the  
580 pH increases (Rinklebe and Shaheen, 2017; Weber et al., 2009). When the flood recedes, oxidation  
581 processes produce protons and decrease the pH (Adewuyi and Osobamiro, 2016; Frohne et al., 2015,  
582 2011; Rinklebe and Shaheen, 2017; Shaheen and Rinklebe, 2017). Furthermore, on exposure to the  
583 atmosphere, when flooding recedes, dissolved organic carbon (DOC) is converted to  $CO_2$ , which  
584 dissolves into porewater as carbonic acid, subsequently further reducing the soil pH (Peacock et al.,  
585 2015). However, this negative correlation between  $E_H$  and pH hasn't always been observed (Du Laing  
586 et al., 2009; Frohne et al., 2015). This is because the degradation of POM such as plant residues, by  
587 soil microbes, may increase the soil pH due to ammonification of the residue N (Xu et al., 2006).

588 As the pH changes, processes such as precipitation, co-precipitation and sorption/desorption  
589 of PTEs from organic matter or clay minerals occur, altering the chemical composition as well as  
590 reaction rates (Frohne et al., 2011). The soil pH plays an important role in mediating the mobility of  
591 PTEs and their availability for plant uptake, as the protons compete with metal cations for exchange  
592 sites on the surface of soils. Some of these exchange sites, particularly those associated with soil  
593 organic matter, are pH-dependent and thus only become deprotonated at high pH. A decrease in pH  
594 is generally accompanied by an increase in the mobility of most PTEs that are metal cations (e.g.  $Cd^{2+}$ ,  
595  $Cu^{2+}$ ,  $Co^{2+}$ ,  $Ni^{2+}$ ,  $Pb^{2+}$  and  $Zn^{2+}$ ) (Gröngröft et al., 2005; Sherene, 2010). Thus, as pH increases there is a  
596 subsequent decrease in the mobility of these PTEs (Giacalone et al., 2005). The extent to which PTEs  
597 mobility decreases in soils during flooding, due to a redox-induced increase in pH, is likely to depend  
598 on the proportion of PTEs in the soil that are associated with pH-dependent exchange sites) which are  
599 typically associated with soil organic matter) and the pH of the soil prior to the flooding event.

600 2.3.6 *Dissolved organic matter (DOM)*

601 The increase in pH of soil solutions with lower redox potential (reducing conditions) is often  
602 accompanied by a release of dissolved organic matter (DOM) and the subsequent formation of soluble  
603 organo-metal complexes (Abgottspon et al., 2015; Alvim Ferraz and Lourenço, 2000; Frohne et al.,  
604 2011). The presence of DOM in floodplain soils acts as a chelating agent which has a strong binding  
605 ability and increases the mobility of PTEs into pore waters and subsequently into river water or  
606 groundwater (Dawson et al., 2010; Du Laing et al., 2009; Shaheen et al., 2014b). The greater the  
607 concentration of DOM in porewater, the more PTEs that are held in solution, and (to maintain an  
608 equilibrium) the more PTEs that desorb from the surfaces of the soil to replenish the free ion  
609 concentrations in the porewater, thus increasing PTEs mobility. Greater concentrations of DOM have  
610 been observed with decreasing  $E_H$ , which may be due to suppressed microbial carbon consumption  
611 under anoxic conditions (Frohne et al., 2015). Shaheen et al. (2014a) highlighted that increases in DOM  
612 associated with lower  $E_H$  may help to catalyse changes in the valence state of PTEs; for example, of  
613 Chromium (III) to (VI).

