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Arsenic distribution and partitioning in multiple media in a typical catchment in the Qinghai-Tibetan plateau: A comparison between freshwater and saltwater lakes

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ABSTRACT

Arsenic (As) has been widely detected in surface media on the Qinghai-Tibetan Plateau (QTP); however, the differences in the As distribution and partitioning characteristics between freshwater and saltwater lakes remain poorly understood. To determine the distribution and partitioning characteristics of As, multimedia environmental samples were collected from a typical small watershed consisting of a river, wetland, and both freshwater and saltwater lakes on the QTP. Results showed that freshwater systems, represented by Hurleg Lake, were high in particulate arsenic (PAs) and low in dissolved arsenic (DAs), whereas the saltwater system represented by Tosen Lake, exhibited the reverse distribution. This discrepancy in As distribution was primarily attributed to evaporation enrichment, competitive adsorption of HCO_3^- and pH variations, as suggested by correlation analysis and stable isotopic composition of water. In the stratified Tosen Lake, an increasing trend of DAs in the water column was observed, potentially driven by the reductive dissolution of Fe (hydr)oxides and bacterial sulfate reduction in the anoxic bottom hypolimnion. Conversely, Hurleg Lake maintained oxic conditions with stable DAs concentrations. Notably, PAs was elevated in the bottom layer of both lakes, possibly due to uptake/adsorption by biogenic particles, as indicated by high levels of chl.α and suspended particulate matter. These findings offer insights into the potential future impact of climate change on As mobilization/redistribution in arid plateau lakes, with implications for management policies that regulate As pollution.

1. Introduction

Arsenic (As), a severely toxic and carcinogenic element, is widely found in various environments. As contamination has been one of the most globally ubiquitous and complex issues over the past decades (Gorny et al., 2015). The distribution of As in aquatic systems is highly heterogeneous, influenced by both natural geological processes and anthropogenic activities (Rosso et al., 2013). Under natural conditions, baseline dissolved As (DAs) in in riverine and lacustrine systems are typically low, ranging between 0.1 and 2 μg/L. However, geogenic sources, such as As-rich bedrock and geothermal discharges, can elevate As levels in certain locales (Smedley and Kinniburgh, 2002). The partitioning of As across water, suspended particulate matter (SPM), and sediments is dynamic, shaped by a suite of environmental drivers including redox conditions (Guo et al., 2011, 2014), pH (Gankhurel et al., 2022; Zhang et al., 2022), salinity (Hong et al., 2018; Jia et al., 2017), and microbial activity (Guo et al., 2016). These factors govern the adsorption of DAs onto SPM and its subsequent sedimentation (Barrett et al., 2018; Ying et al., 2017; Zhang et al., 2022), necessitating a nuanced understanding of the mobilization and partitioning of As in multimedia is of great importance.

Lacustrine systems, pivotal to global aquatic ecology, are typically one of the major As sinks. The mechanisms dictating As distribution and mobility within lakes are complex and site-specific. For instance, thermal stratification, prevalent in temperate lakes, can induce anoxic conditions in deeper waters, favoring reductive As release from SPM

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sediments by the dissolution of Fe/Mn (hydr)oxides (Barrett et al., 2018; Ding et al., 2022). In semiarid or arid areas, intense evaporation can lead to the enrichment of DAs in closed lakes (Jia et al., 2017, 2021; Oremland et al., 2004), where concurrent increases in salinity and pH (Furian et al., 2013) can further affect the release of As. Gankhurel et al. (2022) compared the measured DAs with those predicted by a geochemical model, and suggested that As in lake water could be eliminated in the form of SPM or sediment during evaporation (Gankhurel et al., 2022). Desorption from the mineral surfaces can also enhance As release from sediments or SPM at higher pH or anion concentrations, therefore showing a decreasing distribution coefficient (K_d) for As (Che et al., 2020; Zhang et al., 2022). Iron (Fe) and manganese (Mn), being significant components of fine particulate matter and having high adsorptive surface areas, are likely to retain As under particular pH conditions (Kanematsu et al., 2013; Yuan et al., 2021). Correspondingly, the reductive dissolution of Fe/Mn (hydr)oxides has been considered the crucial controlling process triggering As release under suboxic or anoxic environments (Ding et al., 2022; Jia et al., 2021; Guo et al., 2011). While considerable research has been conducted on As mobilization in lakes, studies on multimedia partitioning patterns of As are still lacking (Gu et al., 2020). Moreover, the influencing factors are site-specific (Hong et al., 2018). Therefore, site-specific field studies on the multimedia partitioning of As are necessary.

The lakes in the Qinghai-Tibetan Plateau (QTP) account for more than half of the total lake area in China (Zhang et al., 2020), which is a critical component of the Asian water tower (Immerzeel et al., 2010). Owing to the high altitude, dry climate, and extensive exposure to solar radiation, approximately 92% of lakes in this region are saline or hypersaline (Song et al., 2018; Zhang et al., 2020). Meanwhile, widespread As-enriched bedrock and geothermal activities supply As to the hydrosphere (Li et al., 2013). Notably, significant variations in As contents have been found in various lakes in the QTP, ranging from 1.80 μg/L in a freshwater lake (Mokiu Co) to an exceptional 10,775 μg/L in a saline lake (Tangyung Co) (Xiong et al., 2018). Most lakes have been reported to have high As concentrations—for example, the dissolved As concentrations are 60 μg/L in Bam Co, 306 μg/L in Tuksitugar, 504 μg/L in Dawa, and 1200 μg/L in Dajiacuo (Xiong et al., 2018).

Salinity's influence on arsenic mobilization has been widely studied in marine environments and groundwater (Ayers et al., 2016; Gu et al., 2020; Hong et al., 2018; Jia et al., 2017, 2021; Yuan et al., 2021). However, the effect of salinity on the mobilization and partitioning of As differs in specific environments. For example, research conducted in the Yongsan Estuary observed that a greater concentration of salt in water resulted in less adsorption of As into SPM and sediment; thus, the field-based K_d for As decreased with a higher salinity (Hong et al., 2018). However, a study in the Taehwa River Estuary did not show a significant difference in K_d between freshwater and saltwater (Hong et al., 2016). Moreover, Yuan et al. (2021) observed that DAs decreased with increasing salinity during autumn but showed no significant correlations during summer in the adjacent area of the Changjiang Estuary. Hence, several uncertainties exist in the effect of salinity on As mobilization and partitioning pattern due to the complexity within aquatic ecosystems. Despite research efforts to understand As enrichment in alkaline saltwater lakes (Che et al., 2020; Xiong et al., 2018), the behaviors of As in arid plateau saltwater lakes remain poorly understood (Li et al., 2013), especially how environmental factors, such as salinity, control the mobilization and partitioning pattern of As between freshwater and saltwater lakes.

