



Review



# Arsenic Contamination in Groundwater of Sea-Land Interaction Zones: Diagnosis and Remediation

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**Abstract:** Sea-land interaction zones (SLIZs) are critical socio-economic and ecological hotspots, whose groundwater serves as the core freshwater supply for over 40% of the global coastal population. Widespread arsenic (As) contamination in SLIZs groundwater, with nearly 50% of global coastal aquifers persistently exceeding the WHO drinking water guideline of 10 µg/L, poses severe chronic health risks and threatens coastal ecosystem integrity. Through comprehensive literature retrieval, classification induction, and comparative analysis of technologies, this review systematically synthesizes current progress in the diagnosis, transformation mechanisms, and remediation of As contamination in SLIZs groundwater. Key quantitative findings reveal that tidal fluctuations drive over 40% diurnal variation in coastal As concentrations, yet approximately 85% of existing field surveys rely on static single-point sampling that fails to capture these transient dynamics. This work clarifies the unique coupled natural-anthropogenic As sources and tide-driven speciation transformation and migration rules specific to SLIZs, critically evaluates the severe adaptability limitations of existing diagnostic and remediation systems in dynamic high-salinity coastal environments, and proposes targeted key challenges and future research priorities. This review provides essential scientific support for As contamination control and water security safeguarding in global coastal regions.

**Keywords:** sea-land interaction zones; groundwater; arsenic contamination; speciation transformation; contamination diagnosis; remediation technology

## 1. Introduction

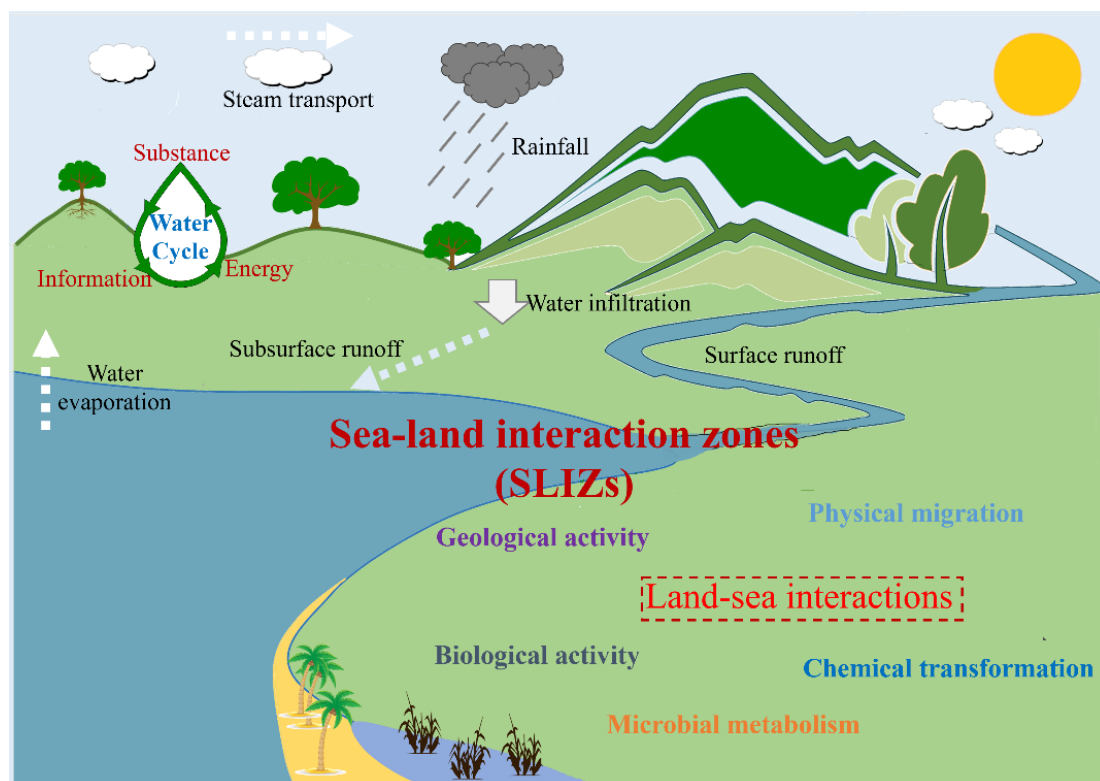
Sea-land interaction zones (SLIZs), which cover estuaries, deltaic systems, and tide-influenced coastal aquifers, are key regions with irreplaceable socio-economic value and ecological functions worldwide [1]. Globally, groundwater systems are increasingly recognized as complex integrated systems where the fate and transport of contaminants are governed by coupled hydrogeochemical, hydrological, and microbial processes. These interconnected processes not only regulate the mobilization of inorganic contaminants, including arsenic, but also control the spread of microbial and viral contaminants, creating co-contamination scenarios that pose compounded risks to human health and ecological security. Unlike general coastal aquifers, which are often less affected by tidal dynamics and saltwater-freshwater mixing, SLIZs are defined by intense land-sea interactions, periodic tidal pumping, frequent saltwater-freshwater interface fluctuations, and strong redox oscillations. These zones extend approximately 10 km landward and 10–15 km seaward, forming one of the most active earth surface systems for the exchange of material, energy, and information flows [2,3]. This unique dynamic environment leads to distinctly different arsenic sources, speciation transformation, and migration mechanisms compared with those