614 2.3.7 *Temperature*

615 As temperatures are predicted to increase as a result of climate change, they may become a  
616 factor that contributes to greater release of PTEs from the soil during a flood (Visser et al., 2012). Soils  
617 are affected by variations in air temperature which, in turn, affects the rate of biogeochemical  
618 processes during a flooding event, including decreasing redox potential and, ultimately, influences the  
619 rate and extent to which PTEs are released/ desorbed from POM into surface water and groundwater  
620 (Arnell et al., 2015; González-Alcaraz and van Gestel, 2015; Sánchez-Rodríguez et al., 2019; Shaheen  
621 et al., 2016; Stahl et al., 2013). Increases in temperature raise the ion activity in soil solution, and also  
622 make plants more active, which may lead to greater plant root uptake of soil water and  
623 dissolved/labile PTEs within this water (Sherene, 2010). Arsenic release from flooded soils was found  
624 to have temperature dependence, with As solubilisation increasing as temperature increased  
625 (Simmler et al., 2017; Visser et al., 2012). Temperature increases are attributed to a decrease in the

626 water viscosity resulting in dissociation of molecules and a subsequent increase in the number of ions  
627 in the solution. For every degree Celsius increase in temperature there is an observed increase in  
628 electrical conductivity of 1.9% (Ander et al., 2016; Ma et al., 2011).

629 Many of the processes (e.g. redox reactions) described in the previous sections are microbially  
630 mediated and temperature dependent, and so the extent to which they affect the mobility of PTEs  
631 depends on their kinetics and the duration that floodplain soils are inundated. Changes in soil pH and  
632 DOM have been shown to exert a greater influence than  $E_H$  on the mobility of PTEs when considering  
633 shorter flood-dry cycles (Shaheen et al., 2014b, 2014a). However, Dang, Liu, and Haigh (2002) found  
634 that with increasing flood duration, more trace elements were transformed from inert phase to  
635 exchangeable fractions, increasing mobilisation. Soil redox processes are important for protecting  
636 environmental health; however, the kinetics and mechanisms remain poorly characterised and  
637 understood (Abgottspon et al., 2015; Borch et al., 2010; Pulchalski, 2003). Many of the studies  
638 reviewed in this manuscript undertook experiments in the laboratory at temperatures that are higher  
639 than the soil temperatures typically found *in-situ* at the location where the soil samples were  
640 collected, and so the rates at which reactions occur and the subsequent mobilisation of PTEs may be  
641 overestimated in these experiments. It is not possible to verify the extent of the overestimation, so  
642 more *in-situ* experiments are needed to investigate and provide quantification of the differences  
643 between laboratory and *in-situ* experiments.

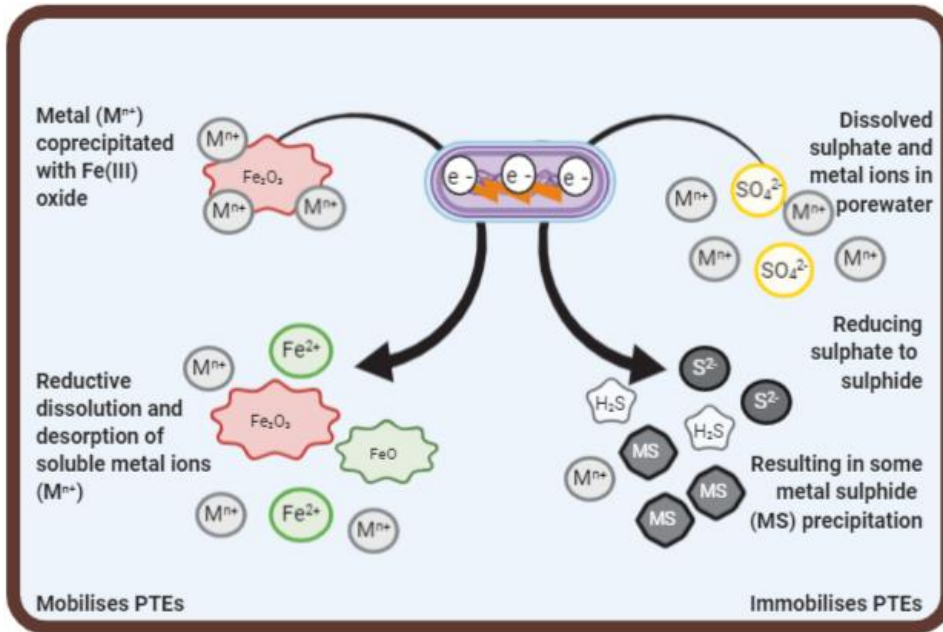
644 Groundwater and river temperatures may have a strong effect on floodplain soil temperatures  
645 (Andersen, 2018). In warmer seasons they will generally be cooler than soil temperatures, but the  
646 opposite occurs in cooler seasons. Also, changes in soil moisture content, as a result of flood events,  
647 will affect the soil thermal properties such as thermal conductivity and heat capacity (Lu et al., 2007),  
648 thereby also affecting the spatial and temporal variation in the soils' temperature regime. A laboratory  
649 microcosm experiment with mining-contaminated topsoil and subsoil samples saturated for up to 41  
650 days at temperatures ranging between 10-25°C, found that soil temperature increased the

