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

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Abstract

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

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

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

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

1. Introduction

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

Soil contamination is among the most serious threats to soil resources globally (Nriagu et al., 2007; Srivastava et al., 2017; Tóth et al., 2016b). Since many commercial, industrial, residential and agricultural developments have historically been situated adjacent to rivers; they contribute to the contamination of river sediments, and these sediments are often deposited onto the floodplain soils downstream by overbanking river water during a flooding event (Arnell et al., 2015; Nshimiyimana et al., 2014; Zhao et al., 1999). Here we use the term PTEs, also referred to in the literature as ‘trace elements’ or ‘heavy metals’, to encompass all metals, metalloids, non-metals and other inorganic elements in the soil–plant–animal system, of which their mobility and potential toxicity to that system and/or humans is largely dependent upon their concentration, bioavailability and chemical form (Hooda, 2010; Rodgers et al., 2015). The term "mobility” is a concept that has been frequently used to estimate the risk of contamination from the soil to the surrounding environment by PTEs (Domergue and Vedy, 1992). Here we define mobile PTEs as those elements that are dissolved in soil porewater or associated with colloids and thus capable of leaching from the soil profile, or being taken up into plants or soil organisms. The mobility and subsequent fate of PTEs in periodically (occasionally) flooded soils (such as floodplain soils) are imperfectly understood. The legacy of historic contamination and continuing increases in emissions from urban activities pose a serious environmental threat globally (de Souza Machado et al., 2016; Srivastava et al., 2017). Human actions to mitigate and adapt to the impacts of climate change may influence the fate of contaminants, with climate change itself also potentially affecting the toxicity of the contaminants within the environment (Stahl et al., 2013).

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

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

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

An IPCC Special Report (SREX) on climate extremes (IPCC, 2012) assessed it is *likely* there have been statistically significant increases in the number of heavy precipitation events in more regions than significant decreases, with strong regional and sub-regional variation. The observed changes to precipitation extremes have been found to be far less spatially coherent or statistically significant compared with changes found in temperature extremes (Kundzewicz et al., 2014). Projected scenarios with 4°C warming showed more than 70% of the global population will face increased flood risk (Alfieri et al., 2017). Increases in flood frequency are expected in; Europe, America, Southeast Asia, eastern Africa, and Peninsular India. Populations in regions such as Bangladesh, Mumbai and Thailand are potentially at higher risk from flooding due to predicted increases in rainfall, coupled with changes in land use (e.g. irrigation schemes and construction of dams), and increasing population size requiring rapid expansion of urban areas (Hirabayashi et al., 2013; Kundzewicz et al., 2014; Tockner et al., 2010). However, climate projections tend to have relatively low levels of model confidence, particularly for the prediction of fluvial floods because there is still relatively limited evidence and the causes of regional changes to flood occurrence are complex (Hirabayashi et al., 2013).

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

The probability of flooding occurring in a particular region is often related to regional processes like El Niño Southern Oscillation (ENSO) cycle and the North Atlantic Oscillation (NAO) that, in turn, cause global impacts. The intensity (frequency and amplitude) of both ENSO and NAO are influenced by other modes of variability, for example; Pacific Decadal Oscillation (PDO) and Interdecadal Pacific Oscillation (IPO) which cause opposite atmospheric and sea surface temperatures and can therefore determine the magnitude of floods (Grimm and Tedeschi, 2009; Johnson et al., 2020). ENSO is a rapid warming of the sea surface temperature (by 1–5 °C) of the equatorial Pacific over the duration of a few weeks, resulting in extreme rainfall and increased cyclone activity in some regions, and risk of drought and forest fires in others (Berz et al., 2001; Grimm and Tedeschi, 2009; Karl and Trenberth, 2003; Kundzewicz et al., 2014; Tedeschi and Collins, 2016). Periods of extreme rainfall and subsequent flooding have been found to correlate with ENSO events in North and South America as well as in Africa (Berz et al., 2001; Brönnimann, 2007; Kundzewicz et al., 2014). NAO is an atmospheric pattern that affects the severity of winter temperatures and precipitation over Europe and eastern North America (Karl and Trenberth, 2003). Intense rainfall is a common cause of river basin flooding; however, in high latitude regions it is changes in temperatures altering the timing of seasonal snowmelt and causing glacier retreat that commonly causes flooding, for example in north-eastern Europe, Central and South America, and in polar regions such as the Russian Arctic (Blöschl et al., 2017; Hirabayashi et al., 2008; Kharin et al., 2007; Kundzewicz et al., 2014; Stagl et al., 2014). Rising global sea-level (11-16cm in the 20th century and a further 0.5m predicted this century) will certainly increase risk of flooding caused by tidal processes, with current estimates that 630 million people live on land below projected annual flood levels for 2100 (Kulp and Strauss, 2019). While there is uncertainty regarding the effect that future climate change will have on river levels (Prudhomme and Davies, 2009), changes made to land-use, and land cover, for example by urbanisation, will drive changes in the local climate (at the kilometre scale) influencing the hydrometeorological regime and resulting in more flooding (Foley et al., 2005; Hirabayashi and Kanae, 2009). Pathirana et al. (2014), using a 3D atmospheric model coupled with a land surface model (WRF-ARW) in southern India, found that in three out of four simulated cases there was a significant increase in local extreme rainfall when urbanisation in the area increased. This work was conducted in southern India, however the model could be applied and validated to other regions to establish whether this correlation is found globally.

