

TECHNICAL REPORT

Trace Elements in the Environment

Mobility of arsenic and vanadium in waterlogged calcareous soils due to addition of zeolite and manganese oxide amendments

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Abstract

Addition of manganese(IV) oxides (MnO_2) and zeolite can affect the mobility of As and V in soils due to geochemical changes that have not been studied well in calcareous, flooded soils. This study evaluated the mobility of As and V in flooded soils surface-amended with MnO_2 or zeolite. A simulated summer flooding study was conducted for 8 weeks using intact soil columns from four calcareous soils. Redox potential was measured in soils, whereas pH, major cations, and As and V concentrations were measured biweekly in pore water and floodwater. Aqueous As and V species were modeled at 0, 4, and 8 weeks after flooding (WAF) using Visual MINTEQ modeling software with input parameters of redox potential, temperature, pH, total alkalinity, and concentrations of major cations and anions. Aqueous As concentrations were below the critical thresholds ($<100 \mu\text{g L}^{-1}$), whereas aqueous V concentrations exceeded the threshold for sensitive aquatic species ($2\text{--}80 \mu\text{g L}^{-1}$). MnO_2 -amended soils were reduced to sub-oxic levels, whereas zeolite-amended and unamended soils were reduced to anoxic levels by 8 WAF. MnO_2 decreased As and V mobilities, whereas zeolite had no effect on As but increased V mobility, compared to unamended soils. Arsenic mobility increased under anoxic conditions, and V mobility increased under oxic and alkaline pH conditions. Conversion of As(V) to As(III) and V(V) to V(IV) was regulated by MnO_2 in flooded soils. MnO_2 can be used as an amendment in immobilizing As and V, whereas the use of zeolite in flooded calcareous soils should be done cautiously.

1 | INTRODUCTION

Behavior of metals and the effects of amendments in metal bioavailability are different in calcareous soils than that of

noncalcareous soils (Moral et al., 2005). The available data is still limited to the risk evaluation of the bioavailable fraction of the potentially toxic elements (PTEs) in calcareous soils (Rezapour et al., 2019), and therefore, more studies are needed to better understand the effects of PTEs in these soils under changing geochemical environments. Arsenic and V are redox-sensitive elements, and their fate in the soil environment is determined by the changes in Eh/pH values, organic C, and the related chemistry of S, Fe, and Mn (O'Loughlin et al., 2021; Ying et al., 2012). Soil amendments

Abbreviations: CCE, calcium carbonate equivalent; Eh, Redox potential; ICP-AES and -MS, inductively coupled plasma atomic emission spectroscopy and mass spectroscopy; MnO_2 , manganese (IV) oxide; PTE, potentially toxic elements; WAF, weeks after flooding; XRD, X-ray diffractometry.

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typically act as adsorbent materials to bind toxic metals to the surface, form precipitates, or change their speciation (Indraratne et al., 2021; Zou et al., 2019). Manganese oxides alter the speciation of redox-sensitive elements and are well-known scavengers for PTEs in contaminated soils (Abernathy et al., 2021; Komarek et al., 2013). Zeolites are microporous aluminosilicates that can be modified to increase their metal adsorption capacity by improving the surface area, porosity, and ion exchange capacity (Zheng et al., 2021). Zeolite can be used for heavy metal immobilization because it has high thermal and chemical stabilities and does not cause secondary pollution (Salehi et al., 2020). Studies on the mobility of As and V in calcareous soils with the addition of amendments, such as zeolite and manganese oxides, are limited.

Arsenates [As(V)] in the forms of H_2AsO_4^- and HAsO_4^{2-} occur in oxidized conditions, whereas arsenites [As(III)] in the forms of H_3AsO_3 and H_2AsO_3 occur in reduced environments (Fan et al., 2014). Some minerals with the ability to oxidize As such as manganese(III/IV) oxides alter the extent of As retention by changing As speciation (Ying et al., 2012). Arsenic removal involved bidentate, binuclear inner-sphere complexation of arsenate at the surfaces of ferric precipitates (Sherman & Randall, 2003; Wang et al., 2021). Arsenates showed inner-sphere complexation with oxyhydroxides of goethite, lepidocrocite, mackinawite, and pyrite and outer-sphere complexation with residual FeS (Farquhar et al., 2002). At oxic-to-anoxic transitions, arsenic is released into the surrounding aqueous medium by reductively dissolving iron hydroxides (Smedley & Kinniburgh, 2002). The industrial production of V has doubled since 2002, and the total global production of V in 2019 was 73 G t (O'Loughlin et al., 2021; Watt et al., 2018). Therefore, V is reemerging as a PTE receiving increased attention due to its possible toxicity and its increased use in society (O'Loughlin et al., 2021; Watt et al., 2018; White & Levy, 2021). Redox-sensitive V prevails in the oxidation states of V(III), V(IV), and V(V) in the soil environment, where the latter has the highest solubility and toxicity (Larsson et al., 2017). Vanadium toxicity in aquatic and terrestrial environments is dependent on the concentration, oxidation state, and species of V (Abernathy et al., 2021).

The Red River floodplain area is flat and contains clay soils with poor permeability, causing frequent flooding, and floodwater carries nutrients and metals to waterways. Therefore, we have been testing different soil amendments, including MnO_2 and zeolite, to prevent P losses from flat floodplain agricultural soils under short-term flooding (Attanayake et al., 2022; Dharmakeerthi et al., 2019; Vitharana et al., 2021), but we did not investigate the fate of PTEs upon the addition of such amendments. Manganese oxide is a well-known PTE scavenger used in contaminated soils (Abernathy et al., 2021), and zeolite is also a proven soil amendment for PTEs (Shi et al., 2009). Previously, we observed changes of Eh and pH in uncontaminated, agricultural soils under short-term

Core Ideas

- MnO_2 amendment decreased the mobility of As in 50% of tested soils.
- Aqueous As concentrations increased with increasing flooding time.
- Amendment zeolite enhanced, whereas amendment MnO_2 reduced the mobility of V.
- The highest V release was observed under oxic and alkaline conditions in soils.
- Amendment MnO_2 delayed the conversion of As(V) to As(III) and V(V) to V(IV).

