Supporting information

***In-situ* stabilization of multiple heavy metals (Pb, Zn, As) by ferrous sulfate – From batch experiments to pilot study**

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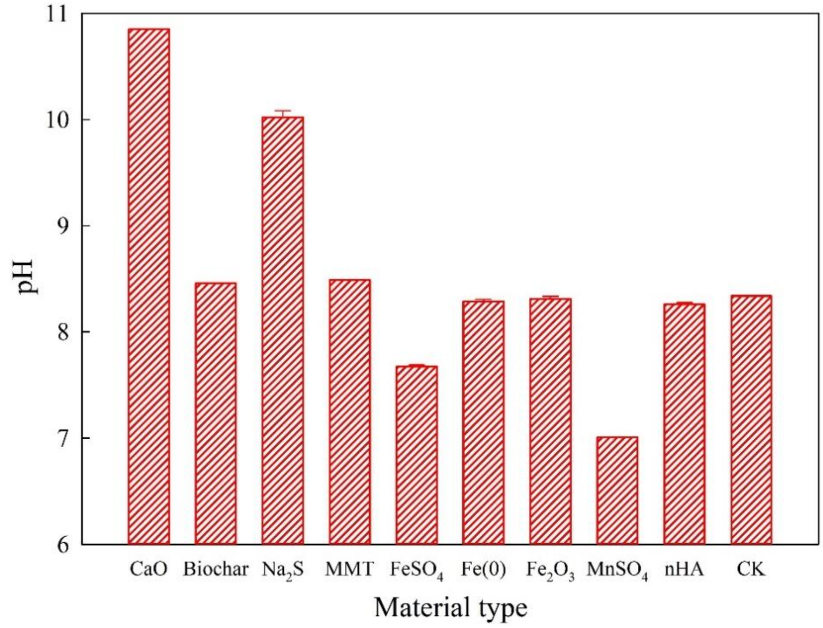

Fig. S1 Soil pH after the addition of different materials for 7 days

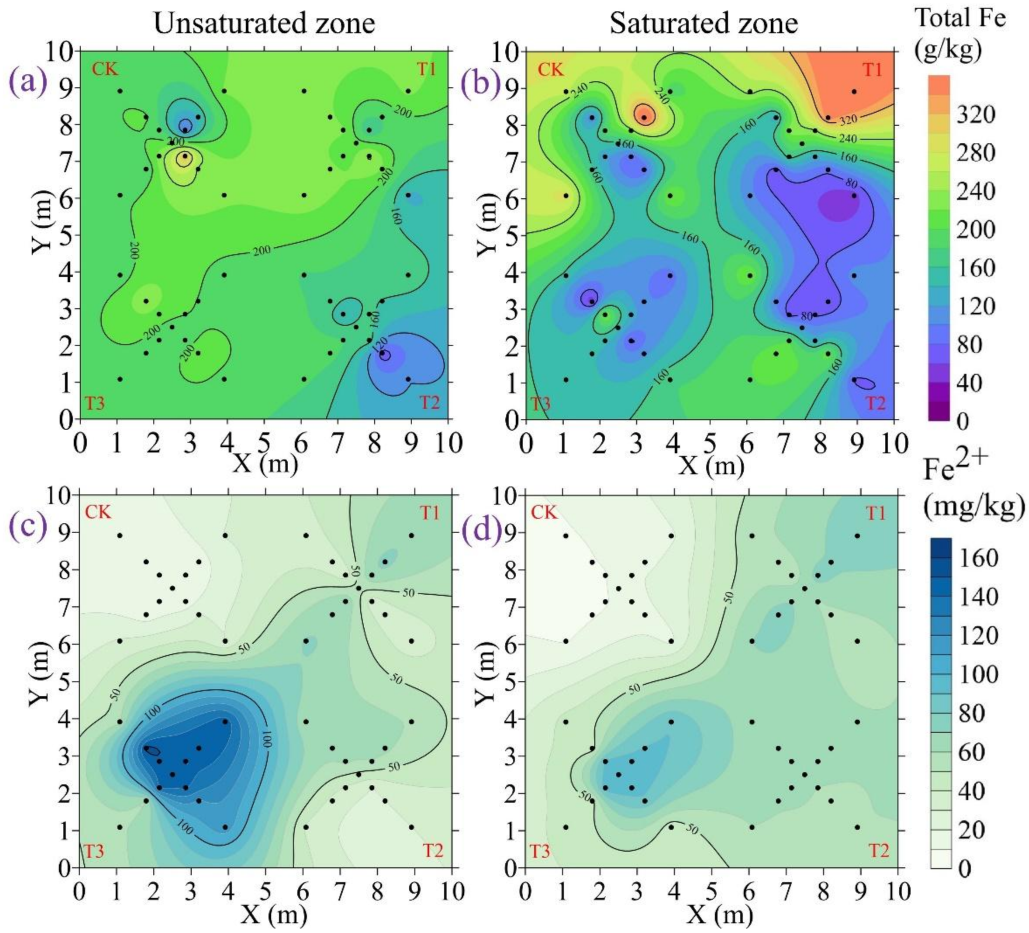

Fig. S2 The distribution of total Fe and Fe^{2+} concentrations after stabilization. (a) Total Fe concentration in the unsaturated zone; (b) Total Fe concentration in the saturated zone; (c) Fe^{2+} concentration in the unsaturated zone; (d) Fe^{2+} concentration in the saturated zone.