1.2. The role of floodplains during floods

Floodplains are by definition dynamic environments subjected to fluctuations between flooding and drying (Vijver et al., 2007). They are distinctive landscape features, often on low-lying ground, and characterised by a high spatio-temporal heterogeneity (Schulz-Zunkel et al., 2015; Stuart and Lapworth, 2011; Tockner et al., 2010; Tockner and Stanford, 2002). Periodic overbank inundation from the adjacent watercourse, overland flow, subsurface flow, and changes to the groundwater levels result in a constantly changing water balance and degree of floodplain saturation (Stuart and Lapworth, 2011; Tockner and Stanford, 2002). Floodplain topography and variations in elevation are usually slight but have an important effect on the degree of soil saturation across the floodplain, depending on the overall water balance from surface and sub-surface run-off (Arnell et al., 2015; Kundzewicz et al., 2014; Qiao et al., 2019).

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

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

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

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

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

1.3. Changes that floodplain soils undergo during and after inundation

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

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

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

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

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

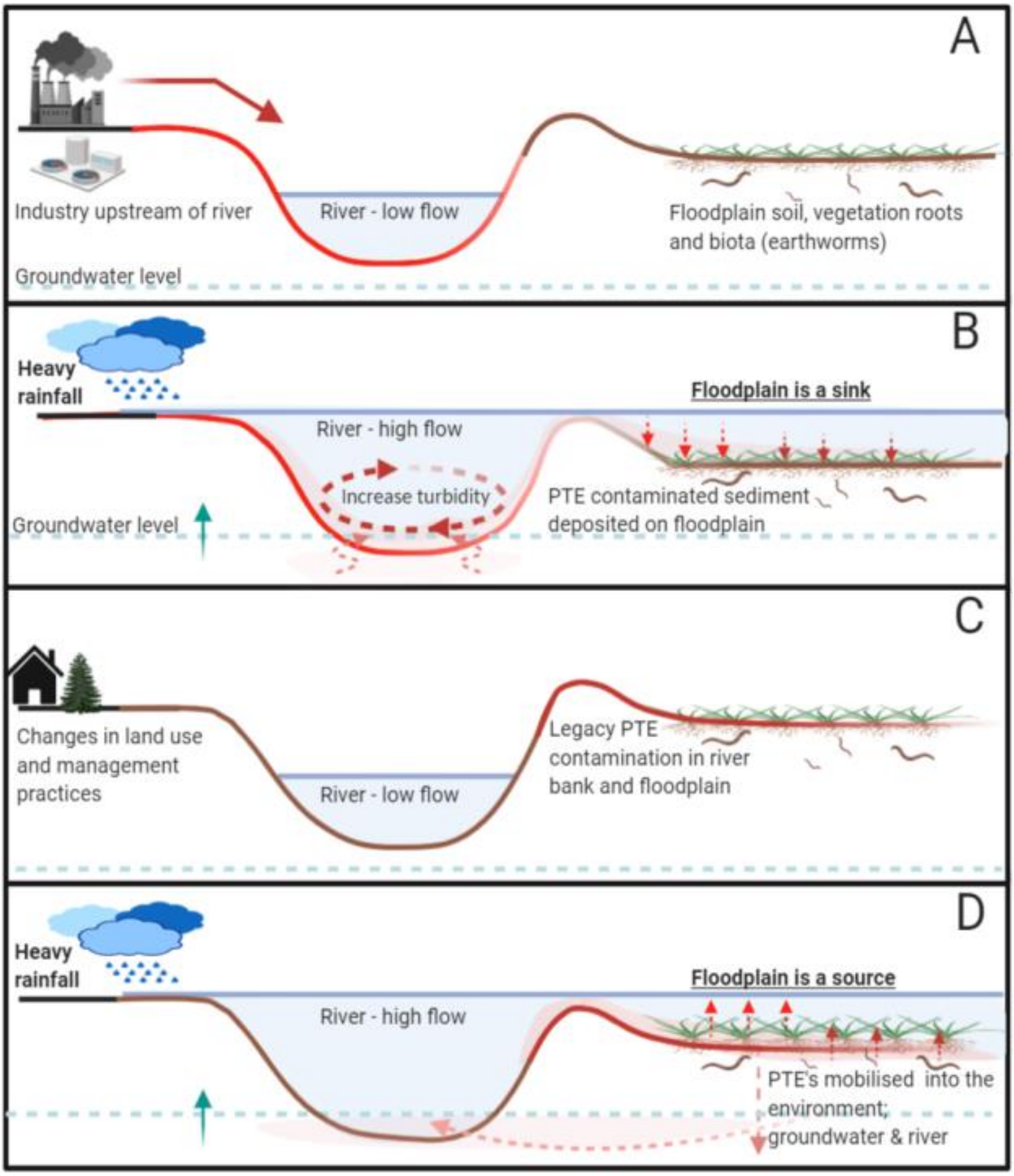

Figure 1: How floodplains may switch from being a sink of pollution to becoming a source of legacy pollution: *A) PTEs contaminated river sediment (red) due to industry in the catchment upstream, B) heavy rainfall influences the receiving catchment (increased river flow and groundwater level), resulting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

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

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

2.1 PTEs in floodplain soil

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

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

2. 2. Influence of flooding on PTEs mobility