The purpose of this study was to determine the distribution and partitioning behavior of As across water, SPM, and sediment, with a focus on comparing freshwater and saltwater lakes. The main objectives were to 1) characterize As contents and distribution in both lake types; 2) compare solid-liquid distribution coefficients between SPM-water and sediment-water phases across lakes; and 3) identify key factors influencing As distribution across media. The results significantly improve understanding of As fate and cycling in lakes within arid plateaus.

2. Materials and methods

2.1. Study area

The Hurleg Lake-Tosen Lake Nature Reserve is located in the northeastern Qaidam Basin, QTP (Yang et al., 2021). The reserve contains a connected freshwater lake and a saltwater lake, which are typical representatives of lakes in the QTP. Hurleg Lake is a freshwater lake, while Tosen Lake is a saltwater lake. The Bayin River flows through Delingha City and Hurleg Lake and finally discharges to Tosen Lake. The region has a typical alpine continental climate, with a yearly average temperature of 1–5 °C (Fan et al., 2012). The average precipitation is only 176.3 mm, and 56–84% of the rain falls during the summer. Compared to precipitation, annual evaporation is up to 2000–2500 mm, approximately 12–17 times the average yearly precipitation (Cui and Li, 2014; Xiong et al., 2018). In addition, relatively high DAs was found by Wang et al. (2018) in Tosen Lake. The specific geographical and climatic conditions and the high As accumulation previously found provide a natural site to investigate the behavioral differences of As in freshwater and saltwater environments.

2.2. Sampling and sample preparation

To investigate the As distribution in the study area, 48 surface water sampling sites were established along the Bayin River, within the wetland, and across the two lakes (see Fig. 1). Profile water samples were also collected in Hurleg Lake (K-11) and Tosen Lake (T-14) to examine the differences in As behaviors between freshwater and saltwater lakes. Water, SPM, and surface sediment samples were simultaneously collected in May 2021.

Surface water samples (0.5 m beneath the surface) and profile water samples were collected with a 10 L acid-washed plastic bucket and filtered with a polyurethane fiber filter membrane (0.45 μm) for the analysis of ions (NH_4^+, Cl^-, SO_4^{2-}, Na+, Mg^{2+}, Ca^{2+}) and water isotopes. Water samples for the analysis of As, Fe, Mn, and Al concentrations were rapidly acidified to a $pH < 2$ and refrigerated at 4 °C. Surface sediment samples were also collected using a Van Veen grab (JC-W801N, China). To obtain the suspended particulate matter (SPM), 5 L of water was filtered through a 0.45 μm polyurethane fiber filter membrane. Afterward, the filtered SPM and sediment samples were freeze-dried in a vacuum freeze dryer (Labconco FreeZone, USA) until they reached a constant weight for the following analysis.

2.3. Analytical methods

The dissolved oxygen (DO), temperature (T), oxidation-reduction potential (ORP), chlorophyll *a* (Chl.α), salinity, and pH were simultaneously measured using a multiparameter water quality sonde (EXO-3, YSI, USA). Ammonium (NH_4^+) was determined using a continuous flow analyzer (Skalar, San++, Netherlands) with a detection limit of 0.02 mg/L. The alkalinity was determined by titrating with a 0.02 mol/L hydrochloric acid concentration within 8 h after sampling. The concentrations of As, Fe, Mn, and Al in water samples were measured by inductively coupled plasma-mass spectrometry (ICP-MS 8900, Agilent, USA). The concentrations of major ions (Cl^-, SO_4^{2-}, Na^+, Mg^{2+}, Ca^{2+}) in water were measured using an ion chromatograph (ICS-1100, Thermo Scientific Dionex, USA) with a detection limit of 0.01 mg/L. IC Standards (Thermo Scientific Dionex, USA) were used as the spiking solution for quality assurance. The standard deviation for spiking solution samples and 10% parallel samples was below 5%.

The SPM and sediment samples were digested with mix-acid by a high-throughput Microwave Digestion System (MARS 6, CEM, USA) (Xuecheng Zhang et al., 2022). In brief, freeze-dried sediment samples were ground and sieved to <0.15 mm then 50 mg of sediment were