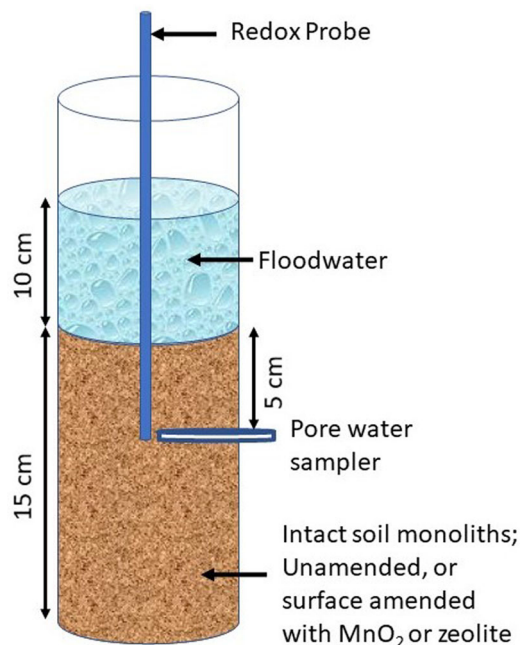
flooded conditions contributing to an enhanced release of otherwise stable PTEs to pore water and floodwater (Indraratne & Kumaragamage, 2018). This study was conducted with the objectives of (a) to determine the concentrations of As and V in pore water and floodwater of calcareous soils during an 8-week flooded period to understand their potential mobility, and (b) to compare the mobility of these elements in unamended soils (control) and soils amended with zeolite or Mn(IV) oxide (MnO_2) under short-term flooded conditions with the support of MINTEQ thermodynamic modeling. We hypothesized that the mobility of As and V would increase with the flooding of calcareous soils because of decreasing Eh and pH resulting in physical and reductive dissolution reactions, and zeolite and MnO_2 amendments will reduce PTE mobility in these flooded calcareous soils through enhanced adsorption.

2 | MATERIALS AND METHODS

2.1 | Soil and material characterization

This experiment was conducted with four soils (Dencross, Lakeland, Osborne, and Scantebury soil series) using intact soil monoliths collected from agricultural fields in the Red River Valley of Manitoba in May 2019, as described in Attanayake et al. (2022). Soil classification information, the analysis of basic properties of soils such as soil texture, pH, calcium carbonate equivalent, Mehlich-3 extractable metals (Ca, Mg, Al, Fe, Mn, As, and V), and cation exchange capacity methods were given in details in Attanayake et al. (2022).

Analysis of total As and V in MnO_2 , zeolite, and the four soil samples were conducted by one-step digestion using 0.25 g of soil/material with perchloric, nitric, and hydrofluoric acids and analyzed by inductively coupled plasma mass spectrometry (ICP-MS; Agilent 7900). The analytical results were corrected for interelement spectral interferences (ALS code ME-MS61).



Experiment setup

FIGURE 1 Experimental setup

2.2 | Simulated summer flooding experiment

Soil monoliths of 15 cm were collected in 30 cm length and 10 cm diameter polyvinyl chloride tubes that had 3-mm holes drilled at 10 cm from the bottom (Figure 1) to extract soil pore water using Rhizon MOM soil solution samplers (Rhizosphere Research Products). Manganese(IV) oxide (98%) and zeolite (zeolite Y: $\text{Na}_{54.0}\text{Al}_{54.1}\text{Si}_{137.9}\text{O}_{384}\cdot 245.1\text{H}_2\text{O}$) purchased from Thermo Fisher Scientific and Sigma-Aldrich, respectively, were used for the study as amendments. Amendments were added to the surface of soil monoliths at the rate of 0.25% w/w (equivalent to 5 Mg ha^{-1}) and then preincubated for 7 days at $22 \pm 2^\circ\text{C}$ at field moisture capacity. Triplicate soil monoliths per each treatment were flooded by adding deionized water (Milli-Q; $18 \text{ M}\Omega \text{ cm}$) from the top. Immediately after flooding, a redox potential probe (Paleo Terra) with a platinum sensor was installed inside the soil monolith at 5-cm depth (Figure 1). Monoliths were incubated for 8 weeks maintaining a constant 10 cm water head to simulate summer flooded conditions ($22 \pm 1^\circ\text{C}$). Pore water (soil solution) and floodwater samples were collected on a biweekly basis for 8 weeks using the Rhizon-MOM samplers with $0.15 \mu\text{m}$ filters and syringed attached $0.45 \mu\text{m}$ membrane filters, respectively. Because there was a difference in filter size between floodwater and pore water, floodwater would include more colloidal PTEs than pore water, which, however, allows comparisons of pore water (or floodwater) across flooding times and treatments. Further details of the collection of pore water and

floodwater and pH measurements are given in Attanayake et al. (2022). Water samples were acidified with $50 \mu\text{l}$ of concentrated nitric acid (trace metal grade) and stored at 4°C to analyze for concentrations of Ca, Mg, Fe, Mn, Al, As, and V using ICP-AES spectroscopy (ICP-AES; Thermo iCAP 6500 Duo, Cambridge, UK). The detection limits of ICP-AES were 0.030 and 0.001 mg L^{-1} for As and V, respectively.

Anions (Cl^- , F^- , NO_3^- , PO_4^{3-} , and SO_4^{2-}) were measured using ion chromatography, and alkalinities of pore water samples were assessed using the titrimetric method at 0, 4, and 8 weeks after flooding (WAF) (Attanayake et al., 2022). Anions and cations were measured within 2 weeks after the final sampling. Redox potential in soil monoliths was measured at 2-week intervals using the permanently installed Pt electrodes relative to an Ag–AgCl reference electrode inserted to the floodwater, and redox status (pe) was calculated in relation to redox potential measurements (Attanayake et al., 2022).

At the end of the incubation period, unamended, MnO_2 -amended, and zeolite-amended soils were subjected to an X-ray diffraction analysis (Bruker D8 ADVANCE, Germany). Flooded soil samples from columns were kept at 4°C cooler until analysis. Osborne soil was selected for the X-ray diffraction (XRD) characterization as both As and V showed a significant amendment effect after the addition of zeolite and MnO_2 . Further information on analysis is given at Attanayake et al. (2022).