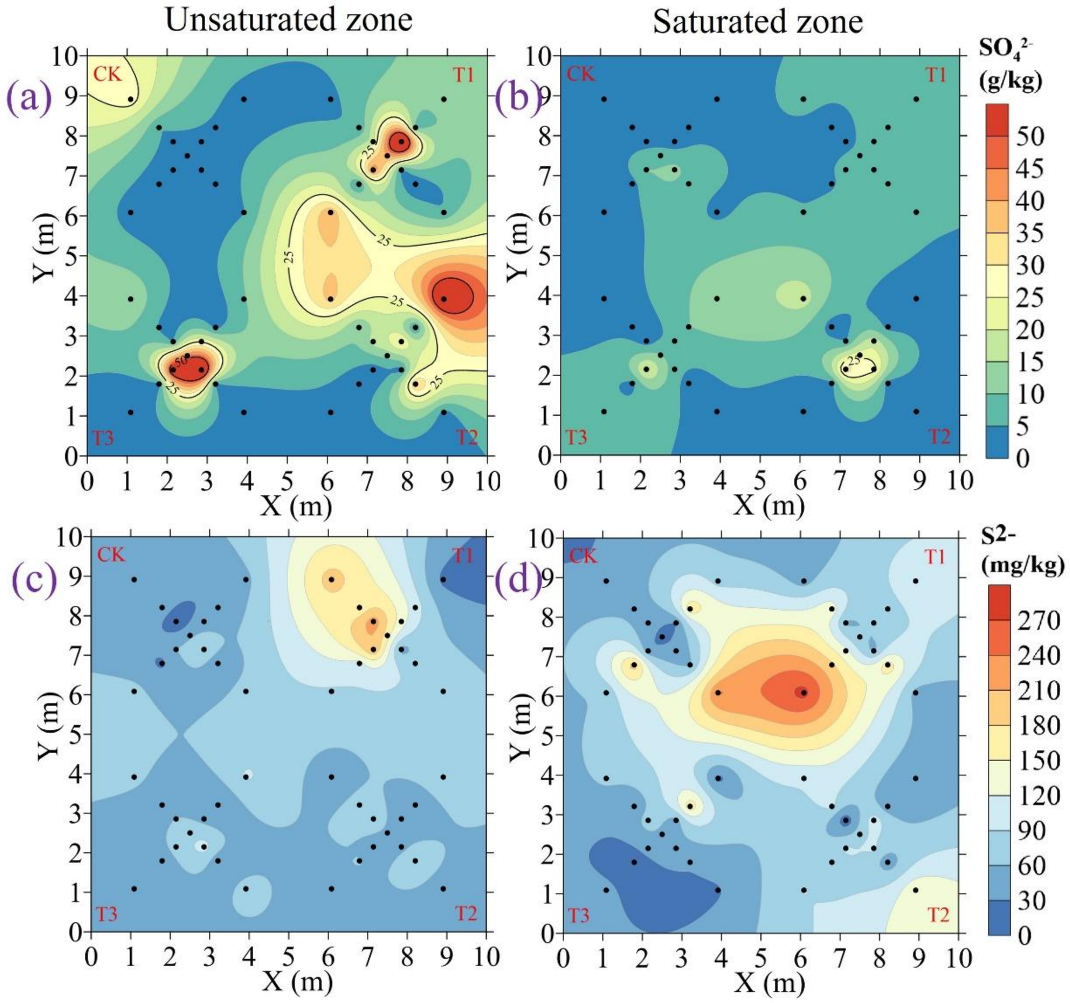

Fig. S3 The distribution of SO_4^{2-} and S^{2-} concentrations in soil after stabilization. (a) SO_4^{2-} concentration in the unsaturated zone; (b) SO_4^{2-} concentration in the saturated zone; (c) S^{2-} concentration in the unsaturated zone; (d) S^{2-} concentration in the saturated zone.