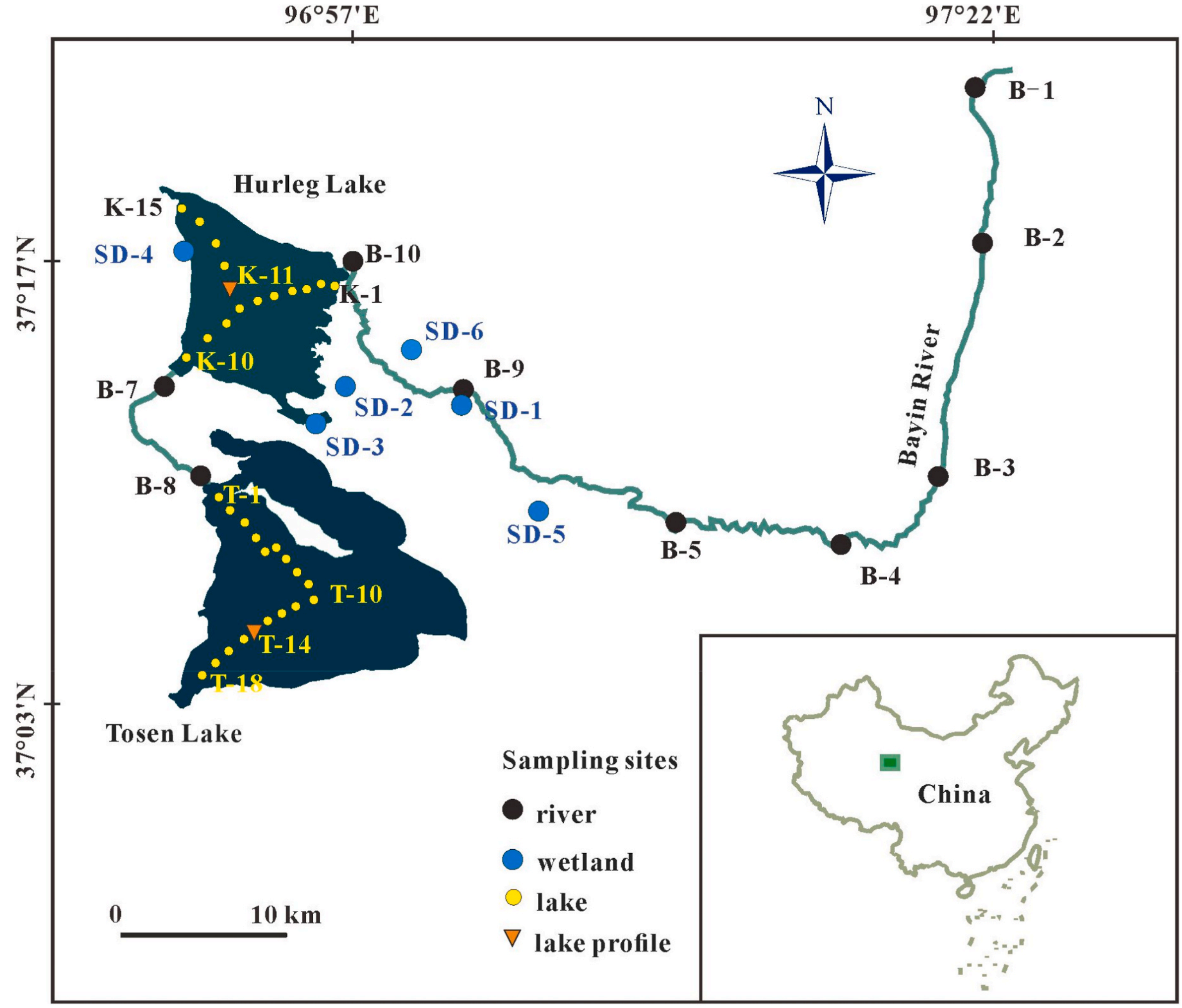


Fig. 1. Location of the study area and sampling sites.

digested by HNO_3, HCl, and HF (HNO_3: HCl: HF = 6: 2: 2) in Telfon vessels in a closed system at 140 °C for 1 h. The digested samples were dried to evaporate acid and then diluted with 3% HNO_3 to measure As, Fe, Mn, and Al by inductively coupled plasma-mass spectrometry (ICP-MS 8900, Agilent, USA). Quality control was performed using the standard reference sediment material (GBW-07309). The recovery of As was 106.2% of the certified value, which was deemed acceptable. The standard deviation for 10% parallel extraction and measurement samples was below 5%. For particle size analysis, weighed about 0.15 g sediment sample in a clean glass baker, then washed the sample with ultrapure water to a pH of 7.0 after adding 10 mL 30% H_2O_2 and 10 mL HCl to remove organic matter. Sediment particle size analysis was conducted using a laser particle size analyzer (Mastersizer 3000, Malvern, UK) with a measured range of 0.02–2000 μm. The particles were categorized into three size ranges: <4 μm (clay), 4–63 μm (silt), and >63 μm (sand) (Gu et al., 2020; Wang et al., 2017).

The stable water isotopes were measured by a wavelength-scanned cavity ringdown spectroscopy (WS-CRDS) instrument (L2140-i, Picarro, USA). Notation is expressed in terms of δ (‰) relative to the Vienna Standard Mean Ocean Water (VSMOW). The precision of the measurements is ±0.4‰ for δD and ±0.05‰ for $\delta^{18}O$.

2.4. Calculations

The distribution coefficient (K_d) of As between dissolved (<0.45 μm) and particulate (>0.45 μm) phases has been widely used to understand the geochemical behaviors of pollutants (Gu et al., 2020; Yuan et al., 2021). In this study, the K_d for As was calculated as follows:

$$K_d = \frac{C_S}{C_w} \tag{1}$$

where C_S is the As concentration in the sediment/SPM (mg/kg), C_w is the As concentration in water (μg/L), and K_d is the liquid-solid distribution coefficient (10^3 L/kg) in this formula.

The relative water column stability (RWCS) is a measure of the vertical stability of the water column. It quantifies the resistance of the water column to vertical mixing and the potential for thermal stratification. It can be calculated using Equation (2) (Becker et al., 2008; Cui et al., 2021):

$$RWCS = \frac{\rho_b - \rho_w}{\rho_4 - \rho_5} \tag{2}$$

where ρ_b is the bottom water density (g/cm^3) and ρ_w is the water density at a specific depth (g/cm^3), ρ_5 and ρ_4 represent the pure water density (g/cm^3) at 5 and 4 °C, respectively. The water density can be calculated from the following equation (3) (Lawson and Anderson, 2007):

$$\rho T = 1000 \times \left[1 - \frac{T + 288.9414}{508929.2 \times (T + 68.12196)} \times (T - 3.9862)^2\right] \tag{3}$$

2.5. Statistical analysis

Unless otherwise noted, figures in this study were plotted using Origin version 8.5 software. The saturation index (SI) was calculated using the PHREEQC version 3.3 software. The relationship between As concentrations/contents and environmental factors was analyzed using Pearson's correlation coefficient. Redundancy analysis (RDA) was performed using Canoco version 4.5 software (ter Braak and Smilauer, 2002) to explore the influences of environmental factors on As distribution.

3. Results and discussion

3.1. General properties of water and sediments

Among the main parameters for surface water (Table S1), the pH and DO ranged from 8.13 to 9.01 and 6.71–10.22 mg/L, respectively, which indicated a weak alkaline oxic environment. Tosen Lake exhibited elevated salinity (17.12‰) and pH (8.91), which were markedly distinct ($p < 0.001$) from other water bodies in the study (Table S1). The stable isotopes of hydrogen (δD) and oxygen ($\delta^{18}O$) are commonly used to identify the intensity of evaporation. In the present study, the values of $\delta^{18}O$ and δD transitioned from negative values in the Bayin River (−9.28‰ to −5.06‰, −59.79‰ to −36.88‰) and Hurleg Lake (−7.00‰ to −5.90‰, −48.64‰ to −42.89‰) to positive values in Tosen Lake (0.59‰–2.07‰, −5.06‰–3.16‰) along the flowing direction. As shown in Fig. 2a, all samples were located around or deviated to the right of the local meteoric water line (LMWL), indicating their meteoric origin and the effect of evaporation enrichment. Especially samples from Tosen Lake, their values deviated far to the right of the LMWL, suggesting a strong influence of evaporation enrichment in Tosen Lake. In arid or semi-arid regions, evaporation enrichment was shown to contribute to water salinization and the release and mobilization of As (Gu et al., 2020; Jia et al., 2021; Qing et al., 2023; Smedley and Kinniburgh, 2002; Zhang et al., 2022). Thus, the subsequent analysis will primarily focus on the influence of evaporation on the salinization process and the spatial distribution of As.

In the water profiles, T and DO decreased with water depth in Tosen Lake (Fig. 3ab), indicating that relatively strong thermal and DO stratification occurred concurrently. Conversely, Hurleg Lake with a maximum depth of 8 m, exhibited only slight thermal stratification and no DO stratification in the water profile (Fig. 3ab). Thermal stratification could reduce particle movement and sediment resuspension, resulting in the accumulation of substances in the bottom waters (Karageorgis et al., 2003; Song et al., 2013). Furthermore, DO stratification formed an anaerobic environment in the hypolimnion, which favored the reductive reaction. The observed differences in stratification between the two lakes might account for the disparate distribution patterns of As across various media.

In terms of the grain-size composition of the surface sediment (Fig. S2), approximately 48.7% of the samples were found to consist of sandy mud (sM), followed by sandy silt (sZ) with 20.5%, silty sand (zS), and sandy clay (sC) with 17.9% and 10.2%, respectively. Except for a few locations where sand constituted over 50%, the majority of sites displayed silt content exceeding 50%, suggesting a potential association of As with finer sediment particles such as silt and clay.