651 solubilisation of As, particularly in the topsoil when saturated for 1-2 weeks (Simmler et al., 2017).  
652 This means, for some PTEs, flooding during warmer seasons may result in greater mobilisation than  
653 flooding during cooler seasons. More seasonal field observations are required to understand the  
654 subtle interactions and feedbacks between soil moisture, floodwater temperature, and mobility of  
655 PTEs.

## 656 2. 4. Soil biological processes that influence PTEs mobility

### 657 *2.4.1 Soil organisms*

658 Floodplain soils contain a great diversity of organisms that are known to contribute to the  
659 physical structure of the soil/sediment through bioturbation which influences the biogeochemical  
660 cycling of PTEs through oxygen diffusion, redox gradient and decomposition of dissolved organic  
661 matter (Classen et al., 2015; He et al., 2019; Hooda, 2010; Selinus et al., 2005). As the soil pore spaces  
662 are filled with water, oxygen diffusion is low so microbial respiration relies on alternative electron  
663 acceptors (e.g.  $\text{NO}_3^-$ , Mn, Fe and S), resulting in reducing conditions (decreasing  $E_H$ ) that  
664 simultaneously increase pH (Matern and Mansfeldt, 2016), and the changes to PTEs mobility (Figure  
665 3) that are described in previous sections. Changes in the chemical speciation of PTEs can also occur  
666 due to microbial processes in reducing conditions, for example, sulphate reducing bacteria can  
667 methylate Hg in anoxic conditions (Ma et al., 2019).



668

669 *Figure 3: Soil microbial processes during inundation of floodplain soil influences mobility of PTEs; (left-*  
 670 *hand side) generic metals with valence state (denoted by  $M^{n+}$ ) are coprecipitated to Fe oxides and are*  
 671 *released due to reductive dissolution and (right-hand side) sulphate reduction (sulphate to sulphide)*  
 672 *results in some metal (denoted by MS) precipitation, some of the metal ( $M^{n+}$ ) remains in the pore water.*  
 673 *Created with BioRender.com.*

674

675 PTEs that are present in floodplain soils are often protected within the soils' aggregates, which  
 676 are stabilised by POM. However, inundation can stimulate the soil microbial community, which is  
 677 sensitive to disturbance, accelerating the refractory organic matter mineralisation and destabilisation  
 678 of aggregates, exposing and increasing the mobility of PTEs in the soil (Du Laing et al., 2009; Gall et al.,  
 679 2015; González-Macé et al., 2016; He et al., 2019; Rawlins et al., 2013). Tack et al. (2006) found that  
 680 the drying of sandy soils caused an increase in soil solution metal concentrations, compared with the  
 681 same soils maintained at field capacity. This observation was attributed to microbial effects, increasing  
 682 the solubility of dissolved organic matter.