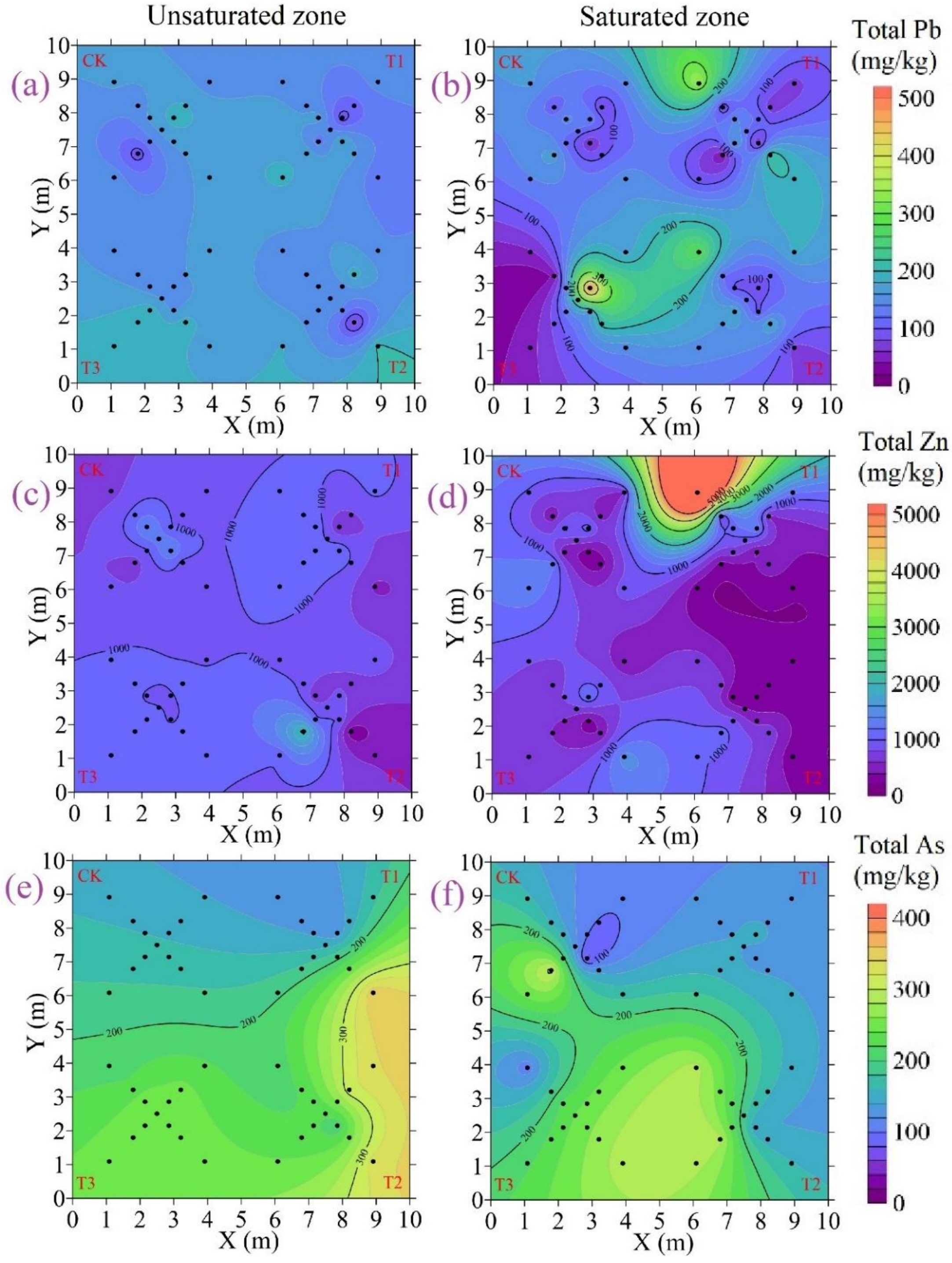

Fig. S4 The distribution of total Pb, Zn and As concentrations after stabilization. (a) Total Pb concentration in the unsaturated zone; (b) Total Pb concentration in the saturated zone; (c) Total Zn concentration in the unsaturated zone; (d) Total Zn concentration in the saturated zone; (e) Total As concentration in the unsaturated zone; (f) Total As concentration in the saturated zone.