3.2. Effect of evaporation on the horizontal distribution of arsenic in the surface environment

The average DAs in surface water varied spatially (Fig. S3a), with the highest value in Tosen Lake (40.88 ± 1.64 μg/L) exceeding the WHO guideline limit of As (10 μg/L) and the lowest value in Hurleg Lake (1.74 ± 0.24 μg/L). Interestingly, particulate arsenic (PAs) showed a different tendency (Fig. S3b); the lowest value was found in Tosen Lake (11.98 ± 3.89 mg/kg), and the highest value was found in wetlands (49.05 ± 59.03 mg/kg), followed by Hurleg Lake (48.59 ± 41.18 mg/kg). However, there was no significant difference in the content of arsenic in sediment (SAs) among the different aquatic systems, while Tosen Lake had slightly higher SAs than the others (Fig. S3c). Notably, high DAs at site SD-2 (40.84 μg/L) were consistent with high salinity (11.55 ‰), a condition potentially attributable to the intensive evaporation processes previously discussed.

The deuterium excess d ($d = \delta D - 8 \times \delta^{18}O$) is available to quantify the contribution of evaporation to salinity (Huang and Pang, 2012; Jia et al., 2017). d-excess remains constant during mineral dissolution and transpiration, decreasing during evaporation (Dansgaard, 1964). Globally, the average d-excess of precipitation is 10‰. When a water body undergoes evaporation, the d-excess will decrease while salinity will increase, exhibiting a negative correlation (Huang and Pang, 2012; Jia et al., 2017). Our analysis revealed a significant negative correlation between d-excess and log salinity ($R^2 = 0.62$, $p < 0.001$; Fig. 2b) and log DAs ($R^2 = 0.66$ $p < 0.001$; Fig. 2c), implicating evaporation enrichment as the important mechanism influencing both arsenic release and water salinization.

In Hurleg Lake, the d-excess values of samples were slightly less than 10‰, reflecting its dynamic water exchange, and mitigating the impact of evaporation on salinity and DAs. In stark contrast, those of Tosen Lake were far below the Line d-excess = 10‰. In addition, Tosen Lake's significantly lower d-excess values and its closed-basin nature suggested that evaporation was a key driver of both salinity and DAs increases. Concurrently, the elevated salinity in Tosen Lake promotes arsenic

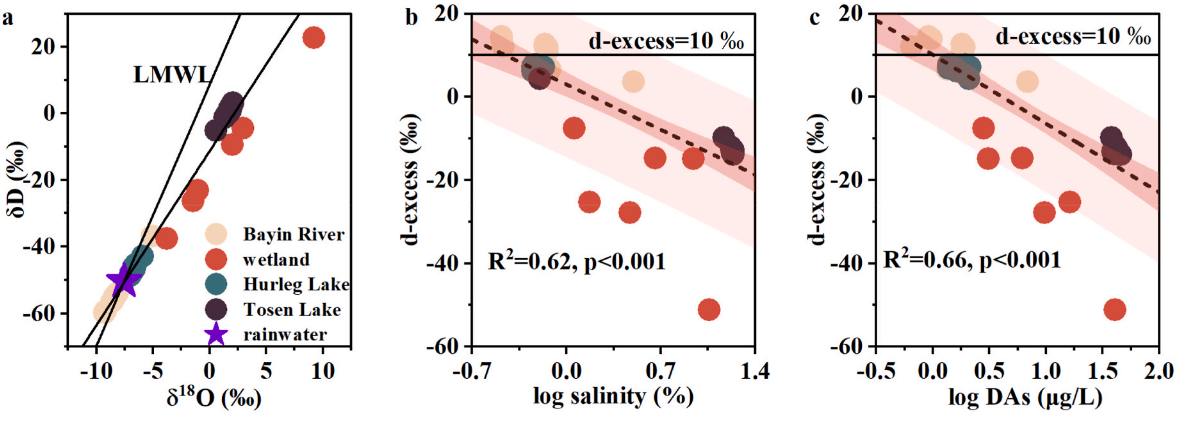


Fig. 2. Plot of δD vs. $\delta^{18}O$ (a) and d-excess vs. log salinity (b) and log DAs (c) in the watershed. Rainwater data and the local meteoric water line (LMWL: $\delta D = 7.83 \times \delta^{18}O + 8.07$) were obtained from reference (Zhu et al., 2015). $\delta D = 5.36 \times \delta^{18}O - 10.59$ (the study area). The lighter-shaded region represents the 95% predi10ction interval, and the darker-shaded area represents the 95% confidence interval.

desorption, likely by inducing competition for adsorption sites (Che et al., 2020; Guo et al., 2014).

According to the correlation heatmap and RDA, salinity was the primary factor in promoting arsenic release, followed by pH (Fig. 4ac). A similar controlling mechanism was responsible for high DAs in hypersaline soda lakes, such as Mono Lake and Searles Lake in California, USA (Kulp et al., 2007; Oremland et al., 2004). In the present study, DAs were positively correlated with various anions, while PAs showed the opposite correlations with those anions (including Cl^-, SO_4^{2-}, and HCO_3^-; Fig. 4a). These patterns suggested a competitive adsorption process, which was consistent with observations from Qinhai Lake (Cao et al., 2021) and Yamdork Lake (Che et al., 2020) in the same region on the QTP.

As shown in Fig. 4c, PFe, PMn, and PAl showed a simultaneous increase in PAs content according to the RDA results, suggesting that As was mainly bound to the surface of Fe/Mn/Al (hydr)oxides. PHREEQC calculation revealed that typical Fe (hydr)oxides, such as $Fe(OH)_3$(a), goethite, and hematite were oversaturated (average SI: 1.18, 6.97, and 15.94, respectively), whereas Fe carbonate/sulfate minerals and manganese minerals were undersaturated (Table S2). This evidence suggested the predominance of Fe (hydr)oxides in As sorption/desorption processes in the oxic conditions of the study area, where As(V) is expected to form inner-sphere complexes (Smedley and Kinniburgh, 2002). Under neutral to alkaline conditions, the competitive capacity of anions for the adsorption of As(V) by ferrihydrite was derived as $CO_3^{2-} > SO_4^{2-} > Cl^-$ (Frau et al., 2010; Kanematsu et al., 2013). Chloride (Frau et al., 2010) and a low concentration of sulfate (<0.05 M (Kinjo and Pratt, 1971);) were adsorbed as outer-sphere complexes that did not affect the inner-sphere complexes by As(V) at the mineral surface. In comparison, bicarbonate displayed a monodentate inner-sphere complex with minerals such as HFO and ferrihydrite (Frau et al., 2010; Villalobos and Leckie, 2001). Given that the concentration of SO_4^{2-} was lower than 0.5 mM in the catchment (Table S1), carbonate was the primary competitive anion influencing As release in Tosen Lake, corroborated by the significant positive correlation between DAs and alkalinity ($R^2 = 0.94$, $p < 0.001$; Fig. 6ac).