During a flooding event, biogeochemical processes occur in the floodplain soil at the oxic-anoxic interface and in the anoxic layers. The kinetics of these processes are of great importance because the location of the oxic-anoxic interface is subject to change due to fluctuating water table levels (Du Laing et al., 2009; Puchalski, 2003). In their review of trace metal behaviour in floodplain sediments, Du Laing et al. (2009) state that the spatial occurrence of processes affecting metal mobility and availability is largely determined by the topography of the floodplain. Remobilisation of PTEs from sediments into the overlying water column during a flooding event depends on the flood regime; the frequency of these intense floods which flush or remobilise contaminated material as well as the duration or alternation of flood with dry spells (Arnell et al., 2015; Foulds et al., 2014; González-Alcaraz and van Gestel, 2015). Whilst research has suggested that the longer the flood duration, the greater the metal mobility (Shaheen et al., 2014a, 2014b), Stafford et al. (2018) suggest that even short periods of soil saturation can have an influence the solubility of PTEs.

There are conflicting results in the literature regarding the effect of flooding on the mobility of PTEs, expressed by changes in PTEs concentration (increase or decrease), in floodplain soils (Table S1). This may largely be the result of different site-specific conditions (e.g. soil pH, texture, mineralogy) or different laboratory set-ups (e.g. submerging soils in deionised water, or the use of inert gas to simulate the anoxic conditions of a flood), illustrating the complexity of the processes involved in mediating PTEs mobility in floodplain soils (Abgottspon et al., 2015; Du Laing et al., 2007; Frohne et al., 2011; Schulz-Zunkel et al., 2015). Many of the considerations in the literature are founded on research of soils or sediments in microcosm experiments, which often involves homogenising the soil samples, resulting in loss of natural soil structure, loss of roots and biota, short-exposure time to flood conditions, and the control of variable factors such as temperature and soil water conditions (Frohne et al., 2011; Rinklebe et al., 2010). Redox conditions are often simulated and controlled through additions of O_2, to increase E_H, and N_2, to lower E_H (Frohne et al., 2014, 2011; Schulz-Zunkel et al., 2015; Shaheen et al., 2016; Shaheen and Rinklebe, 2017). These differences make extrapolation of these laboratory-based findings to field situations difficult (Hooda, 2010).