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in general coastal aquifers, making SLIZs a necessary and independent focus for this review. United Nations reports show that approximately 40% of the global population resides within 100 km of the coastline, and 75% of the world's megacities are concentrated in coastal areas [4]. These regions contribute more than 60% of the global gross domestic product (GDP) through core industries including fisheries, shipping and coastal tourism [4,5]. Because SLIZs exhibit far stronger dynamic heterogeneity, salinity variability, and biogeochemical activity than general coastal aquifers, their groundwater arsenic contamination behavior cannot be fully represented or explained by studies on ordinary coastal systems. Groundwater in SLIZs acts as the core freshwater source for domestic drinking, agricultural irrigation and industrial production of coastal residents, and also serves as a biogeochemical buffer between terrestrial and marine environments (Figure 1), supporting mangroves, intertidal wetlands and ecosystems with high biodiversity, while effectively mitigating natural hazards such as seawater intrusion and storm surges [1]. However, the widespread arsenic (As) contamination in the groundwater of these zones is posing a severe threat to this foundation of sustainable development. Inorganic arsenic is classified as a Group 1 human carcinogen by the World Health Organization (WHO), with a drinking water guideline limit of 10  $\mu\text{g/L}$  [6,7]. The chronic exposure to arsenic can induce malignant tumors in multiple organs, including the skin, lung and bladder, and cause non-carcinogenic damages such as neurotoxicity, endocrine disruption and cardiovascular diseases, with more than 150 million people affected globally [8]. At the ecological level, arsenic bioaccumulates in aquatic food webs, exerts acute and chronic toxicity to benthic organisms and fish, and severely damages the ecological structure and fishery resource security of estuarine wetlands such as those in the Yellow River and Yangtze River basins [9]. Therefore, clarifying the migration and transformation mechanisms of arsenic in SLIZs groundwater and developing targeted remediation technologies have become an urgent requirement for safeguarding regional water security and ecological balance.



**Figure 1.** Sea-land interaction zones (SLIZs) and land-sea interactions.

Notably, as key global hotspots of groundwater arsenic contamination, SLIZs have a widespread occurrence of arsenic concentrations exceeding the WHO limit of 10  $\mu\text{g/L}$  in typical coastal aquifers. This contamination pattern is particularly severe in the Ganges-Brahmaputra-Meghna Delta, where more than 60% of the shallow aquifers have arsenic concentrations above 10  $\mu\text{g/L}$ , with peak values even reaching thousands of  $\mu\text{g/L}$  [10,11]. Similar high-risk areas are also widely distributed in the Mekong Delta, the Yangtze River Delta and Pearl River Delta of China, the Atlantic coastal aquifers of the United States, the Baltic Sea coastal areas of Europe, and the Niger River Delta in Africa [7,12]. The unique hydrogeochemical processes driven by land-sea interactions endow the arsenic mobilization and migration mechanisms in this region with significant specificity. On the one hand, the dynamic fluctuation of the saltwater-freshwater interface induces intense redox oscillations, which accelerate the