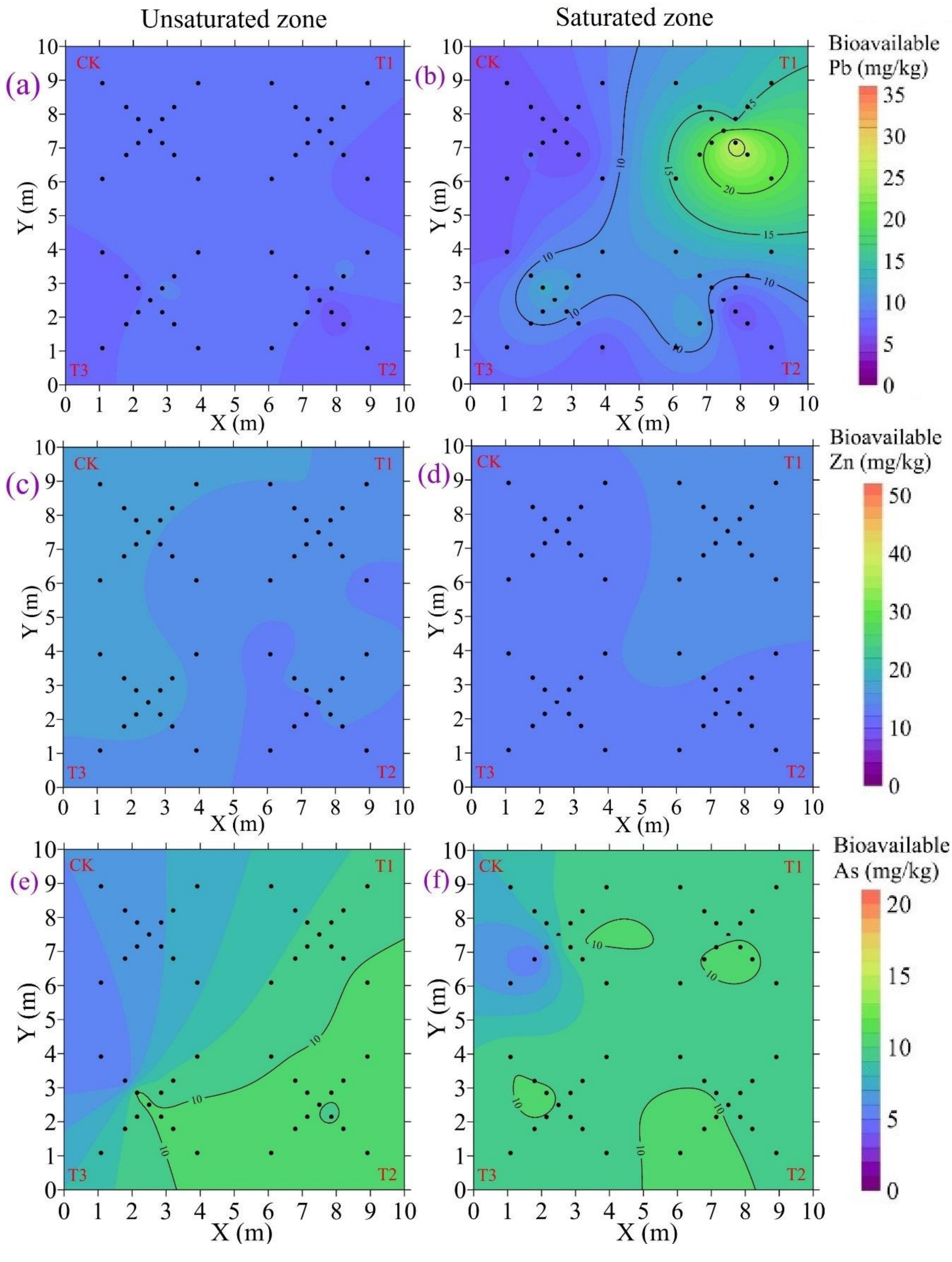

Fig. S5 The distribution of bioavailable Pb, Zn and As concentrations in soil before stabilization. (a) Bioavailable Pb concentration in the unsaturated zone; (b) Bioavailable Pb concentration in the saturated zone; (c) Bioavailable Zn concentration in the unsaturated zone; (d) Bioavailable Zn concentration in the saturated zone; (e) Bioavailable As concentration in the unsaturated zone; (f) Bioavailable As concentration in the saturated zone.