683 Flooding has been found to shift the soil biological community structure and function. These  
 684 changes include a reduction of Gram-positive bacteria, mycorrhizal fungi and earthworms found under  
 685 flooded conditions (Gregory et al., 2015; Harvey et al., 2019; Unger et al., 2009). Harvey et al., (2019)  
 686 found that flooding induced short-term alterations to soil microbial biomass but these changes did

687 not persist in the long term; they concluded that temperate systems may be resilient to winter flood  
688 stress. The seasonal timing of floods influences the effect that flooding has on the soil microbial  
689 community, and so may result in different effects on, and recovery of, the soil microbial community.  
690 Sánchez-Rodríguez et al., (2019) subjected a UK agricultural grassland soil in an intact laboratory  
691 microcosm to flooding and found that summertime flooding (25°C), resulted in a loss of actinomycetes  
692 and arbuscular mycorrhizal fungi, and that these changes persisted post-flood. They expected  
693 microbial biomass to increase with flooding at higher temperatures, due to degradation of vegetation  
694 releasing labile carbon. However, they found that maintaining live roots and an active rhizosphere  
695 were more important for preserving the microbial community in grassland soils. Earthworms also play  
696 a role in increasing the mobility and availability of PTEs in floodplain soil through their activity causing  
697 changes to the soil microbial populations, pH, DOC or metal speciation (Sizmur et al., 2011; Sizmur  
698 and Hodson, 2009) which in turn influences PTEs mobility as discussed in the above sections.

699 As the PTEs are released into the aqueous phase and mobilised in the environment, they  
700 present a potential risk to soil organisms (Ehlers and Loibner, 2006; González-Alcaraz and van Gestel,  
701 2015). Soil organisms uptake PTEs via ingestion of polluted soil, food or pore water and/or via dermal  
702 uptake or absorption of soil water, with the soil water being the more important of the two pathways  
703 (Chrzan, 2016; Hobbelen et al., 2006; Sivakumar and Subbhuraam, 2005). Vijver et al. (2007) found  
704 that the frequency of flooding did not result in consistent changes in the internal PTEs concentrations  
705 of earthworms. Earthworms accumulate PTEs in their chloragogenous tissue and have a mechanism  
706 that allows them to regulate their internal PTEs concentrations, so when they are introduced to  
707 contaminated soils the earthworms reach an equilibrium and when they are returned to  
708 uncontaminated/"clean" soils they are able to detoxify and eliminate essential metals through  
709 excretion (e.g. Cu and Zn), but not non-essential metals (e.g. Cd and Pb) as detoxification processes  
710 involve sequestration within an inorganic matrix or organic ligand (Sizmur and Hodson, 2009;  
711 Spurgeon and Hopkin, 1999). While microbes can tolerate larger quantities of essential PTEs, in excess  
712 both essential and non-essential PTEs (e.g., Al, As, Cd, Hg, Pb, Zn) can adversely affect microbial



713 communities by altering community structure and taxonomic richness; reducing the microbial  
714 biomass and lowering their enzyme activity which results in a decrease of soil diversity (Gadd, 2010;  
715 Gall et al., 2015; Wuana et al., 2011).

716

#### 717 2.4.2 Plants

718 In many cases, PTEs are concentrated in the upper part of the soil profile where roots reside,  
719 meaning that increased mobility is likely to affect plants growing in floodplain soils. Wetland plants  
720 growing on inundated floodplain soils can also affect the mobility of PTEs because they are specially  
721 adapted to have air-filled tissues, or aerenchyma, which create patches of oxygenated soil around  
722 their roots, resulting in an increase in the volume of the oxic/anoxic interface and remobilising PTEs  
723 thus increasing their availability (Du Laing et al., 2009; Wright et al., 2017). However, in arable and  
724 pasture fields that are generally drier, flooding can cause crops to become stressed, as they are not  
725 adapted to wet soils. As oxygen levels decrease there is a build-up of carbon dioxide, methane and  
726 nitrogen gases that leads to the roots suffocating and dying (Hippolyte et al., 2012).