reductive dissolution of iron-manganese (Fe/Mn) oxyhydroxides and promote the release of bound arsenic [12]. On the other hand, tidal and fluvial processes transport terrestrial organic matter (e.g., arsenic-rich industrial waste residues), which transforms the occurrence speciation of arsenic through microbial metabolic pathways [13]. Meanwhile, competitive adsorption of anions (e.g.,  $\text{HCO}_3^-$ ,  $\text{Cl}^-$ ) under salinity gradients can also significantly weaken the arsenic retention capacity of aquifer media. This phenomenon is particularly prominent in the production workshop areas adjacent to river banks. Frequent fluctuations of groundwater level lead to vertical migration of arsenic with seepage flow, while point source pollution, such as gold mine waste residues, causes arsenic enrichment in downstream riverbed sediments by approximately 10 times the standard limit through surface runoff [14,15]. Notably, the high proportion of exchangeable arsenic in soils further exacerbates the threat of groundwater contamination [7]. Existing research frameworks for inland arsenic contamination mechanisms or marine arsenic biogeochemical cycles are difficult to adapt to the highly dynamic, heterogeneous and complex environments of SLIZs [10]. Therefore, there is an urgent need to construct a dynamic diagnostic model integrating multi-interface processes (soil-water-sediment) and a tide-responsive remediation technology system to break the remediation dilemma of this unique pollution barrier in SLIZs.

Over the past decades, systematic research on arsenic contamination in inland groundwater has established a well-developed theoretical framework and technical system. Global-scale surveys have clearly delineated hotspots of high-arsenic aquifers, including the Hetao Basin in China, the Central Valley of the United States, and the Chaco-Pampean Plain in Argentina [10,16]. The mobilization mechanisms of arsenic in these regions, including the reductive dissolution of Fe/Mn oxyhydroxides, competitive adsorption by coexisting anions (e.g.,  $\text{PO}_4^{3-}$ ), and pH-driven desorption, have been fully validated through multi-scale studies integrating field monitoring, laboratory simulation, and numerical modeling [12]. Mature remediation technologies, such as permeable reactive barriers (PRBs) and in-situ immobilization, combined with diagnostic methods including multi-isotope tracing ( $\delta^{18}\text{O}$ - $\delta^2\text{H}$ - $\delta^{34}\text{S}$ ) and in-situ high-resolution sampling with microelectrodes, have achieved remarkable performance in homogeneous inland aquifers [17,18]. In the field of marine groundwater systems, existing studies mainly focus on the arsenic flux from submarine groundwater discharge (SGD), which contributes 5–15% of the total arsenic input to the global ocean [19,20]. The methylation/demethylation speciation transformation of arsenic in sediments, and the impacts of salinity on adsorption-desorption equilibrium, with core attention paid to the ecological risk assessment of the marine arsenic biogeochemical cycle. In recent years, scattered case studies on SLIZs' arsenic contamination have emerged, including regional contamination surveys in the Ganges-Brahmaputra-Meghna Delta, Mekong Delta, and China's Yangtze River Delta and Pearl River Delta, as well as process studies revealing that salt-freshwater mixing and tidal pumping can accelerate arsenic release from Fe/Mn oxyhydroxides [21,22]. A small number of studies have also preliminarily explored the application of iron-based adsorbents and microbial remediation in local coastal sites [23]. However, these studies remain highly fragmented and lack systematic integration. Approximately 85% of field surveys adopt a static single-point sampling mode, which makes it difficult to resolve the transient migration flux of arsenic under the coupled effects of tides, sea-level rise, and storm surges [15]. For example, monitoring data show that river level fluctuations cause a diurnal variation of coastal arsenic concentration exceeding 40% [4]. Meanwhile, research on the coupled effects of arsenic in the terrestrial soil-coastal groundwater-offshore sediment continuum is still lacking. Furthermore, existing remediation technologies exhibit severely limited adaptability to the highly dynamic, high-salinity environments of SLIZs, highlighting the fundamental limitations of current technical systems when applied to these highly heterogeneous coastal aquifers. Notably, no comprehensive review has yet established a full-chain research framework covering source identification, speciation transformation, migration mechanism, diagnostic technology and remediation application specifically for SLIZs arsenic contamination, which has become a major bottleneck restricting the development of targeted pollution control strategies.

Aiming at the critical research gaps identified above, this review provides the first systematic and critical synthesis of the state of knowledge on groundwater arsenic contamination in SLIZs, filling the gap left by previous reviews that focused exclusively on inland or marine systems. The core objectives of this review are as follows: first, to systematically sort out and critically evaluate the full-chain diagnostic methods for groundwater arsenic contamination in SLIZs, addressing the limitations of static sampling approaches. Second, to comprehensively collate and assess the applicability, engineering performance, environmental adaptability, advantages, and limitations of existing arsenic remediation technologies in complex coastal aquifer settings, with a specific focus on their tolerance to high salinity and tidal fluctuations. Finally, to clarify the key unresolved scientific issues and core application bottlenecks restricting the theoretical development and engineering practice in this field, and propose targeted future research priorities. In terms of research scope, this review focuses on geogenic and anthropogenic arsenic contamination in groundwater systems within typical SLIZs, with core research targets being coastal aquifers and estuarine groundwater environments. This review explicitly excludes studies on arsenic

contamination in surface water bodies, including estuarine surface water, coastal seawater, and terrestrial surface runoff, as well as studies on arsenic contamination in purely inland or deep-sea groundwater systems beyond the land-sea interaction interface.