The correlation between pH and salinity (Fig. 4ac) aligned with previous works conducted on a regional scale or with limited samples (Fan et al., 2012; Furian et al., 2013; Liu et al., 2018). The region's waters exhibited a positive calcite residual (alkalinity > Ca^{2+} in millimoles of charge $mmol_c$, Table S1), suggesting an alkaline evaporation pathway. A similar geochemical process was common across all continents in arid and semiarid continental environments (Barbiero et al., 2008; Furian et al., 2013). This pH increase was crucial, alongside salinity, in promoting As release from SPM (Fig. 4c). This was supported by the phenomenon that increasing DAs coincided with increasing pH (Fig. 4ac) and the significant negative correlation between PAs and pH ($R^2 = -0.51$, $p < 0.001$; Fig. 4a). For instance, site T-16 in Tosen Lake recorded a pH of 8.85 and a PAs of 13.80 mg/kg; in comparison, site T-10 exhibited a of pH of 9.01with a reduced PAs of 8.58 mg/kg. As adsorption was pH-dependent, with adsorption efficacy diminishing above pH 9 for Al oxides and pH 7 for Fe (hydr)oxides, illite, and kaolinite (Richter et al., 2019). Similar pH-dependent As release has already been demonstrated in arid and semiarid inland alkaline lakes, such as Gobi Lake (Gankhurel et al., 2022) and Qinhai Lake (Cao et al., 2021), and groundwater in the Hetao Basin (Guo et al., 2011), Guide Basin (Xing et al., 2022), and Loess Plateau (Zhang et al., 2022).

3.3. *Effect of stratification on the vertical distribution of arsenic in water columns*

In Tosen Lake, depth profiles revealed increasing DAs and PAs with greater depths, inversely correlating with observed T and DO levels. Notably, the lake's hypolimnion, marked by anaerobic conditions, displayed significant arsenic accumulation. Contrastingly, Hurleg Lake's DAs remained stable vertically, while PAs exhibited a modest increase in the sub-bottom (Fig. 3cd).

In this study, we observed that stratification had an impact on shaping the vertical As distribution. Thermal stratification reduced the vertical movement and sinking rate of As, resulting in the accumulation of As in the bottom water, especially PAs. Specifically, higher concentrations of PAs in Hurleg Lake (6 m, 68.03 mg/kg) and Tosen Lake (22 m in the T-14 profile, 64.73 mg/kg), accompanied by high DAs (58.88 μg/L) were noticed in the bottom waters (Fig. 3cd). This was further corroborated by the increasing concentration of SPM in the hypolimnion (Fig. S1b). Similar results have been reported from polymictic shallow lakes (Fung et al., 2022), stratified urban ponds (Song et al., 2013), seasonally stratified reservoirs (Ding et al., 2022), and As-contaminated groundwater (Ying et al., 2017).

Furthermore, DO stratification influenced the vertical distribution of As in Tosen Lake as well. High DAs at the bottom waters were accompanied by elevated levels of reduced species like H_2S (>4 mg/L), DFe (>20 μg/L), and DMn (>10 μg/L), indicating that redox-driven processes thus appear to control As mobility (Ding et al., 2022; Guo et al., 2014; Ying et al., 2017). To identify the impact of DO stratification, the correlation relationship with RDA (Fig. 5) of the bottom (below 16 m) anoxic layer in Tosen Lake was conducted. Reductive dissolution of Fe (hydr)oxides corroborated by a positive correlation between DAs and DFe, emerged as an important mechanism for arsenic release in anoxic conditions., which was also the most accepted mechanism for high As in stratified lakes (Che et al., 2020; Ding et al., 2022) and groundwaters (Guo et al., 2011; Xing et al., 2022). Additionally, bacterial sulfate reduction (BSR) may play a significant role in As release under anoxia, with the reduction of Fe (hydr)oxides by sulfur intermediates from BSR potentially mobilizing bound arsenic when the available Fe(III) oxides have low solubility (Guo et al., 2016; Poulton et al., 2004). This was substantiated by the significant negative association between H_2S and DAs ($R^2 = -0.91$, $p < 0.05$, Fig. 5a) and DFe ($R^2 = -0.89$ $p < 0.05$, Fig. 5a), alongside the previously discussed low solubility of Fe (hydr) oxides in Section 3.2.

Interestingly, the negative correlation between PAs and PFe ($R^2 = -0.83$ $p < 0.05$, Fig. 5a) suggested that Fe (hydr)oxides were not the only host for As in the hypolimnion. Previous studies have noted the supplementation of sedimentary resuspension to the overlying water. However, in this study, the arsenic in the sediment (SAs) showed significant correlations with just a few physical properties (particle size) and sediment aluminum content (SAl) (Fig. 4bd), suggesting that As was probably scavenged by sediment fine particles, such as kaolinite ($Al_4(Si_4O_{10})(OH)_8$) or montmorillonite ($Al_2[Si_4O_{10}](OH)_2$). In contrast to sedimental As, particulate As was bonded to Fe (hydr)oxides, as delineated in Section 3.2. This distinction in compositional association implied that the input of resuspended sediment may not significantly contribute to the observed PAs increase in the benthic waters.

Instead, phytoplankton might accumulate As via adsorption or active uptake of As(V) as a byproduct of phosphate transport and dominate the increase in PAs (Yang et al., 2016; Azizur Rahman et al., 2012; Sanders and Riedel, 1998). This could be supported by the obvious rise in chl.α and C_{SPM} in the bottom layer of both lakes (Fig. S1bc). For instance, in Hurleg Lake, the bottom waters exhibited PAs, C_{SPM}, and chl.α of 27.75 mg/kg, 1.72 mg/kg, and 0.91 μg/L, respectively, significantly higher than surface water values (63.08 mg/kg, 3.07 mg/L, and 1.80 μg/L, respectively). Positive correlations between C_{SPM} and chl.α with PAs at the bottom of Tosen Lake further substantiate this relationship (Fig. 5ab). This pattern was consistent with observations in other stratified aquatic systems, such as the Skagerrak Seas, where particulate accumulation on/above the pycnocline was entirely biogenic, often comprising dinoflagellates (Karageorgis et al., 2003). Moreover, many previous studies have considered that a relatively high concentration of DAs could be taken up by marine macroalgae. In fact, the concentration of As in phytoplankton has been observed to be a magnitude higher than that of waterborne As (Barrett et al., 2018; Hong et al., 2016; Caumette et al., 2011, 2012).