2.3 | Thermodynamic modeling

Thermodynamic modeling is an indirect method that would predict the mineral phases that may present in soils based on soil solution parameters. It has been used to predict the chemical transformations that occur in soils as a result of changing soil environment caused by soil–water management practices and natural causes (Schröder et al., 2005; Xu et al., 2019). The geochemical equilibrium speciation modeling software, Visual MINTEQ 3.1 (Gustafsson, 2013), was used to model aqueous As and V species in MnO_2 -amended, zeolite-amended, and unamended soils at 0, 4, and 8 WAF, respectively.

Experimental input parameters used for Visual MINTEQ in unamended, MnO_2 -amended, and zeolite-amended soils are given elsewhere (Attanayake et al., 2022). Redox-coupled reactions, Fe(II)/Fe(III) , Mn(II)/Mn(IV) , As(III)/As(V) , and V(IV)/V(V) , were defined in the software.

2.4 | Statistical analysis

Analyses of variance for floodwater and pore water elemental concentrations were performed for each soil separately

using the generalized linear mixed model (GLIMMIX) in SAS (SAS Institute Inc., 2013), using treatment as the fixed effect, replicate as random effects, and time as the repeated measures factor. When ion concentrations were below detection limits, a value corresponding to half of the detection limit of the element was assigned before analysis (Wong et al., 2002). Principal component analysis (PCA) was conducted using the PROC FACTOR procedure in SAS (SAS Institute Inc., 2013) for pore water PTE concentrations and related parameters to identify possible clusters between variables and factors influencing the release of PTEs to pore water (more details in the [Supporting Information](#) section). For all statistical analyses, significance was determined at $\alpha = 0.05$.

3 | RESULTS

3.1 | Initial soil properties

All soils were clay in texture, neutral in pH, and high in organic matter (8.7%–11.0%) and CEC (36–86 $\text{cmol}^{(+)} \text{kg}^{-1}$; Table S1). The soils were strongly calcareous with CCE values ranging from 89 to 167 g kg^{-1} . Mehlich-3-extractable As and V in soils varied in a narrow range among soils, whereas total As and V varied from 8.3 to 10.9 mg kg^{-1} and 89 to 167 mg kg^{-1} , respectively. Total As concentrations were 0.5 and 2.1 and total V concentrations were <1 and 3 mg kg^{-1} , respectively, in zeolite and MnO_2 amendments. The XRD of MnO_2 revealed a mineralogical signature of amorphous MnO_2 with traces of cryptomalene-like solid phases (Attanayake et al., 2022).

3.2 | Redox (Eh), pH, and redox status (pe + pH) changes in flooded soils

Redox (Eh), pH, and redox status changes of flooded soils were reported in detail elsewhere (Attanayake et al., 2022). In brief, the pore water pH in unamended soils decreased from 6.79 to 6.59 in Dencross, 7.76 to 7.18 in Lakeland, and 7.37 to 6.99 in Osborne and increased from 6.59 to 6.79 in Scantbury soil during the flooded time. Zeolite-amended soils had higher pore water pH than unamended and MnO_2 -amended soils because zeolite was Na-based (Na 7.2%) and alkaline (Attanayake et al., 2022).

Initial soil Eh (0 WAF) ranged from +350 to +425 mV in all treatment combinations, and Eh gradually declined to +38 mV to –88 mV by the end of the flooding period (8 WAF) in unamended and zeolite-amended soils (Attanayake et al., 2022). Soils amended with MnO_2 maintained a higher Eh compared to the unamended and zeolite-amended treatments (+196 to +43 mV).

Essington (2015) classified pe + pH values (redox status) of soils as oxic (pe + pH > 14), sub-oxic (pe+pH = 9–14), and anoxic (pe + pH < 9). MnO_2 -amended treatments became sub-oxic at ~4 WAF for Osborne soil and at ~6–8 WAF for the other three soils, compared to unamended and zeolite-amended treatments, which became sub-oxic at ~2 WAF (Attanayake et al., 2022).

Pore water Fe and Mn concentrations increased during flooding in all treatments of all four soils. Zeolite-amended soils had the lowest Mn, whereas MnO_2 -amended soil had the lowest Fe in pore water throughout the flooding period (Attanayake et al., 2022).

3.3 | Changes in As and V concentrations in pore water and floodwater

Arsenic concentrations were higher in pore water than in floodwater for all soils (Figure S1a). Maximum As concentration in floodwater was 35 $\mu\text{g L}^{-1}$ in unamended control and less than the detection limit (0.030 mg L^{-1}) for all zeolite and MnO_2 -amended soils except for zeolite-amended Osborne soil at 6 WAF. Pore water As concentrations were below detection limit for all treatment combinations at 0 WAF but reached high concentrations up to ~100 $\mu\text{g L}^{-1}$ in unamended Dencross soil at 6 WAF and 70 $\mu\text{g L}^{-1}$ in unamended Scantbury soil at 8 WAF. Our previous studies using soils from uncontaminated, calcareous, and agricultural lands have shown that pore water As concentrations ranged from <3.0 to 65 $\mu\text{g L}^{-1}$, whereas floodwater concentrations were two-to-threefold less than that of pore water (Indraratne & Kumaramage, 2018). In a review article, Smedley and Kinniburgh (2002) also reported high concentrations of As in pore waters extracted from unconsolidated sediments, often with sharp contrasts to the concentrations observed in overlying surface waters.