## 2. Review Methodology

This review was conducted following a standardized and reproducible literature review methodology. A comprehensive literature retrieval was performed across three core academic databases: Web of Science Core Collection, Scopus, and ScienceDirect, covering publications from 2000 to 2026. The search strategy combined the following keywords: (“arsenic contamination” or “arsenic pollution”) and (“groundwater” or “aquifer”) and (“sea-land interaction zones” or “coastal zones” or “estuaries” or “deltas”) and (“speciation transformation” or “migration mechanism” or “contamination diagnosis” or “remediation technology”). Peer-reviewed original research articles and critical reviews were prioritized, while non-English publications, conference abstracts, book chapters, and gray literature were excluded. A total of 327 relevant studies were initially retrieved, and after screening for relevance and quality, 113 high-impact studies were included in the final synthesis. The selected literature was systematically categorized into three thematic modules corresponding to the core objectives: (1) arsenic sources, speciation transformation, and migration mechanisms; (2) contamination diagnostic technologies and risk assessment methods; (3) remediation technologies and their environmental adaptability. Critical comparative analysis was then conducted across each module to evaluate the strengths, limitations, and application scopes of existing theories and technologies, and to identify persistent research gaps and future research priorities.

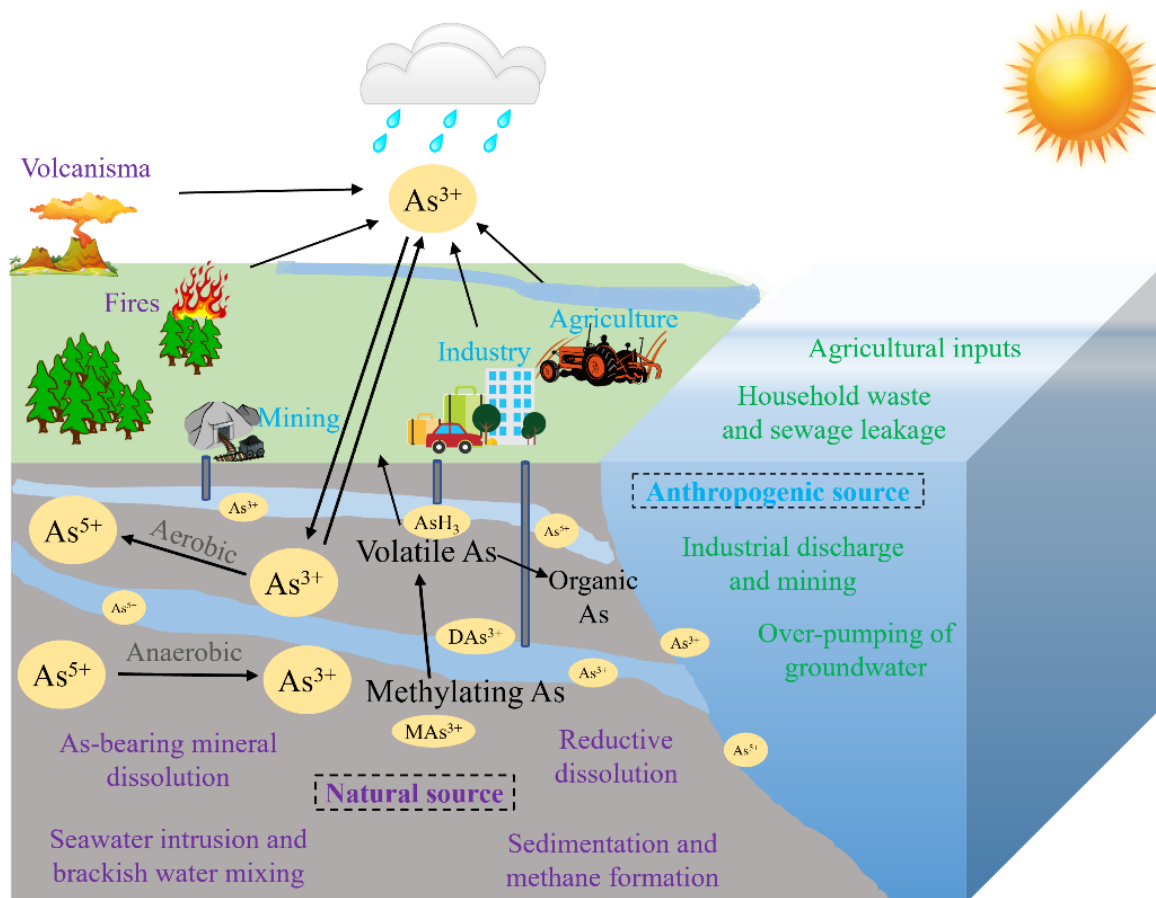
## 3. Arsenic Sources, Speciation Transformation and Migration Mechanisms in SLIZs Groundwater