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

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

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

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

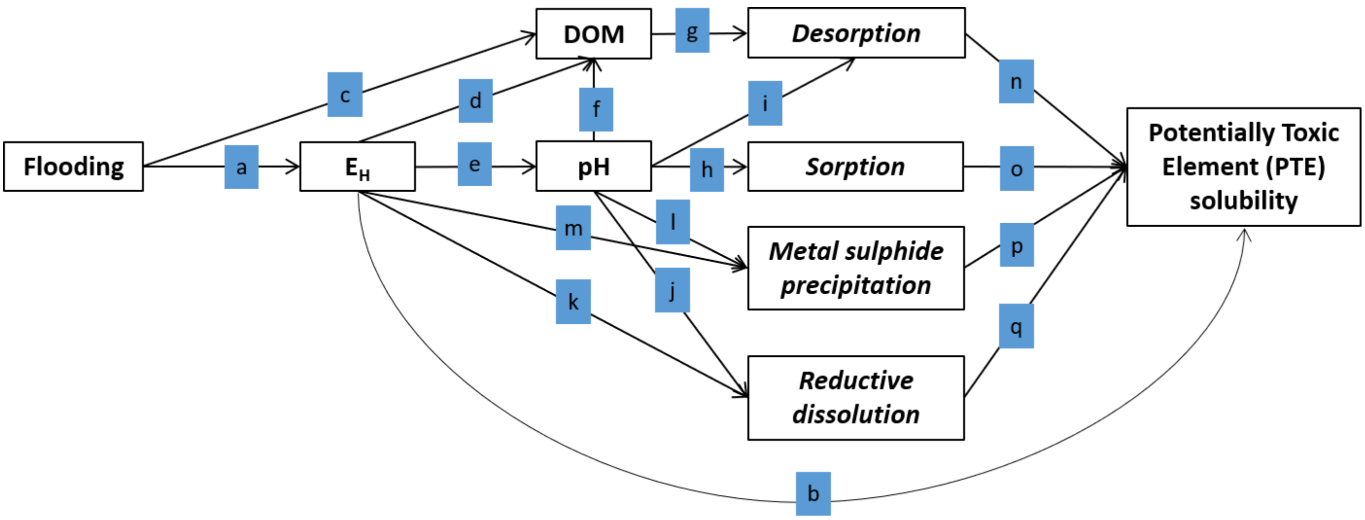

Figure 2: Conceptual model depicting the key processes influencing the solubility of Potentially Toxic Elements (PTEs) after a soil becomes flooded.
a) Oxygen is rapidly consumed by microbial and root respiration, decreasing the redox potential (E_H). b) Decreasing E_H can lead to redox sensitive elements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

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

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

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

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

2.3.1 Soil texture and related properties

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

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

2.3.2 Organic matter

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

2.3.3 Salinity

Salinity is proportional to the conductivity of a sample solution; which is a measure of its ability to conduct or carry electric current and depends on the presence of charged ion species (anions and cations) (Ander et al., 2016; de Souza Machado et al., 2018; De Vivo et al., 2008)). Increasing salinity in flood water is associated with an increase in major cations that compete with PTEs for sorption sites. This competition promotes PTEs desorption from the floodplain soil in the absence of sulphides and hence increases total PTEs concentrations in the soil porewater (Rinklebe and Du Laing, 2011). The presence of Ca-salts releases more PTEs into the soil solution compared with Na-salts that are less competitive for sorption (Du Laing et al., 2009; Hahne and Kroontje, 1973).

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

2.3.4 Redox potential (E_H)

Waterlogging of soils generally results in a reduction in oxygen availability due to rapid consumption of oxygen by soil microbial activity and root respiration (Du Laing et al., 2007; Rinklebe and Du Laing, 2011) and because the dissolution of oxygen through water is many times slower than through air (Alloway, 1995; Du Laing et al., 2009; Frohne et al., 2015; Schulz-Zunkel et al., 2015). The soil microbial community (e.g. bacterial species such as *Thiobacillus ferroxidans*, *Thiobacillus thiooxidans* and *Leptospirillum ferrooxidans*) then uses alternative electron acceptors (such as nitrate, sulphate and Fe/Mn oxides), in anaerobic respiration, which results in a decrease in redox potential (E_H) (Maluckov, 2017) as the floodplain soils change from oxic ([O2]>30 µmol L^{-1}) to anoxic ([O2]<14µmol L^{-1}) conditions (Bellanger et al., 2004). Associated alkalinity generation drives increases in soil pH, a change which can be observed after a few days (Du Laing et al., 2007; Johnston et al., 2014; Karimian et al., 2017). Soil temperature has been found to dictate the rate and type of redox reactions; with soils at low temperatures (1 - 4 °C) requiring greater durations of saturation (20 days) before the onset of reducing conditions were seen, whereas soils at higher temperatures (above 9 °C) only required 2 days of saturation (Vaughan et al., 2009).