727 It is well established that symbiotic fungi, associated with plant roots, regulate the supply of  
728 micronutrients and reduce the uptake of non-essential PTEs by plants (Classen et al., 2015; Gadd,  
729 2010; Tack, 2010). Plants, such as *Artemisia* and *Phalaris* species, on the floodplain excrete exudates  
730 during inundation which stimulates the activity of microbial symbionts in the rhizosphere, allowing  
731 PTEs to be taken up into the vegetation (Gall et al., 2015; Sullivan and Gadd, 2019; Violante et al.,  
732 2010; Xu et al., 2020). PTEs are often accumulated in plant root tissues and can sometimes be  
733 translocated into the plant shoots. However this is regulated in plants by the Casparian strip and  
734 therefore limited (Hooda, 2010; Nouri et al., 2009; Shahid et al., 2017). The uptake and accumulation  
735 of PTEs is element and plant-specific (Niu et al., 2007; Rinklebe et al., 2016; Tack, 2010; Violante et al.,  
736 2010; Xu et al., 2020). The mobilisation and uptake of PTEs by plants may pose a potential  
737 environmental risk (Shaheen and Rinklebe, 2014). European floodplains are most commonly used as  
738 grassland for grazing cattle or hay production, whereas in other regions e.g. India, they are used for

739 crops like rice, which raises concerns for possible pollutant transfer from the floodplain soil into the  
740 surrounding water bodies, then uptake and potential biomagnification of PTEs into the food chain  
741 (Martin et al., 2014; Overesch et al., 2007; Tóth et al., 2016a). However, the hyperaccumulation of  
742 PTEs by some plants (e.g. sunflower, mustard (Brassicaceae), alfalfa and Ricinus) has resulted in them  
743 being considered for phytoremediation of contaminated floodplain soils (Gall et al., 2015; Niu et al.,  
744 2007; Nouri et al., 2009; Shaheen et al., 2016; Violante et al., 2010).

745           Factors influencing plant uptake of PTEs include soil pH, electrical conductivity and the total  
746 concentrations of PTEs in the soil (Nouri et al., 2009). PTEs uptake also depends on the concentrations  
747 in the soil solution, governed by plant exudates and root-induced changes to pH and DOM (Gall et al.,  
748 2015). Quantifying the total content of PTEs transferred into the food chain via plants growing on  
749 contaminated soil is difficult (Gröngröft et al., 2005). The concentrations of PTEs found in floodplain  
750 plants are not always directly reflected in the PTEs content found in the soil, due to both physiological  
751 and biochemical differences between different plant species; for example differences in the age of the  
752 plant biomass (seasonal trends in growth and therefore uptake of nutrients). Moreover, the rooting  
753 depth influences metal mobilisation/immobilisation and element specific uptake into the roots which  
754 also affects the transfer into the shoots(Chrzan, 2016; Overesch et al., 2007). Thapa et al. (2016) also  
755 demonstrated a change in semi-arid Australian floodplain vegetation productivity in response to  
756 flooding and drying cycles; flooding brings nutrients which increases net primary productivity. These  
757 changes in vegetation productivity could also initiate structural changes in floodplain vegetation  
758 communities in natural and semi-natural ecosystems (Overesch et al., 2007).

### 759 3. Summary and further research needs

#### 760 3.1. Summary of current understanding

761           Floodplain soils downstream of urban catchments contain elevated concentrations of PTEs as  
762 a legacy of human activity and these PTEs could potentially be remobilised by future flooding events.  
763 A number of processes occur within the soil, ultimately determining PTEs fate. These processes

764 include: sorption, desorption, complexation, precipitation and dissolution, transport of water and  
765 heat, and biological activity. The processes are influenced by the changing conditions that flooding  
766 brings particularly with regards to soil moisture content, temperature and redox potential. The  
767 mobility of PTEs in flooded soils is closely related to changes in redox potential which, in turn, is altered  
768 by flooding. These changes can have direct impacts on the mobility of redox sensitive PTEs (e.g. As  
769 and Cr). Furthermore, the reduction of Mn and Fe can cause reductive dissolution of co-precipitated  
770 PTEs, but the reduction of sulphate can result in the precipitation of PTEs as insoluble metal sulphides.  
771 Which of these processes dominates will depend on the mineralogy of the soil. PTEs precipitated as  
772 metal sulphides may oxidise after floodwaters recede and mobilise, accelerated by the pH reduction  
773 caused by production of sulfuric acid. There are important interactions between redox potential and  
774 other soil properties, such as soil pH, moisture content, POM, DOM, temperature, and salinity which  
775 also have a strong impact on PTEs mobility (Vaughan et al., 2009). Many of these reactions are  
776 microbially mediated, temperature dependent and the kinetics in real-world scenarios are poorly  
777 understood. However, it seems that changes associated with alterations to pH and dissolved organic  
778 carbon are relatively fast, while changes to  $E_H$  are slower and only become apparent after extended  
779 periods of flooding. In many cases, PTEs deposited due to legacy pollution events are concentrated in  
780 the upper part of the soil profile, meaning that increased mobility is likely to affect plants growing in  
781 floodplain soils and potentially lead to contamination of the surrounding environment, including  
782 overlying surface waters.