3.4. Solid-liquid partitioning behavior of arsenic

The partitioning coefficients K_d values for As between water and solid phases in freshwater and saltwater were found to vary between regions. The average log K_d values for SPM-water were 4.34 ± 0.29, 3.59 ± 0.59, 4.36 ± 0.27, and 2.45 ± 0.13 L/kg in the Bayin River, wetland, Hurleg Lake, and Tosen Lake, respectively. The log K_d values for SPM-water in Hurleg Lake are relevant to those in other freshwater environments, such as Taihu Lake (4.3 L/kg) and the Diaojiang River (3.9 L/kg). However, those values in Tosen Lake are much lower than those in other saltwater environments, such as the Taehwa River Estuary (3.40 L/kg) and Changjiang Estuary (8.7 L/kg). The log K_d values for sediment-water were 3.77 ± 0.31, 2.85 ± 0.54, 3.77 ± 0.15, and 2.42 ± 0.25 L/kg in Bayin River, wetland, Hurleg Lake, and Tosen Lake, respectively (Fig. S5). Obviously, the log K_d values of SPM-water were higher than those of sediment-water due to the large specific surface areas of SPM facilitating As adsorption. Moreover, the K_d values for As were significantly higher in freshwater than in saltwater, corroborating previous studies in the eastern Hainan Estuary, Huanghe River Estuary in China, and Yongsan River Estuary in Korea (Balzer et al., 2013; Hong et al., 2016; Wang et al., 2017).

Several previous studies suggested that salinity (Hong et al., 2018; Wang et al., 2017; Balzer et al., 2013; Huang and Pang, 2012), pH (Yang et al., 2012), and SPM concentrations (Hong et al., 2016; Yang et al., 2016) were the dominant factors affecting the K_d value for As. As discussed in Section 3.2, increasing salinity and pH correlated with reduced As adsorption, as high HCO_3^- levels compete with As for binding sites on Fe (hydr)oxides, and elevated pH favored the release of As from SPM to water. This relationship was underscored by the significant negative correlation between log K_d and salinity ($R^2 = 0.91$, $p < 0.001$, Fig. S6a), and a significant negative correlation with pH ($R^2 = 0.54$, $p < 0.001$, Fig. S6b), emphasizing the sensitivity of As partitioning to these parameters. However, the “particle concentration effect” was not observed in the present study ($p > 0.05$, Fig. 4a). In contrast, PAs exhibited an increasing trend with C_{SPM} in the profiles (Fig. 3d, Fig.S1b), potentially due to the uptake/adsorption by biogenetic particles in deeper waters, as discussed in Section 3.3.

3.5. Arsenic mobilization in QTP lakes within the context of ongoing climate change

Climate change, manifesting in temperature and precipitation shifts, directly impacts hydrological dynamics within a lake basin, especially in the QTP (Liu et al., 2019). Changes in lake volume and temperature have been pronounced under the background of warming and wetting in recent decades (Ma et al., 2022). These alterations would change lakes’ physical (e.g., the intensity of evaporation and stratification) and chemical (e.g., salinity and pH) properties and thus affect As mobilization in QTP lakes.

Our study revealed a pattern of low DAs in the river outflow and freshwater lakes, with concentrations intensifying in closed saline lakes due to evaporation. This process was exacerbated by the concurrent rise

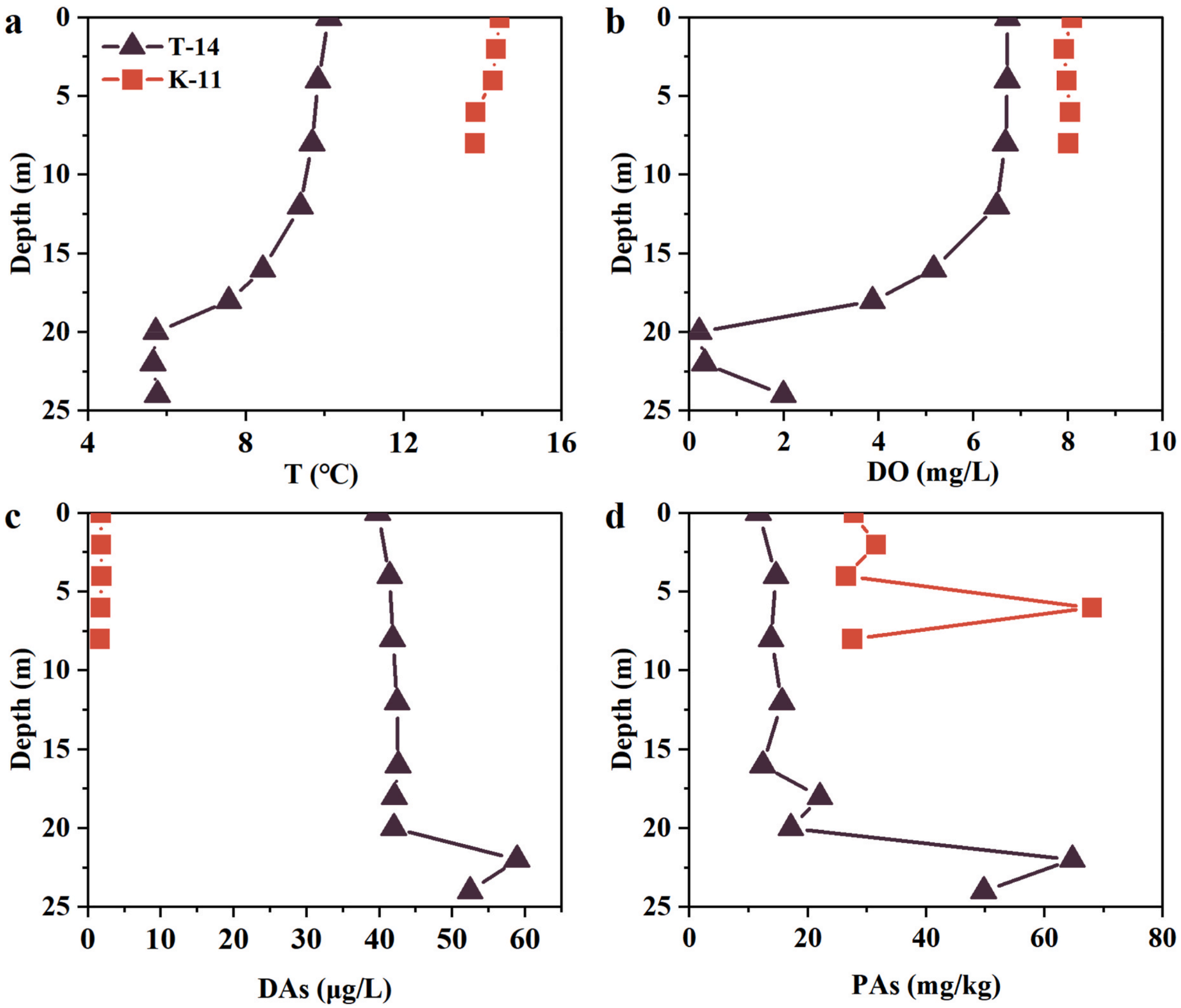


Fig. 3. Variation in temperature T (a), cd DO (b), DAs (c), PAs (d) in the water columns of the two lakes. K-11 was the water column in Hurleg Lake water; T-14 were the water column in Tosen Lake.