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

The presence of variable charge minerals, such as Fe and Mn oxides, phosphates, carbonates and sulphides provide a reaction surface for sorption processes, allowing PTEs to bind and become immobilised (Antoniadis et al., 2018; De Jonge et al., 2012; Sipos et al., 2014; Violante, 2013). Reducing conditions change the oxidation state of Fe and Mn, increase their solubility and may have indirect effects (known as reductive dissolution) on the mobility of associated metal cations (e.g. As, Cd, Cu, Ni, Pb, and Zn), releasing them from the solid phase to pore waters, depending on flood duration (Abgottspon et al., 2015; Ciszewski and Grygar, 2016; Du Laing et al., 2009; Frohne et al., 2011; Karimian et al., 2017; Rinklebe and Du Laing, 2011; Schulz-Zunkel et al., 2015; Shaheen et al., 2016, 2014b; Vaughan et al., 2009). Redox processes are a key factor for the reductive dissolution of Mn and Fe (hydr)oxides, these processes are often catalysed by microorganisms and result in the release of PTEs from the sediment (Du Laing et al., 2009; Frohne et al., 2011; Stafford et al., 2018; Yang et al., 2015). Relatively insoluble Fe(III) and Mn(IV) prevail under aerobic soil conditions providing sorption surfaces for many metals, whereas under anaerobic conditions Mn(IV) and Fe(III) are reduced to more soluble forms (Mn(II) and Fe(II)) with consequential dissolution of Mn and Fe hydrous oxides, co-sorbed PTEs ions (e.g. As, Cd, Cr, Ni and Pb), are released into soil solution (Simmler et al., 2017; Stafford et al., 2018; Yang et al., 2015). After inundation, Fe and Mn may re-precipitate as oxides and can bind (by desorption or co-precipitation) the trace metals back into the solid state (Ciszewski and Grygar, 2016; Davranche et al., 2011; Du Laing et al., 2009).

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

2.3.5 Soil pH

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

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

2.3.6 Dissolved organic matter (DOM)

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

2.3.7 Temperature

As temperatures are predicted to increase as a result of climate change, they may become a factor that contributes to greater release of PTEs from the soil during a flood (Visser et al., 2012). Soils are affected by variations in air temperature which, in turn, affects the rate of biogeochemical processes during a flooding event, including decreasing redox potential and, ultimately, influences the rate and extent to which PTEs are released/ desorbed from POM into surface water and groundwater (Arnell et al., 2015; González-Alcaraz and van Gestel, 2015; Sánchez-Rodríguez et al., 2019; Shaheen et al., 2016; Stahl et al., 2013). Increases in temperature raise the ion activity in soil solution, and also make plants more active, which may lead to greater plant root uptake of soil water and dissolved/labile PTEs within this water (Sherene, 2010). Arsenic release from flooded soils was found to have temperature dependence, with As solubilisation increasing as temperature increased (Simmler et al., 2017; Visser et al., 2012). Temperature increases are attributed to a decrease in the water viscosity resulting in dissociation of molecules and a subsequent increase in the number of ions in the solution. For every degree Celsius increase in temperature there is an observed increase in electrical conductivity of 1.9% (Ander et al., 2016; Ma et al., 2011).

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

Groundwater and river temperatures may have a strong effect on floodplain soil temperatures (Andersen, 2018). In warmer seasons they will generally be cooler than soil temperatures, but the opposite occurs in cooler seasons. Also, changes in soil moisture content, as a result of flood events, will affect the soil thermal properties such as thermal conductivity and heat capacity (Lu et al., 2007), thereby also affecting the spatial and temporal variation in the soils' temperature regime. A laboratory microcosm experiment with mining-contaminated topsoil and subsoil samples saturated for up to 41 days at temperatures ranging between 10-25^{o}C, found that soil temperature increased the solubilisation of As, particularly in the topsoil when saturated for 1-2 weeks (Simmler et al., 2017). This means, for some PTEs, flooding during warmer seasons may result in greater mobilisation than flooding during cooler seasons. More seasonal field observations are required to understand the subtle interactions and feedbacks between soil moisture, floodwater temperature, and mobility of PTEs.