783

### 784 3.2 Knowledge gaps and recommendations for future research

785 Floods are dynamic events that expose floodplain soils to water with rapidly changing flows,  
786 chemical composition, and sediment load. They can be difficult to predict, due to their different types  
787 (e.g. overbanking or groundwater flooding), and the high variation in their magnitude, duration, and  
788 frequency of recurrence. Therefore, chemical, physical and biological data from floodplain soils  
789 immediately before and immediately after a flooding event are often lacking. However these data

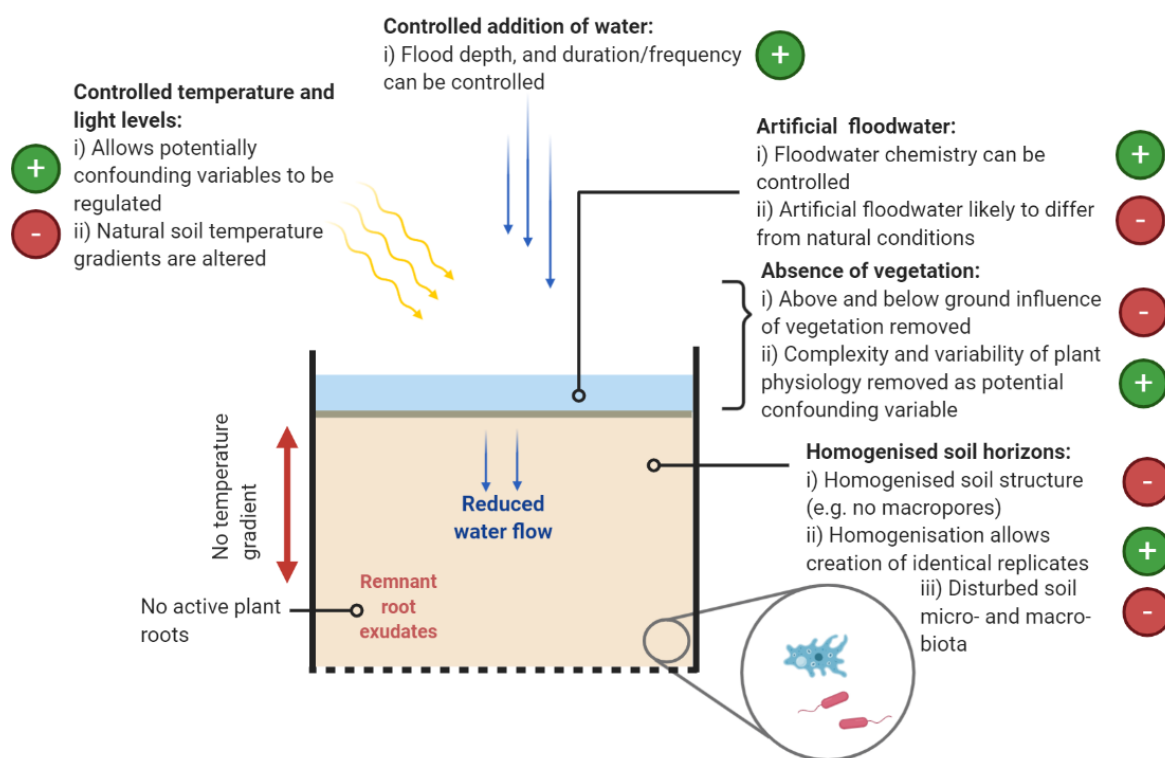
790 would provide the necessary insights into the factors and processes involved in altering the mobility  
791 of PTEs during and after a real flooding event (Barber et al., 2017). The effect of flooding on PTEs  
792 mobility can be difficult to predict due to there being several factors (e.g. speciation, release through  
793 biological degradation and competitive action of other ions) or interactions between factors (e.g.  
794 changes in  $E_H$  caused degradation of POM) influencing PTEs mobility (Tack and Verloo, 1995).