Vanadium in unamended soils varied from 5 to 45 $\mu\text{g L}^{-1}$ in pore water and 2 to 10 $\mu\text{g L}^{-1}$ in floodwater (Figure S1b). Zeolite-amended soils had higher aqueous V concentrations (ranging from 5 to 108 $\mu\text{g L}^{-1}$ across soils) than the unamended control, with lower concentrations in Dencross (<40 $\mu\text{g L}^{-1}$) and Scantbury (<15 $\mu\text{g L}^{-1}$) soils. Floodwater V concentrations in Lakeland and Osborne soils were <30 and <50 $\mu\text{g L}^{-1}$, respectively, whereas respective pore water concentrations were 12-fold and 5-fold higher than the corresponding floodwater concentrations. Previous studies with microcosm experiments also reported higher pore water V concentrations with short-term flooding (Shaheen, Rinklebe, Rupp, et al., 2014). High aqueous V concentrations in unamended and zeolite-amended soils could be a concern due to toxic impacts on sensitive aquatic species as the thresholds for aquatic species range from 1.2 to 80 $\mu\text{g L}^{-1}$ (Abernathy et al., 2021). Reemerging attention on

V led to impose a new drinking water standard ($15 \mu\text{g L}^{-1}$) by the State of California, USA (Schlesinger et al., 2018). This again signifies the potential environmental concerns on reported V concentrations in this study.

For all soils, treatment \times time interaction effect was not significant for As concentrations in pore water. Therefore, As concentrations were averaged across treatments to evaluate the time effect and averaged across time to evaluate treatment effect (Figure 2). Significantly lower pore water As concentrations were found in MnO_2 -amended treatments in Osborne and Scantebury soils, compared to unamended and zeolite-amended counterparts (Figure 2). The effect of flooding time on As concentrations in pore water was significant for all soils but varied among soils. Dencross, Lakeland, and Osborne soils showed increasing As concentrations with flooding time up to 4–6 WAF and then declining thereafter, whereas Scantebury showed steadily increasing concentrations up to the end of the flooding period.

Pore water V concentrations showed treatment \times time significant interaction effect for Scantebury soil, whereas the interaction effect was not significant for Dencross, Lakeland, and Osborne soils. Zeolite-amended treatments had significantly higher V concentrations than unamended, whereas MnO_2 -amended treatments had significantly lower concentrations than unamended (Figure 3). The highest pore water V concentrations were found at the beginning of the flooding period (oxic conditions) and declined over the time in all soils.

3.4 | Principal component analysis of elements and soil properties

Three PCs with eigenvalues higher than 1 (before and after rotation) were extracted for amended (MnO_2 and zeolite) treatments, whereas two PCs were retained for unamended treatment (Table S2). In unamended, the first PC (PC1) loaded Eh (0.93), Ca (−0.91), Mn (−0.91), V (0.90), Mg (−0.78), Fe (−0.75), and Al (0.6), whereas PC2 loading As (0.88), pH (−0.83), and Fe (0.55). MnO_2 -amended soils grouped Mn (0.91), V (0.81), Fe (−0.68), Ca (0.60), and Mg (0.44) to PC1, Eh (0.91) and Al (0.80) to PC2, and As (0.80) and pH (−0.81) to PC3. Tested parameters of Eh (0.93), Ca (−0.89), Mn (−0.70), V (0.68), and Al (0.60) loaded to PC1, Fe (0.78) and As (0.84) to PC2, and V (0.69), Al (−0.55), and pH (0.91) loading to PC3 in zeolite-amended treatment.

3.5 | PTE-associated minerals

Visual MINTEQ model did not predict solid mineral phases of As and V. The saturation indices (SIs) of several Fe/Mn oxy(hydr)oxides, calcite, gypsum, Mn-carbonates, and sul-

fides are given in Tables S3–S6. Pore water was always undersaturated ($\text{SI} < 0$) with all Mn oxy(hydr)oxides in all soils. It was supersaturated ($\text{SI} > 0$) with goethite, hematite, hercynite, and lepidocrocite at all times in all soils, except in a few instances. In general, the pore water was supersaturated with ferrihydrite, maghemite, and magnesioferrite at 0 and 4 WAF. It became undersaturated with those minerals later during flooding, except in MnO_2 -amended Scantebury soil, in which the soils were supersaturated throughout the flooding period. Pore water was supersaturated with Mn(II) carbonates at 4 and/or 8 WAF in all soils. Further, it was in equilibrium with calcite ($\text{SI} = 0$) throughout the flooding. Pore water was undersaturated with gypsum, pyrite, manganese(II) sulfide, and arsenic(III) sulfide at all times in all soils.

The XRD spectra showed clear diffraction peaks for quartz ($d = 0.33 \text{ nm}$) and clay minerals of montmorillonite ($d = 1.4 \text{ nm}$), illite ($d = 1.0 \text{ nm}$), kaolinite ($d = 0.7 \text{ nm}$), and goethite (0.36 nm) in unamended and amended Osborne soils (Figure 4). Doloresite ($\text{V}^{4+}_3\text{O}_4(\text{OH})_4$), a mineral intermixed with other vanadium oxide minerals, was found in the MnO_2 -amended Osborne soil (Mineralogy Database at <http://webmineral.com/help/XRayDiffraction.shtml#.YhaP--hMFPY>).

3.6 | Soil solution speciation of As and V

We are presenting predicted soil solution As and V species for soils that showed significant treatment effects. At the beginning of flooding, only As(V) species (100%) was predicted in pore water in all treatments of both Osborne and Scantebury soils (Figure 5a). Zeolite-amended and unamended Osborne soil was predicted to have almost 100% of total As in pore water as As(III) species at 4 WAF, whereas in MnO_2 -amended treatment, only 84% of total As present was predicted to be in As(III) species at 8 WAF. MnO_2 -amended Scantebury soil predicted only As(V) species present in pore water throughout the flooding time. In contrast, zeolite-amended and unamended treatments were predicted to have 55% and 100% total As in As(III), respectively, at 8 WAF. At 0 WAF, all soils were predicted to have 0% V(IV) in unamended and amended treatments. By the end of the incubation period, predicted V(IV)% varied from 46% to 99% in unamended, 5% to 21% in zeolite-amended, and 0% to 30% in MnO_2 -amended among four soils (Figure 5b).