2. 4. Soil biological processes that influence PTEs mobility

2.4.1 Soil organisms

Floodplain soils contain a great diversity of organisms that are known to contribute to the physical structure of the soil/sediment through bioturbation which influences the biogeochemical cycling of PTEs through oxygen diffusion, redox gradient and decomposition of dissolved organic matter (Classen et al., 2015; He et al., 2019; Hooda, 2010; Selinus et al., 2005). As the soil pore spaces are filled with water, oxygen diffusion is low so microbial respiration relies on alternative electron acceptors (e.g. NO_3^-, Mn, Fe and S), resulting in reducing conditions (decreasing E_H) that simultaneously increase pH (Matern and Mansfeldt, 2016), and the changes to PTEs mobility (Figure 3) that are described in previous sections. Changes in the chemical speciation of PTEs can also occur due to microbial processes in reducing conditions, for example, sulphate reducing bacteria can methylate Hg in anoxic conditions (Ma et al., 2019).

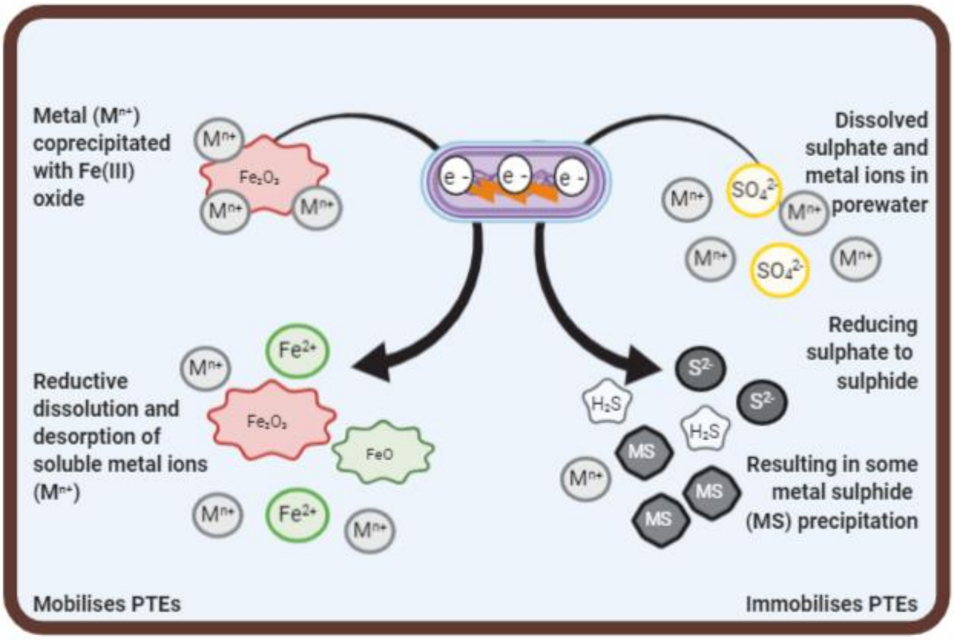

Figure 3: Soil microbial processes during inundation of floodplain soil influences mobility of PTEs; (left-hand side) generic metals with valence state (denoted by M^{n+}) are coprecipitated to Fe oxides and are released due to reductive dissolution and (right-hand side) sulphate reduction (sulphate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

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

Flooding has been found to shift the soil biological community structure and function. These changes include a reduction of Gram-positive bacteria, mycorrhizal fungi and earthworms found under flooded conditions (Gregory et al., 2015; Harvey et al., 2019; Unger et al., 2009). Harvey et al., (2019) found that flooding induced short-term alterations to soil microbial biomass but these changes did not persist in the long term; they concluded that temperate systems may be resilient to winter flood stress. The seasonal timing of floods influences the effect that flooding has on the soil microbial community, and so may result in different effects on, and recovery of, the soil microbial community. Sánchez-Rodríguez et al., (2019) subjected a UK agricultural grassland soil in an intact laboratory microcosm to flooding and found that summertime flooding (25°C), resulted in a loss of actinomycetes and arbuscular mycorrhizal fungi, and that these changes persisted post-flood. They expected microbial biomass to increase with flooding at higher temperatures, due to degradation of vegetation releasing labile carbon. However, they found that maintaining live roots and an active rhizosphere were more important for preserving the microbial community in grassland soils. Earthworms also play a role in increasing the mobility and availability of PTEs in floodplain soil through their activity causing changes to the soil microbial populations, pH, DOC or metal speciation (Sizmur et al., 2011; Sizmur and Hodson, 2009) which in turn influences PTEs mobility as discussed in the above sections.