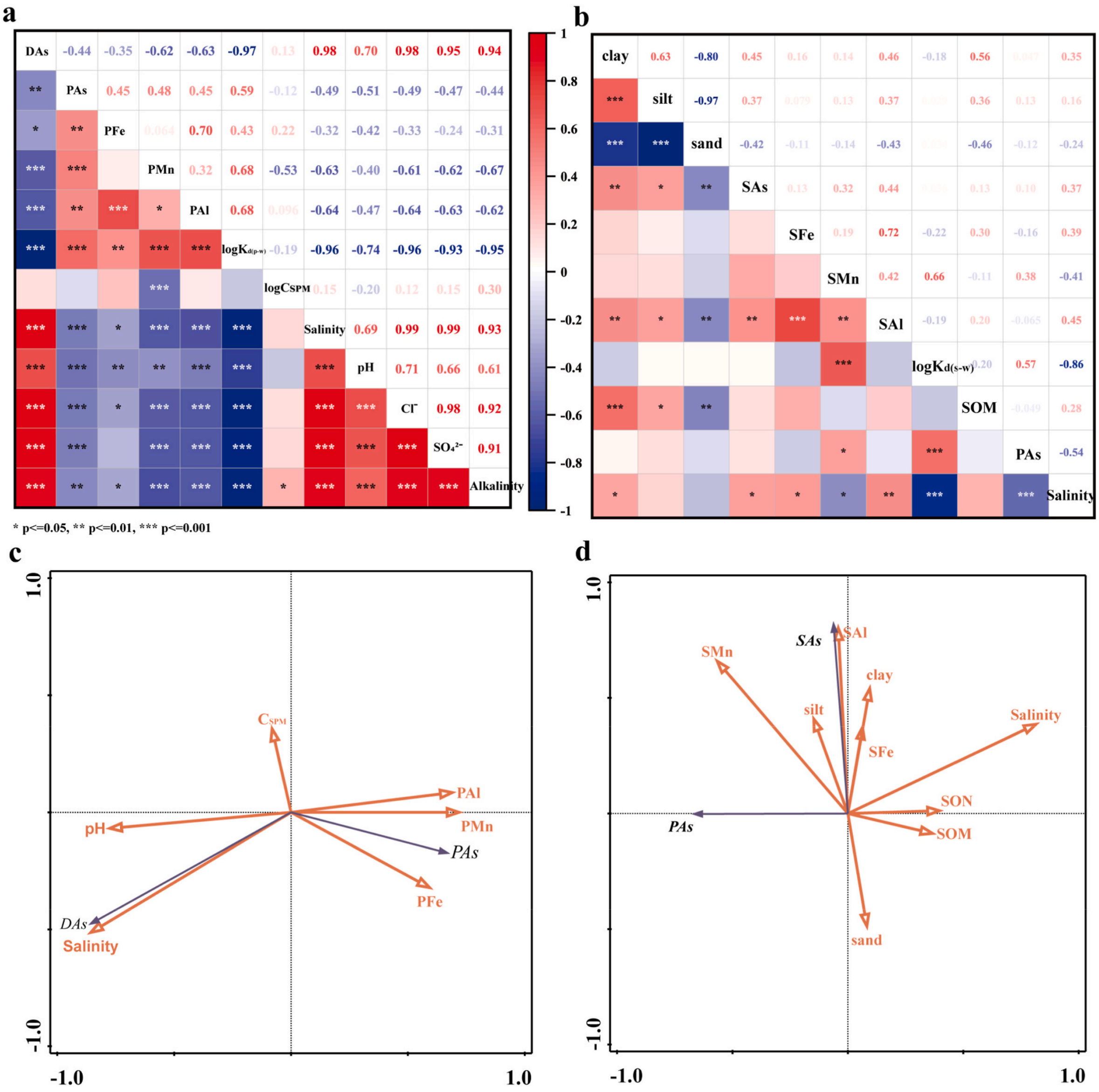


Fig. 4. Correlation analysis of arsenic and environmental variables in water phases (a: water phases; b: sediment phase) and the redundancy analysis of arsenic and environmental factors (c: water phases; d: sediment phase). Since the predominant species of dissolved inorganic carbon in the pH range in the study area was bicarbonate (HCO_3^-), the alkalinity obtained in this study was nearly equal to the concentration of HCO_3^-. *: $p \leq 0.05$; **: $p \leq 0.01$; ***: $p \leq 0.001$.

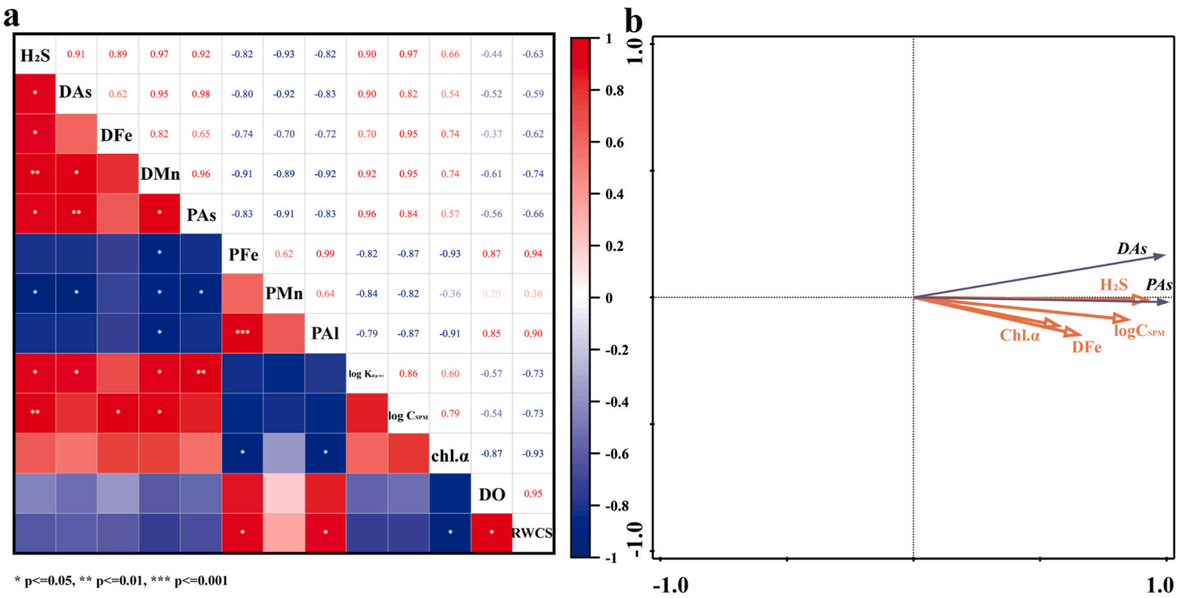
in salinity and pH, which enhanced As release from the SPM (Fig. 6). Under the context of ongoing climate change, lake expansion and shrinkage have been widely observed in the QTP since the 1970s (Zhang et al., 2020). Lake expansion, driven by increased precipitation, could dilute DAs and reduce salinity, promoting As adsorption onto the SPM. For example, the salinity of Caiji Co on the QTP declined dramatically from 15.84 g/L in the 1980s to 1.35 g/L in 2021 (Liu et al., 2021). Such a dramatic decline was likely to influence the distribution of As in aqueous systems, preventing the release of As from SPM or sediment to water. In contrast, lake shrinkage enhances As mobilization under intense evaporation, and exacerbates As mobilization, with residual waters becoming more saline and alkaline.