4 | DISCUSSION

Total As concentrations in soils were below the threshold levels given by the Canadian Council of the Environment (CCME, 2006; 12 mg kg^{-1}). Total V in Dencross, Osborne,

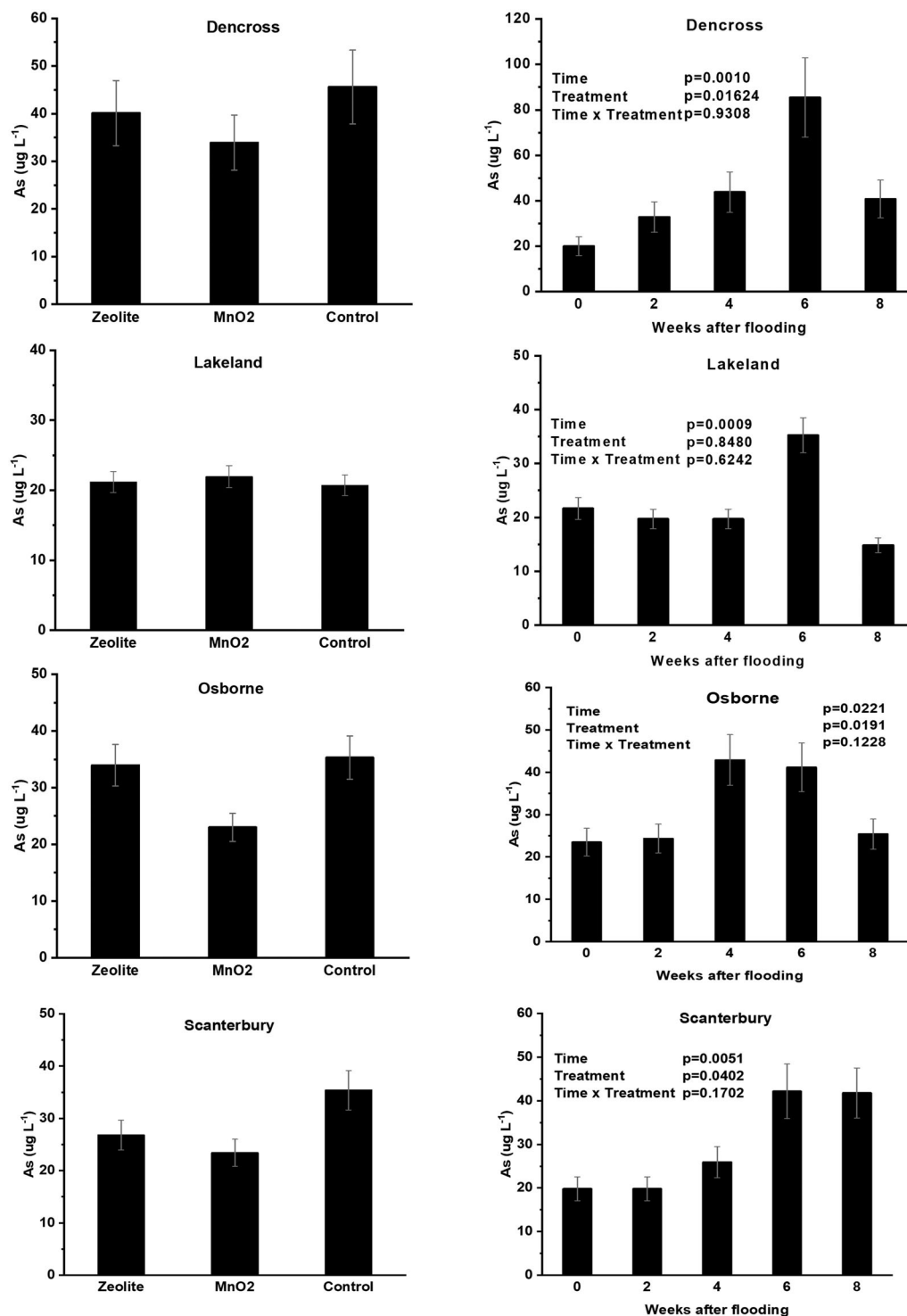


FIGURE 2 Effects of treatment and time on As concentration in pore water during the flooding time in four soils. Vertical bars show standard error of mean

and Scanterbury soils were higher than the CCME threshold value for V in soil, which is 130 mg kg⁻¹ (CCME, 2006). Nevertheless, total V were within the values reported for Canadian prairies that were in the range of 31.7–180.1 mg kg⁻¹ (Mermut et al., 1996). Zeolite-amended soils significantly increased the pH of pore water, compared to unamended.

All amended and unamended treatments were at oxic levels at the start of flooding and became anoxic ($p_e + pH < 8$) in unamended and zeolite-amended treatments, whereas in MnO₂-amended treatments, the soils were at the margin of anoxic to sub-oxic levels ($p_e + pH$, 10 – 8) by 8 WAF. Pore water Fe and Mn increased during flooding in all treatments