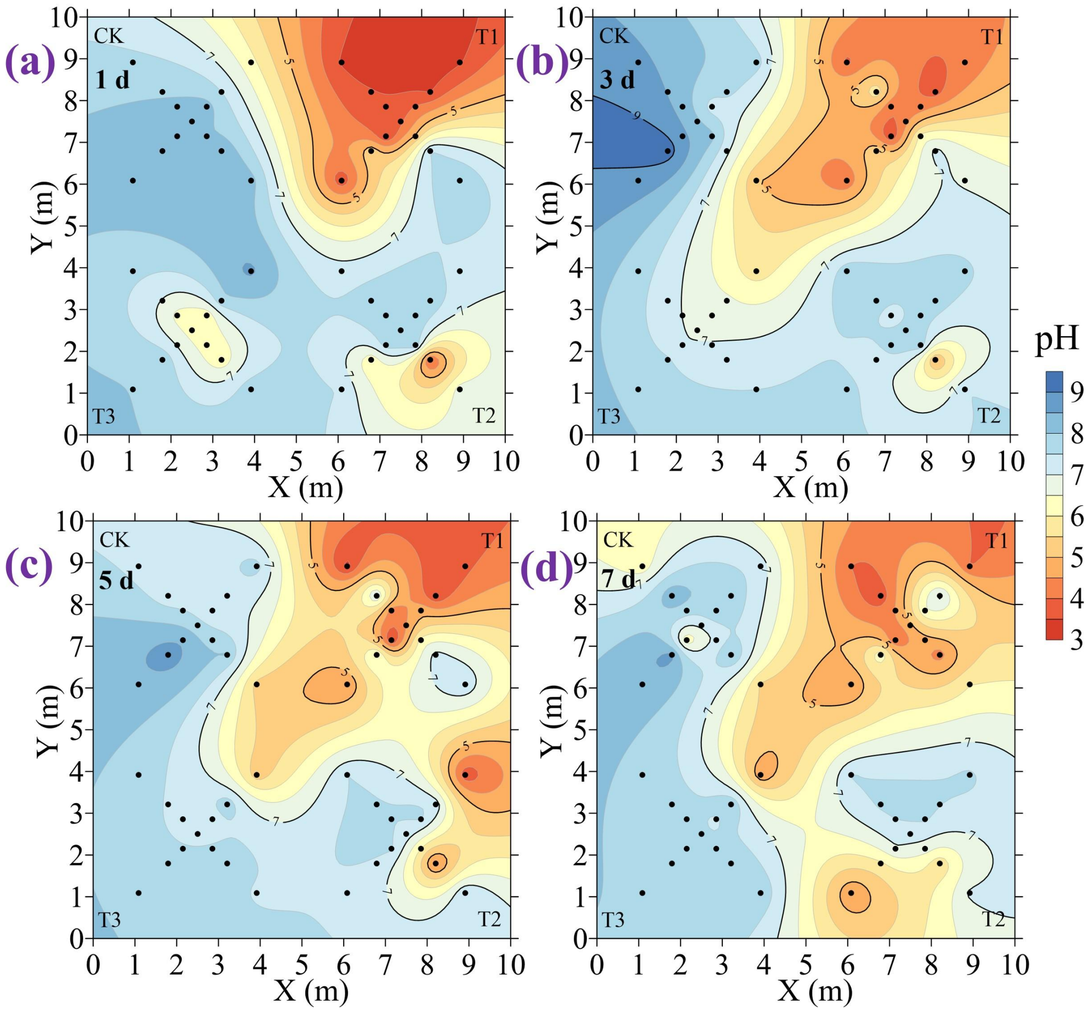

Fig. S6 The distribution of groundwater pH after the injection of $FeSO_4$ solution for 1 day (a), 3 days (b), 5 days (c) and 7 days (d) respectively.

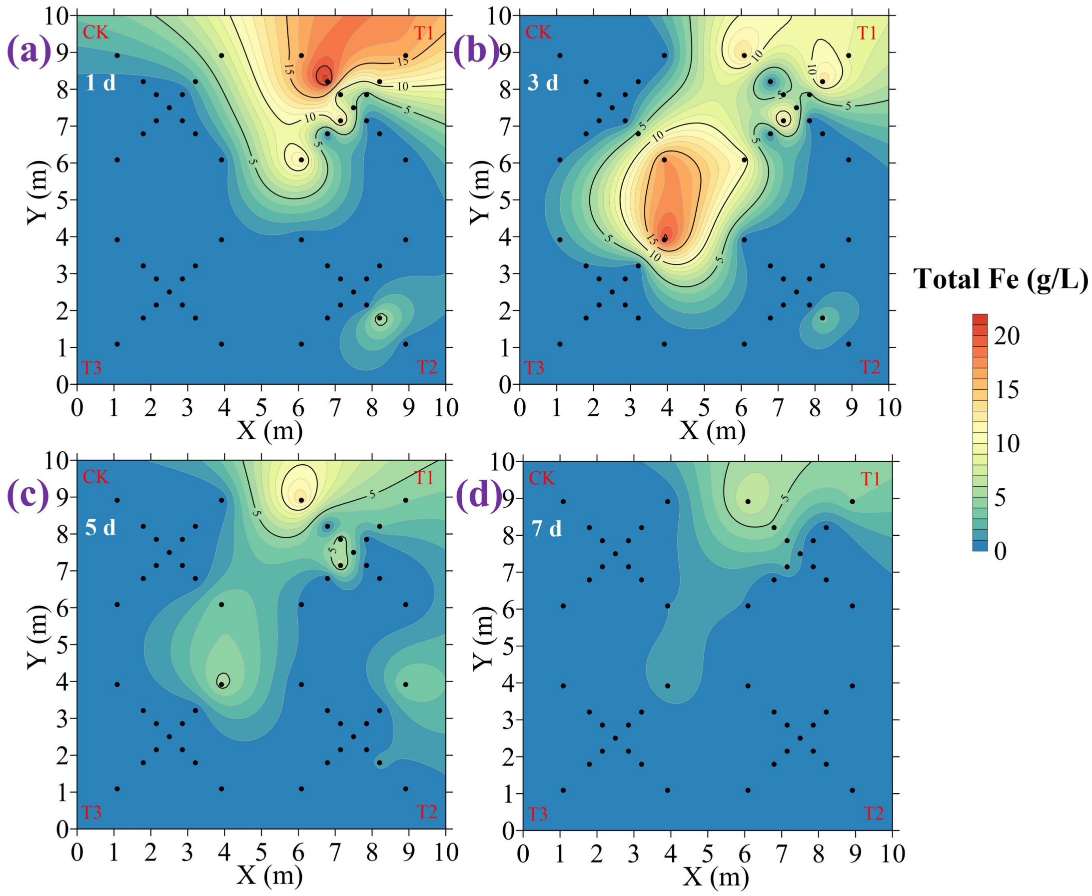

Fig. S7 The distribution of total Fe concentration in groundwater after the injection of $FeSO_4$ solution for 1 day (a), 3 days (b), 5 days (c) and 7 days (d) respectively.