795 Contamination of soil with PTEs receives most attention in highly contaminated urban,  
796 industrial, mining and waste disposal sites (Adamo et al., 2014; Resongles et al., 2015; Simmler et al.,  
797 2017; Wuana et al., 2011) with relatively little attention given to more 'typical' floodplains  
798 downstream of catchments with a history of urban and industrial development. Much of the work  
799 conducted to date (see Table S1) has been undertaken in Europe, America, Canada, China, Indonesia,  
800 Australia and New Zealand. Just over half of the studies cited in Table S1 were undertaken in Germany  
801 and Belgium (52%), with a particular research effort around the River Elbe and Wupper River in  
802 Germany (Du Laing et al., 2009; Förstner, 2004; Frohne et al., 2011; Overesch et al., 2007; Rennert et  
803 al., 2017; Rinklebe et al., 2013; Shaheen et al., 2017). However, research examining the relationship  
804 between PTEs mobility and flooding in other parts of the world that are expected to see an increase  
805 in the frequency and magnitude of flooding events, for example in Asia, Africa and India, is limited.

806 A number of factors were identified that contribute to whether the mobility of PTEs will  
807 increase or decrease during inundation of a floodplain, which may be interconnected or work in  
808 combination to affect PTEs mobility. As a result, different soils with differing mineralogy and thus  
809 different biogeochemical and physical properties, will likely respond differently to flooding. Individual  
810 studies tend to focus on one floodplain site. However, knowledge based on one river catchment may  
811 not be particularly useful for predicting the impacts of flooding at another site with different  
812 mineralogy and physical and chemical characteristics. A more fundamental mechanistic understanding  
813 is required to inform the development of predictive models. Therefore, more coordinated work  
814 encompassing multiple contrasting sites is required to understand the relative importance of key soil

815 properties (e.g. mineralogy, POM, soil pH, texture; and how these affect derived soil properties such  
816 as hydraulic and thermal soil properties) on influencing the impact of flooding on the mobility of PTEs.

817           Many of the findings in the literature are based on research of soils or sediments in laboratory-  
818 based artificial flooding environments (Figure 4), which often involve; homogenisation of samples and  
819 removal of plant roots, short-exposure time for soil microorganisms and incubation under controlled  
820 conditions, such as temperature (often higher than *in-situ* temperatures) and soil water conditions  
821 (often wetting the samples with deionised water which is slightly acidic) (Frohne et al., 2011; Izquierdo  
822 et al., 2017; Rinklebe et al., 2010; Weber et al., 2009). This makes extrapolation of laboratory-based  
823 findings to field situations difficult (Hooda, 2010). Attempts to model the concentration of PTEs in  
824 floodplain pore waters have demonstrated the complexity of predicting how different variables such  
825 as soil moisture content and temperature interact and alter mobility (Rennert et al., 2017), with site  
826 or catchment-specific information being of great importance to establish and capture spatial  
827 differences sufficiently (Schulz-Zunkel et al., 2015). While much research undertaken in controlled  
828 conditions in laboratory microcosms is undoubtedly useful because independent replicates can be  
829 assigned to treatments without confounding variables (Figure 4), there is a clear research need for on-  
830 site experiments on the effect of flooding on PTEs mobility using real-time field-based observations  
831 that capture the kinetics of processes before, during, and after a flooding event under ambient  
832 temperatures and in geochemically contrasting soils.



833

834 *Figure 4: Strengths (+) and weaknesses (-) of laboratory-based studies for researching the impact of*  
 835 *flooding on mobility of PTEs. Created with BioRender.com.*

836

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840

841 Supplementary Material

842 One supplementary table (Table S1) is provided

843

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