In addition to altering salinity and pH, climate change may also be responsible for the intensification and prolongation of thermal and DO stratification in lakes (Zhang et al., 2015). This, in turn, can initiate the redistribution of As in multimedia. Under the context of climate warming, the capacity of the lake to retain oxygen diminishes as temperatures rise, due to a decrease in oxygen solubility (Zhang et al., 2015). According to the current study, the DO stratification established an anoxic situation in the bottom layer, triggering the mobilization of As via the reductive dissolution of Fe (hydr)oxides and SBR in the deepest parts of the lake, such as Tosen Lake (Fig. 6). Hence, climate warming could increase As mobilization in stratified lakes due to intensified DO depletion.

These findings emphasize the potential for climate change-driven alterations in salinity, pH, and DO to influence As redistribution in multimedia in arid plateau lakes. However, how the synergistic or reactive effects between environmental factors affect As mobilization remains to be quantitatively understood. Therefore, it is imperative to conduct further exploration on the distribution and mobilization of toxic arsenic within the context of ongoing climate change.

4. Conclusion

The spatial distribution of As in multiphase media was investigated by comparing freshwater and saltwater lakes in a typical small watershed in the QTP. The saltwater lake was high in DAs and low in PAs, whereas the freshwater lake exhibited the opposite pattern. Tosen



H_2S	0.91	0.89	0.97	0.92	-0.82	-0.93	-0.82	0.90	0.97	0.66	-0.44	-0.63
*	DAs	0.62	0.95	0.98	-0.80	-0.92	-0.83	0.90	0.82	0.54	-0.52	-0.59
*		DFe	0.82	0.65	-0.74	-0.70	-0.72	0.70	0.95	0.74	-0.37	-0.62
**	*		DMn	0.96	-0.91	-0.89	-0.92	0.92	0.95	0.74	-0.61	-0.74
*	**		*	PAs	-0.83	-0.91	-0.83	0.96	0.84	0.57	-0.56	-0.66
			*		PFe	0.62	0.99	-0.82	-0.87	-0.93	0.87	0.94
*	*		*	*		PMn	0.64	-0.84	-0.82	-0.36	0.20	0.36
			*		***		PAl	-0.79	-0.87	-0.91	0.85	0.90
*	*		*	**				$\log K_{d(p\text{-}w)}$	0.86	0.60	-0.57	-0.73
**		*	*						$\log C_{SPM}$	0.79	-0.54	-0.73
					*		*			chl.α	-0.87	-0.93
											DO	0.95
					*		*			*	*	RWCS

* p<=0.05, ** p<=0.01, *** p<=0.001

Fig. 5. Correlation analysis (a) and redundancy analysis (b) of arsenic and environmental variables in the water phase in the bottom layer (below 16 m) in profile T-14. *: $p \leq 0.05$; **: $p \leq 0.01$; ***: $p \leq 0.001$.

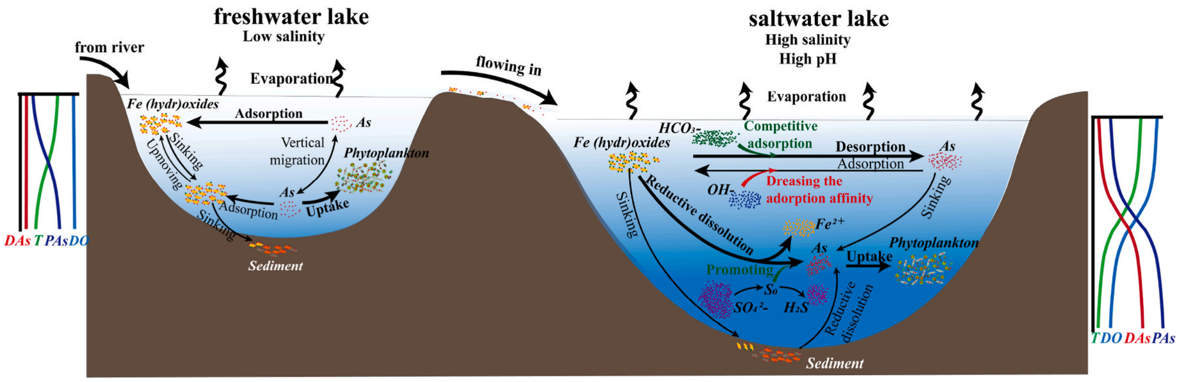


Fig. 6. Conceptual diagram showing different processes controlling As mobilization in the freshwater lake and the saltwater lake. The direction and width of the arrows refer to the transfer process and magnitude, respectively.

Lake's pronounced evaporation facilitated DAs enrichment, with concurrent increases in HCO_3^- and pH levels promoting As desorption from SPM, yielding higher water-SPM As partitioning coefficients. Depth profiles in Tosen Lake revealed ascending DAs and PAs, whereas Hurleg Lake registered a rise in PAs alone, with stable DAs. RDA analysis corroborated stratification's critical role in As distribution within water columns. In the saline lake, pronounced DO stratification enabled As release through the reductive dissolution of Fe (hydr)oxides and bacterial sulfate reduction under the anoxic environment. Elevated chl.α and SPM levels in both lakes' profundal zones indicated significant uptake/adsorption by biogenic particles to PAs increments. Thermal stratification further restricted vertical As transfer, resulting in PAs accretion in the bottom water. This investigation advanced the comprehension of As behavior, revealing its intricate distribution and partitioning in plateau lacustrine ecosystems.

CRediT authorship contribution statement

Dongli Li: Formal analysis, Resources, Writing – original draft, Writing – review & editing. **Haibo He:** Conceptualization, Funding acquisition, Writing – review & editing. **Mengdi Yang:** Writing – review & editing. **Xuecheng Zhang:** Resources. **Tianhao Guan:** Resources. **Wenjing Dai:** Writing – review & editing. **Yan Li:** Writing – review & editing. **Hang Shao:** Resources. **Shiyuan Ding:** Conceptualization, Funding acquisition, Writing – review & editing. **Xiaodong Li:** Conceptualization, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envres.2024.118132.

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