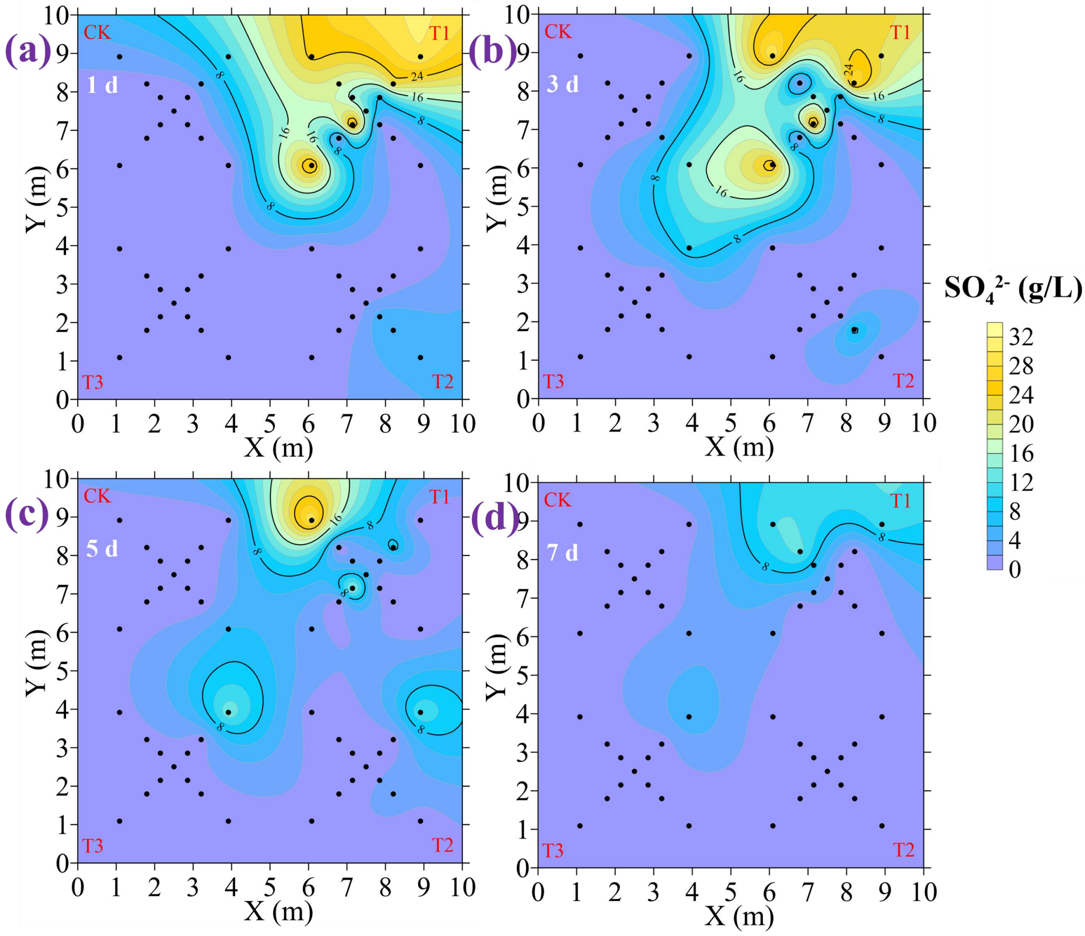

Fig. S8 The distribution of SO_4^{2-} concentration after the injection of $FeSO_4$ solution for 1 day (a), 3 days (b), 5 days (c) and 7 days (d) respectively.