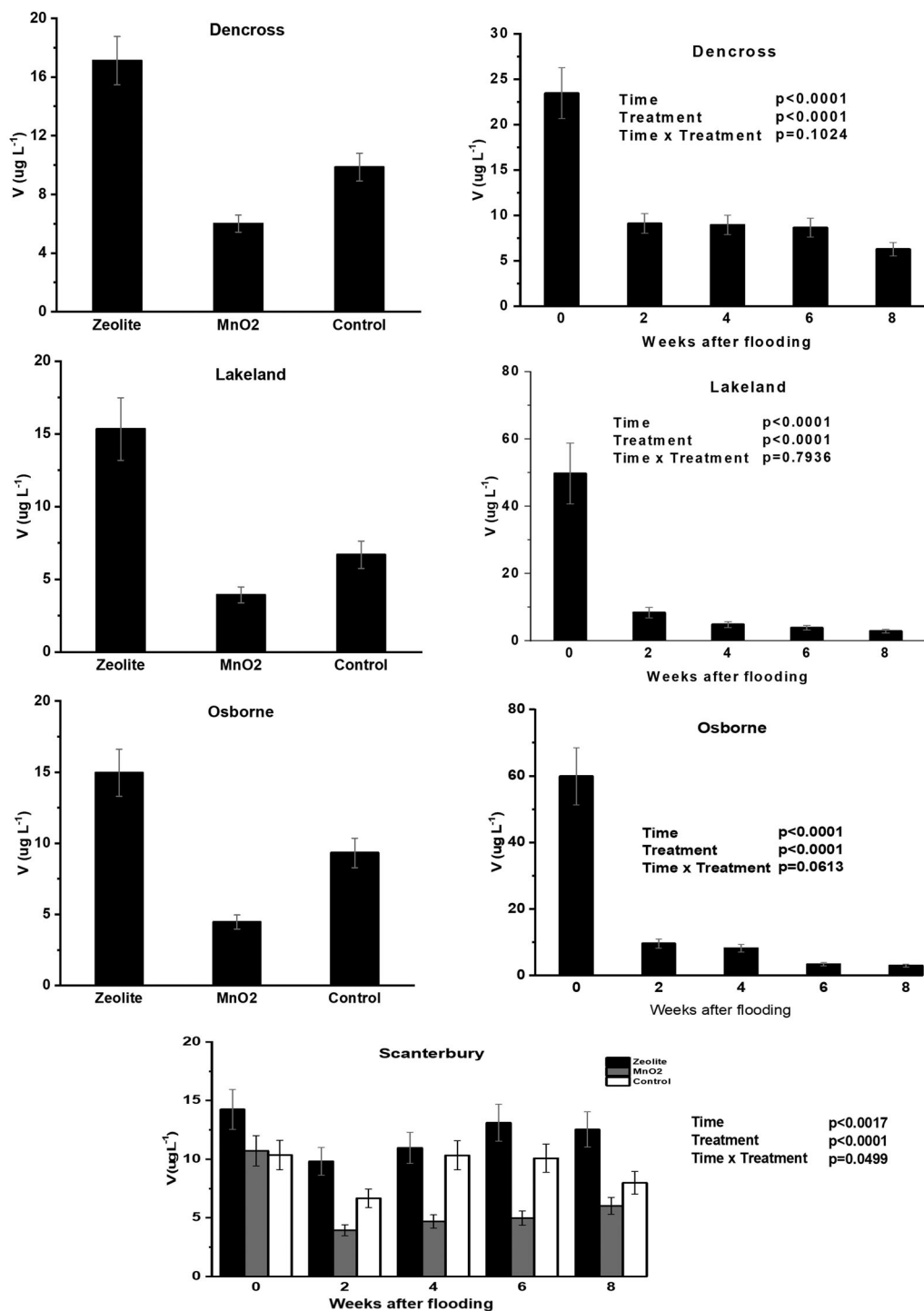


FIGURE 3 Effects of treatment and time on V concentrations in pore water during the flooding time in four soils. Vertical bars show standard error of mean. WAF, weeks after flooding

of all soils. The variation of SI in ferrihydrite, maghemite, and magnesioferrite indicates the reductive dissolution of these minerals during flooding. Although Mn concentration in pore water increased during flooding, the thermodynamic analysis did not indicate the dissolution of any Mn oxy(hydr)oxides. Therefore, considering Eh, Fe, Mn, and SI changes with the time of flooding, the reductive dissolution of Fe/Mn

seems to be favored in unamended and zeolite-amended treatments, whereas MnO₂-amended treatments were less favored.

In general, PTE concentrations in floodwater were less than that of pore water, probably due to slow diffusion rate from pore water to floodwater and the dilution of floodwater with the addition of deionized water to maintain a constant

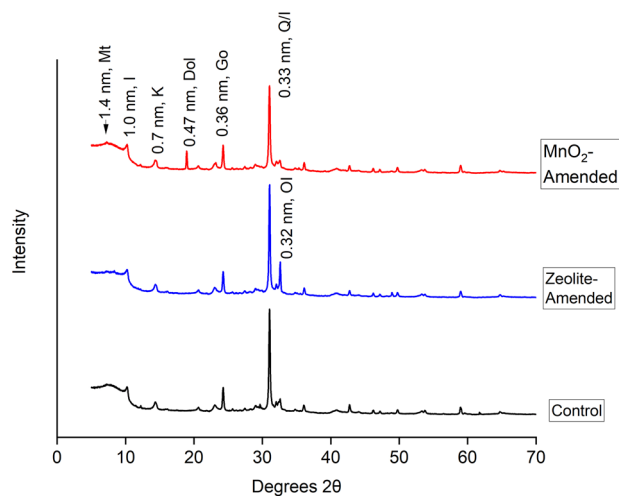


FIGURE 4 X-ray diffraction patterns for unamended, zeolite-amended, and MnO_2 -amended Osborne soil. Dol, Doloresite; Go, Goethite; I, illite; K, kaolinite; Mt, montmorillonite; Oli, oligoclase; and Q, quartz

water-head during the flooding period. Irrigation water quality guidelines given by the CCME (2006) were $100 \mu\text{g L}^{-1}$, for both As and V. Recently acute and chronic hazardous concentrations of V endangering 5% of species (HC5) were estimated as 0.64 and 0.05 mg L^{-1} , respectively (Schiffer & Liber, 2017). Reported aqueous V concentrations in this study were higher than these given threshold values. Accordingly, V concentrations in pore water reached the threshold concentration for some soils, whereas As concentrations were below the threshold values.

Adsorption–desorption reactions are dominant over precipitation–dissolution reactions in uncontaminated or marginally contaminated soils with PTEs (Attanayake et al., 2017; Stevens et al., 2018). As expected, our Visual MINTEQ did not predict the presence of any As or V minerals. Arsenic concentrations in pore water and floodwater increased with flooding time with sub-oxic conditions developed by 4–6 WAF, indicating contributions from reductive dissolution reactions of Fe and Mn minerals. Arsenic species in soils are commonly found adsorbed onto or coprecipitated with hydrous Fe and Mn oxides (Couture et al., 2013; Gupta & Chen, 1978; Ying et al., 2012), which can be released depending on Fe and Mn mineral dissolutions at low Eh under flooded conditions (Cherry et al., 1979; Fan et al., 2014). Takahashi et al. (2004) also found Fe-(hydr)oxide to be a major sorbent of As at oxic conditions, whereas under anoxic conditions, As was released due to the reductive dissolution of Fe (hydr)oxide converting As(V) to As(III). Arsenic release to pore water and groundwater is possibly due to the reductive dissolution of Fe-hydroxides when oxic-to-anoxic transitions take place (Smedley & Kinniburgh, 2002).