As the PTEs are released into the aqueous phase and mobilised in the environment, they present a potential risk to soil organisms (Ehlers and Loibner, 2006; González-Alcaraz and van Gestel, 2015). Soil organisms uptake PTEs via ingestion of polluted soil, food or pore water and/or via dermal uptake or absorption of soil water, with the soil water being the more important of the two pathways (Chrzan, 2016; Hobbelen et al., 2006; Sivakumar and Subbhuraam, 2005). Vijver et al. (2007) found that the frequency of flooding did not result in consistent changes in the internal PTEs concentrations of earthworms. Earthworms accumulate PTEs in their chloragogenous tissue and have a mechanism that allows them to regulate their internal PTEs concentrations, so when they are introduced to contaminated soils the earthworms reach an equilibrium and when they are returned to uncontaminated/"clean" soils they are able to detoxify and eliminate essential metals through excretion (e.g. Cu and Zn), but not non-essential metals (e.g. Cd and Pb) as detoxification processes involve sequestration within an inorganic matrix or organic ligand (Sizmur and Hodson, 2009; Spurgeon and Hopkin, 1999). While microbes can tolerate larger quantities of essential PTEs, in excess both essential and non-essential PTEs (e.g., Al, As, Cd, Hg, Pb, Zn) can adversely affect microbial communities by altering community structure and taxonomic richness; reducing the microbial biomass and lowering their enzyme activity which results in a decrease of soil diversity (Gadd, 2010; Gall et al., 2015; Wuana et al., 2011).

2.4.2 Plants

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

It is well established that symbiotic fungi, associated with plant roots, regulate the supply of micronutrients and reduce the uptake of non-essential PTEs by plants (Classen et al., 2015; Gadd, 2010; Tack, 2010). Plants, such as *Artemisia* and *Phalaris* species, on the floodplain excrete exudates during inundation which stimulates the activity of microbial symbionts in the rhizosphere, allowing PTEs to be taken up into the vegetation (Gall et al., 2015; Sullivan and Gadd, 2019; Violante et al., 2010; Xu et al., 2020). PTEs are often accumulated in plant root tissues and can sometimes be translocated into the plant shoots. However this is regulated in plants by the Casparian strip and therefore limited (Hooda, 2010; Nouri et al., 2009; Shahid et al., 2017). The uptake and accumulation of PTEs is element and plant-specific (Niu et al., 2007; Rinklebe et al., 2016; Tack, 2010; Violante et al., 2010; Xu et al., 2020). The mobilisation and uptake of PTEs by plants may pose a potential environmental risk (Shaheen and Rinklebe, 2014). European floodplains are most commonly used as grassland for grazing cattle or hay production, whereas in other regions e.g. India, they are used for crops like rice, which raises concerns for possible pollutant transfer from the floodplain soil into the surrounding water bodies, then uptake and potential biomagnification of PTEs into the food chain (Martin et al., 2014; Overesch et al., 2007; Tóth et al., 2016a). However, the hyperaccumulation of PTEs by some plants (e.g. sunflower, mustard (Brassicaceae), alfalfa and Ricinus) has resulted in them being considered for phytoremediation of contaminated floodplain soils (Gall et al., 2015; Niu et al., 2007; Nouri et al., 2009; Shaheen et al., 2016; Violante et al., 2010).

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

3. Summary and further research needs

3.1. Summary of current understanding

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

3.2 Knowledge gaps and recommendations for future research

Floods are dynamic events that expose floodplain soils to water with rapidly changing flows, chemical composition, and sediment load. They can be difficult to predict, due to their different types (e.g. overbanking or groundwater flooding), and the high variation in their magnitude, duration, and frequency of recurrence. Therefore, chemical, physical and biological data from floodplain soils immediately before and immediately after a flooding event are often lacking. However these data would provide the necessary insights into the factors and processes involved in altering the mobility of PTEs during and after a real flooding event (Barber et al., 2017). The effect of flooding on PTEs mobility can be difficult to predict due to there being several factors (e.g. speciation, release through biological degradation and competitive action of other ions) or interactions between factors (e.g. changes in E_H caused degradation of POM) influencing PTEs mobility (Tack and Verloo, 1995).