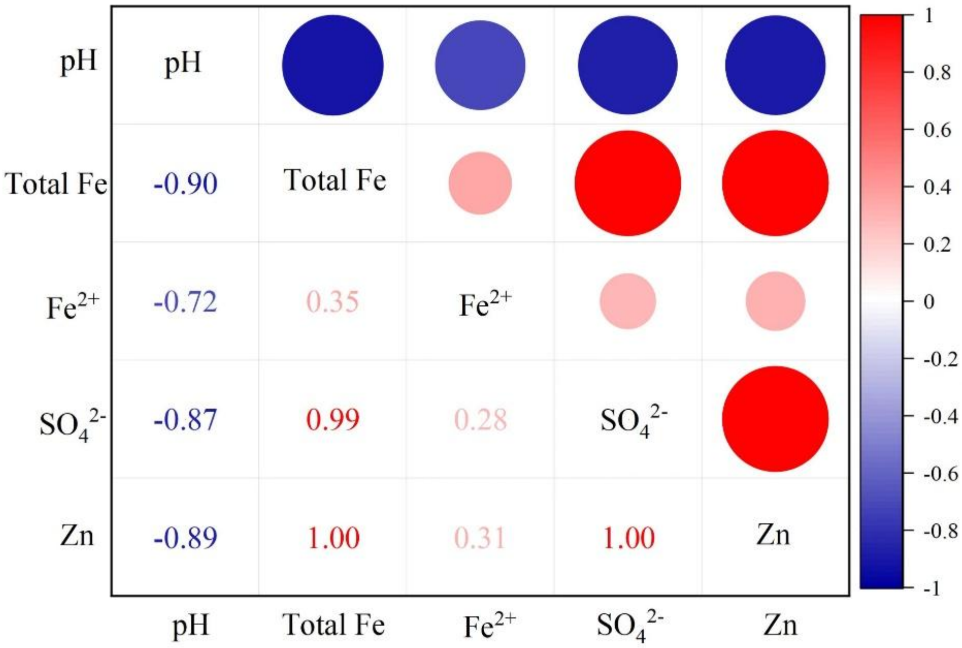

Fig. S9 Matrix of correlation coefficient among different parameters for groundwater

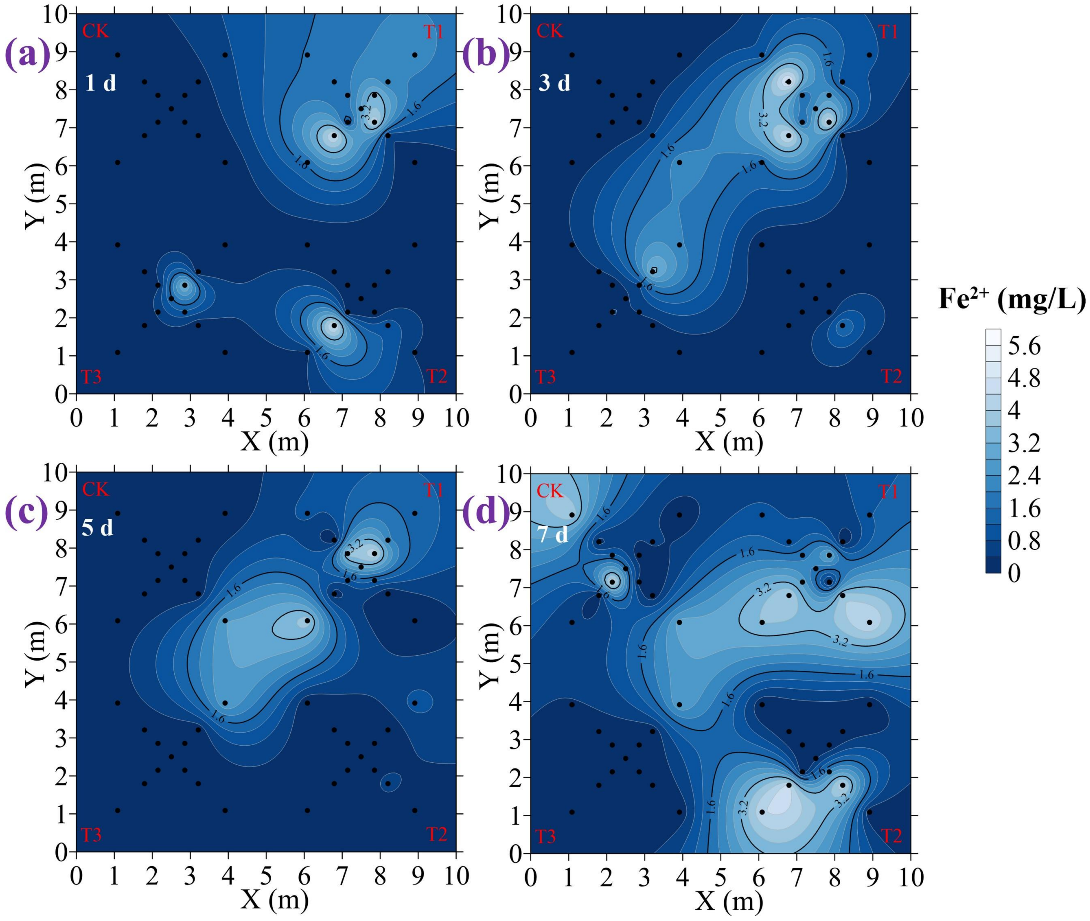

Fig. S10 The distribution of Fe^{2+} concentration in groundwater after the injection of $FeSO_4$ solution for 1 day (a), 3 days (b), 5 days (c) and 7 days (d) respectively.