PCA showed positive relationships between As and Fe in unamended and zeolite-amended treatments, suggesting the possibility of the reductive dissolution of As-associated Fe minerals releasing both Fe and As under anoxic conditions. For Osborne and Scantbury soils, MnO_2 amendment decreased As release to pore water in comparison to unamended, possibly by controlling reductive dissolution ($8 > \text{pe} + \text{pH}$) of As containing Fe and Mn minerals. This was further confirmed by the differences in predicted As(V) and As(III) speciations in MnO_2 -amended, zeolite-amended, and unamended treatments. As(III) forms are more mobile and toxic than As(V) species in soils (Ying et al., 2012), and As(V) species are more dominant in oxic and sub-oxic systems (Essington, 2015; Zhang & Selim, 2008). In unamended and zeolite-amended soils with $\text{pe} + \text{pH} < 8$, it is likely that the reductive dissolution of Fe oxides increased As concentrations in pore water and converted As(V) to As(III). In MnO_2 -amended soils, higher As(V)% and lower As(III)% were predicted than that of in unamended and zeolite-amended soils. When As(V) was added to an anaerobically treated soil, As(III) was detected in the supernatant, and that concentration of As(III) increased with an increase in total As concentration (Fan et al., 2014). Formation of As–S was not predicted in our study by MINTEQ results. According to Dixit & Hering (2003), the sorption of As(V) and As(III) onto amorphous Fe oxides and goethite is pH dependent; the adsorption of As(V) is favored than As(III) below pH 5–6, whereas the adsorption of As(III) is favored than As(V) above pH 7–8. Therefore, As(III) adsorption onto clay mineral surfaces (Manning & Goldberg, 1997) and to carbonate surfaces in alkaline soils (Mehmood et al., 2009) could be the probable reasons for the decreased As in pore water at 8 WAF.

Both pore water and floodwater V concentrations decreased with increasing flooding time indicating greater solubility and mobility of V under oxic conditions that decreased with the development of anoxic conditions. Vanadate [V(V)], the most common V species under oxic conditions, is the most soluble, mobile, and toxic species of V (Ma & Fu, 2009; Peacock & Sherman, 2004; Vessey & Lindsey, 2020). Aqueous V species in natural systems occur mainly as V(V) and V(IV), where V(V) is thermodynamically stable under oxic conditions, and V(IV) is stable under sub-oxic conditions (Huang et al., 2015). Under anoxic conditions, the mobilization of V decreased because of the reduction of V(V) to relatively less soluble V(IV) (Shaheen et al., 2019). Shaheen, Rinklebe, Frohne et al. (2014) reported the presence of higher V and aromatic C compounds under oxidized than reduced conditions and suggested that V release is due to the dissolution of V-aromatic carbon complexes under oxidized conditions. Reduction of V(V) to V(IV) can decrease solubility through the precipitation of V (hydr)oxides or enhanced sorption at mineral surfaces. Vessey and Lindsay (2020) explained that the reduction of V(V) to V(IV) and the formation of complexes on Fe compounds