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

A number of factors were identified that contribute to whether the mobility of PTEs will increase or decrease during inundation of a floodplain, which may be interconnected or work in combination to affect PTEs mobility. As a result, different soils with differing mineralogy and thus different biogeochemical and physical properties, will likely respond differently to flooding. Individual studies tend to focus on one floodplain site. However, knowledge based on one river catchment may not be particularly useful for predicting the impacts of flooding at another site with different mineralogy and physical and chemical characteristics. A more fundamental mechanistic understanding is required to inform the development of predictive models. Therefore, more coordinated work encompassing multiple contrasting sites is required to understand the relative importance of key soil properties (e.g. mineralogy, POM, soil pH, texture; and how these affect derived soil properties such as hydraulic and thermal soil properties) on influencing the impact of flooding on the mobility of PTEs.

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

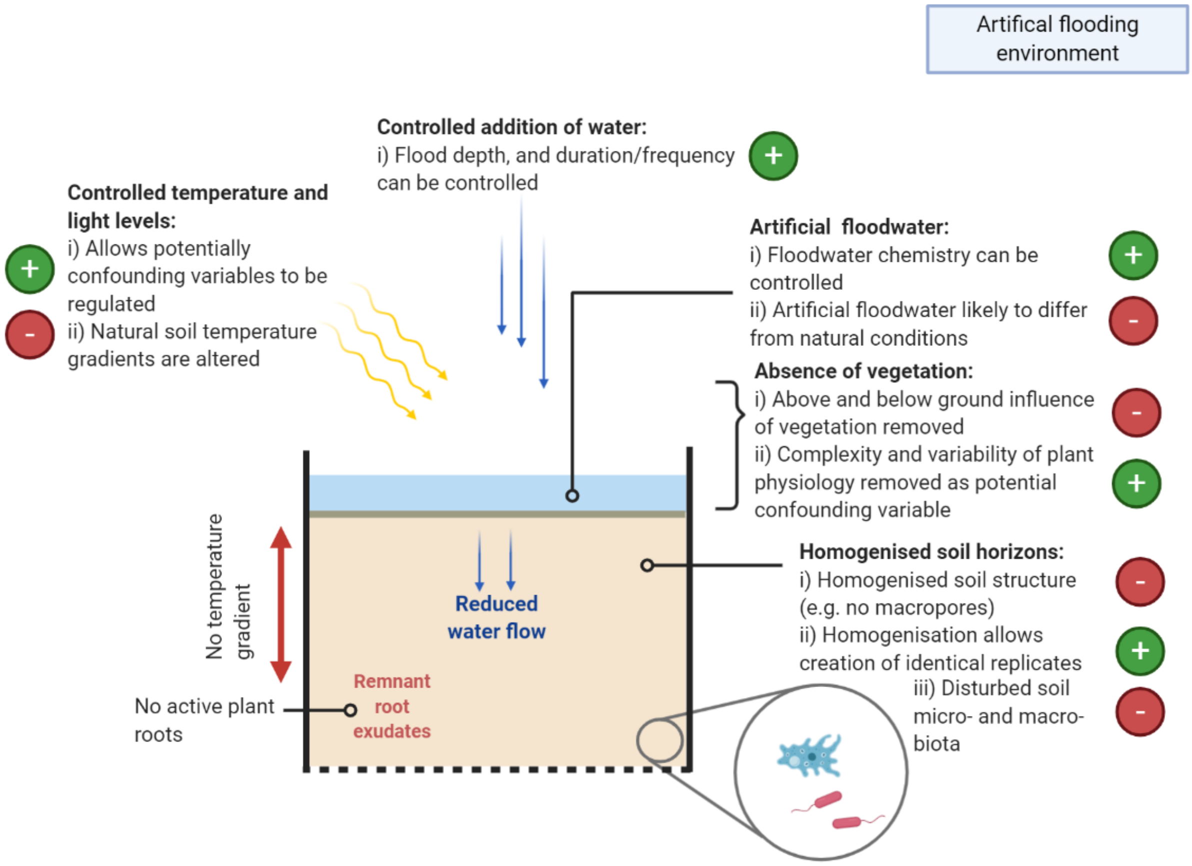

Figure 4: Strengths (+) and weaknesses (-) of laboratory-based studies for researching the impact of flooding on mobility of PTEs. Created with BioRender.com.

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Supplementary Material

One supplementary table (Table S1) is provided

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