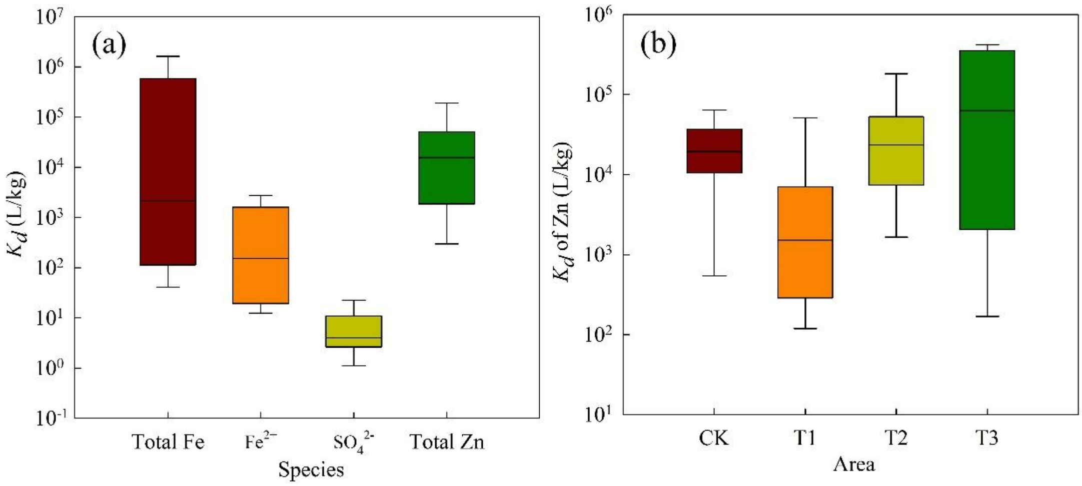

Fig. S11 The water-solid distribution coefficient (K_d) in the contaminated site after stabilization for 7 days. (a) K_d for different species; (b) K_d of total Zn in different areas.

Text S1 The calculation of solution radius of influence

Assuming that the thickness of unconfined aquifer was 1.0 m and the porosity was 0.3, The solution radius of influence could be calculated by the following equation:

$$V = \pi R^2 H\phi \rightarrow R = \sqrt{\frac{V}{\pi H\phi}} = \sqrt{\frac{0.8m^3}{3.14 \times 1.0m \times 0.3}} = 0.921m \quad \text{(Eq. S1)}$$

Text S2 The calculation of theorical concentration of Fe^{2+} and SO_4^{2-} in soil after injection of $FeSO_4$ solution

It was assumed that soil porosity (φ) was 0.3 and the density of soil particles (ρ_p) was 2.55 × 10^3 kg/m^3. If 1 L $FeSO_4$ solution was injected into dry soil, the volume of soil that was filled by the $FeSO_4$ solution could be calculated by the following equation:

$$V_s = \frac{V_l}{\varphi} = \frac{1L}{0.3} = 3.33L \quad \text{(Eq. S2)}$$

Soil weight in such a volume could be calculated by the following equation:

$$M_s = V_s(1-\varphi)\rho_p = 3.33L \times (1-0.3) \times 2.55kg/L = 5.94kg \quad \text{(Eq. S3)}$$

Therefore, the theoretical increased Fe^{2+} and SO_4^{2-} concentration in T1 could be calculated by the following equations:

$$\Delta C(Fe^{2+})_s = \frac{C_l V_l M_{Fe}}{M_s} = \frac{0.27mol/L \times 1L \times 56g/mol}{5.94kg} = 2.55g/kg \quad \text{(Eq. S4)}$$

$$\Delta C(SO_4^{2-})_s = \frac{C_l V_l M_{SO_4^{2-}}}{M_s} = \frac{0.27mol/L \times 1L \times 96g/mol}{5.94kg} = 4.36g/kg \quad \text{(Eq. S5)}$$

The theoretical increased Fe^{2+} and SO_4^{2-} concentration in T2 could be calculated by the following equations:

$$\Delta C(Fe^{2+})_s = \frac{C_l V_l M_{Fe}}{M_s} = \frac{0.52mol/L \times 1L \times 56g/mol}{5.94kg} = 4.90g/kg \quad \text{(Eq. S6)}$$

$$\Delta C(SO_4^{2-})_s = \frac{C_l V_l M_{SO_4^{2-}}}{M_s} = \frac{0.52mol/L \times 1L \times 96g/mol}{5.94kg} = 8.40g/kg \quad \text{(Eq. S7)}$$

The theoretical increased Fe^{2+} and SO_4^{2-} concentration in T3 could be calculated by the following equations:

$$\Delta C(Fe^{2+})_s = \frac{C_l V_l M_{Fe}}{M_s} = \frac{0.90mol/L \times 1L \times 56g/mol}{5.94kg} = 8.48g/kg \quad \text{(Eq. S8)}$$

$$\Delta C(SO_4^{2-})_s = \frac{C_l V_l M_{SO_4^{2-}}}{M_s} = \frac{0.90mol/L \times 1L \times 96g/mol}{5.94kg} = 14.55g/kg \quad \text{(Eq. S9)}$$