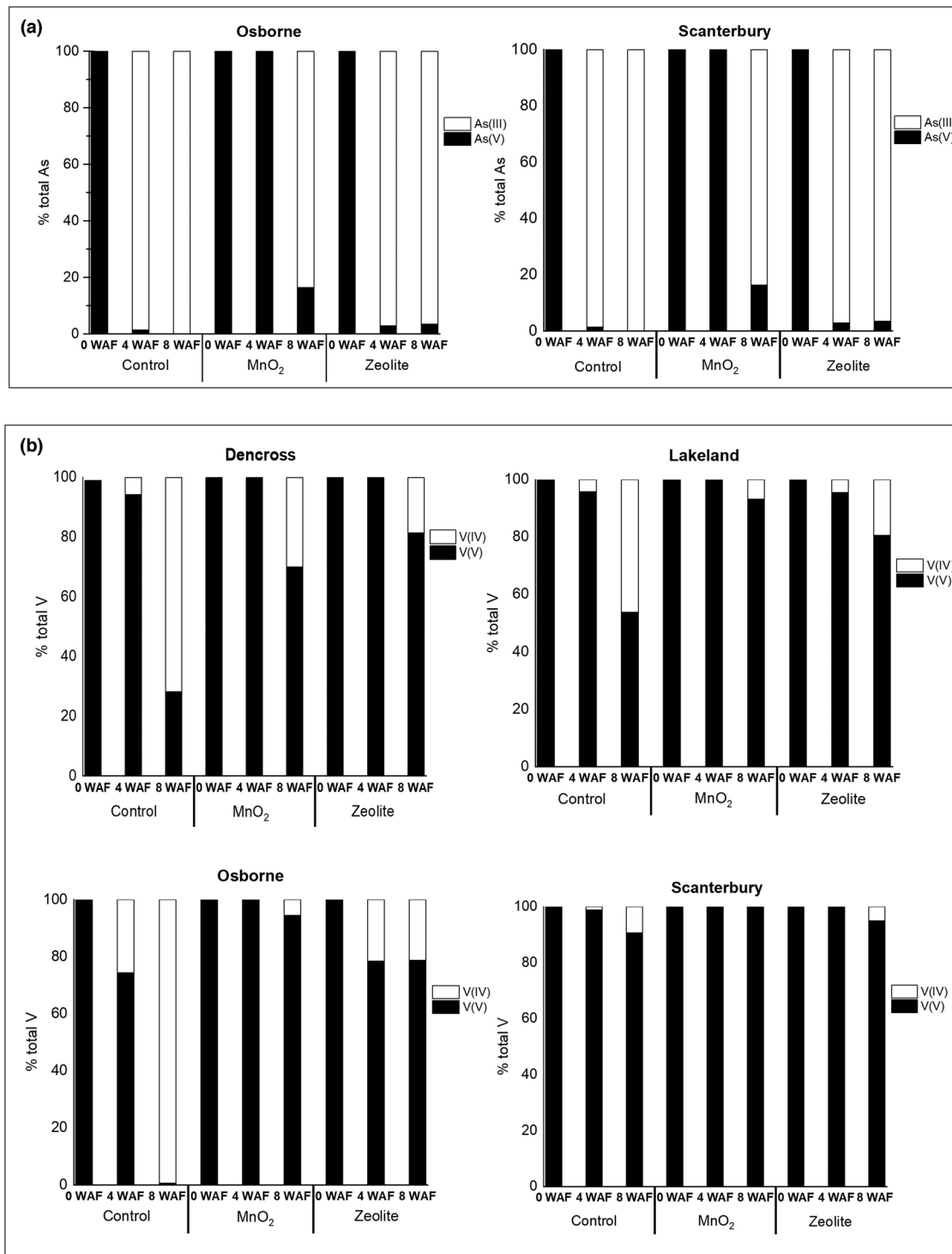


FIGURE 5 Arsenic (a) and V (b) solution speciation during the flooding predicted by the Visual MINTEQ model. WAF, weeks after flooding

and/or with Fe²⁺(aq) reaction products as a reason for decreased vanadate concentrations under reduced conditions.

The V concentrations in pore water and floodwater significantly decreased in the order of zeolite-amended > unamended > MnO₂-amended. In all soils, an application of zeolite enhanced dissolution/desorption reactions of V, whereas MnO₂ enhanced precipitation/adsorption reactions

compared to unamended treatment. In PCA, pore water pH and V concentrations in zeolite-amended soils grouped positively, but not in unamended and MnO₂-amended soils, suggesting the desorption of V at alkaline pH. These results imply that increasing soil pH promotes the mobility of V from soils. Increasing adsorption of V with decreasing pH has been reported previously on ferrihydrite surfaces in

aqueous solutions (Larsson et al., 2017). The higher mobility of V in alkaline soils than in acidic soils was also reported by Shaheen and Rinklebe (2018), where soil organic matter, total sulfur, and carbonates correlated with V. Maximum adsorption capacity of V(V) onto silica was reported at the pH range between 3 and 5 (Gan et al., 2020). Creating more negative charges on organic and inorganic soil colloids at high pH facilitates the release of negatively charged vanadates to soil solution and would have been another reason to have higher V in pore water in zeolite-amended treatments than other treatments. Further, competition between OH⁻ and vanadate for available sites on mineral surfaces at high pH would decrease the adsorption of vanadates. Hence, the pH-driven desorption of V from soil minerals in zeolite-amended soils may be responsible in increasing V concentrations in zeolite-amended soils.

MnO₂-amended soils maintained sub-oxic conditions throughout the flooding period, whereas the corresponding unamended soils became anoxic by the end of the flooding. Based on model predictions, at the initial stage of flooding, all V were in V(V) (100%) form that subsequently reduced to V(IV), in MnO₂-amended soils (~30%) and in unamended soils (>70%) with the time of flooding. Therefore, the MnO₂-amended system, which maintained higher Eh than unamended, may have retained more V as V(V) in the system. Behavior of V in soils is governed by V(V) adsorption to Fe-, Mn-, and Al-(hydr)oxides, allophane, silicate clay minerals, and organic matter (Larsson et al., 2017, 2015). Surface complexation with metal (oxyhydr)oxides was also reported as a mechanism controlling aqueous V(V) concentrations in oxic environments (Vessey & Lindsay, 2020). Abernathy et al. (2021) observed redox reactions between birnessite and V, where birnessite reduced to Mn²⁺, whereas V(IV) oxidized to V(V) consequently forming a bidentate-mononuclear edge-sharing complex between V(V) and birnessite. Formation of such surface complexes of MnO₂ and V(V) is possible in our study because we observed a positive grouping of V and Mn in PCA analysis in the MnO₂-amended system. Compared with zeolite-amended and unamended XRD patterns, MnO₂-amended Osborne soil showed an extra peak for doloresite minerals. The decrease in pore water V concentrations in MnO₂-amended soils in contrast to unamended soils could also be due to the formation of V(IV) oxide minerals. Therefore, redox-driven reactions, surface complexation reactions, and the formation of V(IV) oxide minerals seem to be responsible for lower V concentrations in MnO₂-amended soils than in the unamended soil.

5 | CONCLUSIONS

Manganese(IV) oxide and zeolites due to high sorption capacity were promising amendments in reducing the bioavailabil-

ity of metals. Applying MnO₂ and zeolite followed by 8 weeks of flooding altered the geochemical environment, which changed the mobility of As and V from uncontaminated, calcareous, and agricultural soils. With the development of reducing conditions as flooding progressed, the release of As increased due to reductive dissolution of As-bearing Mn and Fe oxides. Sub-oxic redox status in MnO₂-amended soils maintained a low As concentration in pore water in comparison to unamended soils. Alkaline pH in zeolite-amended soils released higher V to the pore water, likely due to the weak adsorption of vanadates to soil colloids, whereas the MnO₂-amended system maintained a low aqueous vanadate concentration because of strong V adsorption to Al/Fe/Mn minerals and formation of V oxides. The greater release of As upon flooding and V release with the addition of zeolite should be further investigated in calcareous floodplain soils because increasing mobility could lead to environmental risks.

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CONFLICT OF INTEREST

The authors declare that there is no conflict of interest regarding the publication of this article.

AUTHOR CONTRIBUTIONS

Srimathie Priyanthika Indraratne: Conceptualization; formal analysis; funding acquisition; methodology; validation; visualization; writing—original draft; writing—review and editing. Chammi Attanayake: Conceptualization; Data curation; Formal analysis; methodology; visualization; writing—original draft; writing—review and editing. Darshani Kumaragamage: Conceptualization; Funding acquisition; investigation; project administration; resources; supervision; visualization; writing—review and editing. Eragoda Amarawansa: Data curation; Formal analysis; Investigation; Methodology. Doug Goltz: Conceptualization; Project administration; Resources; Supervision; Writing — review & editing. Dan Applin: Methodology.

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