### 3.1. Sources of Arsenic Contamination

Groundwater arsenic contamination in SLIZs is fundamentally characterized by the coupled superposition of natural geogenic sources and anthropogenic inputs, and this complex contamination pattern is deeply modulated by the unique dynamic hydrogeochemical conditions inherent to coastal environments (Figure 2). Natural geogenic sources are the dominant contributors to high-arsenic coastal aquifers worldwide. First, arsenic-bearing sulfide minerals (e.g., arsenopyrite) release soluble arsenite ( $\text{As}^{3+}$ ) or arsenate ( $\text{As}^{5+}$ ) during oxidative weathering in aquifers [24]. Second, Fe/Mn oxide-bearing sediments act as secondary arsenic reservoirs, where reductive dissolution occurs under anaerobic conditions, leading to the massive release of adsorbed arsenic, and this process is significantly enhanced by microbially mediated reduction driven by organic matter decomposition [25,26]. Experimental results show that up to 62% of highly labile arsenic (exchangeable fraction + carbonate-bound fraction) in soils can be released within 96 h under anaerobic conditions [27,28]. Most critically, salt-freshwater interaction induced by seawater intrusion exacerbates the spread of arsenic contamination via dual pathways. On the one hand, seawater cations ( $\text{Na}^+$ ,  $\text{Ca}^{2+}$ ) displace arsenic adsorbed on the binding sites of clay minerals or Fe/Mn hydroxides, resulting in elevated dissolved arsenic concentrations. On the other hand, the chemosmotic zone formed by saltwater intrusion promotes the metabolism of sulfate-reducing bacteria, and the generated sulfide combines with  $\text{As}^{3+}$  to form highly mobile thioarsenate complexes (e.g.,  $\text{H}_2\text{As}_3\text{S}_6^-$ ) [29,30]. Isotope tracing ( $\delta^{18}\text{O}$ ,  $\delta^2\text{H}$ ) has quantitatively verified the mobilization effect of seawater on sedimentary arsenic, which forms a highly heterogeneous process with a spatial coefficient of variation exceeding 200% in estuarine areas [31,32].

Anthropogenic contamination further elevates the natural background levels of arsenic via multiple pathways. In agricultural production, arsenic-containing pesticides (e.g., lead arsenate) and phosphate fertilizers enter aquifers via leaching by precipitation after application, and the infiltration of heavy rainfall during the rainy season particularly causes a significant increase in the fluctuation of groundwater arsenic concentrations [33,34]. For example, arsenic-containing feed additives such as roxarsone are used in coastal aquaculture, whose metabolites infiltrate shallow aquifers after biotransformation [6]. The impacts of industrial activities are more direct. First, arsenic-laden wastewater and waste residues discharged by metallurgical, mining and other industries contaminate groundwater via surface runoff or leakage [35,36]. In addition, dissolved organic matter (DOM) in leachate from municipal landfills promotes the desorption of arsenic from sediments via complexation. Notably, arsenic contamination caused by unregulated wastewater discharge from tanning, textile and other industries is characterized by a higher spatial coefficient of variation and localized pollution hotspots. Second, closed mining sites continuously release arsenic via underground runoff-seepage interactions, serving as an important source of groundwater arsenic contamination [37]. The synergistic amplification effect of these anthropogenic sources and natural processes, such as the influence of tide-driven water level fluctuations on the vertical migration rate of arsenic, ultimately forms a contamination chain of terrestrial input-tidal driving-interfacial mobilization. This

results in arsenic concentrations in nearly 50% of global coastal aquifers persistently exceeding the WHO drinking water guideline limit, and poses a long-term threat of chronic health risks [7,10,11].



**Figure 2.** Sources (natural and anthropogenic) and transformation of arsenic in groundwater in the sea-land interaction zone. (Note: methylarsenate,  $\text{MAS}^{3+}$ ; dimethylarsenate,  $\text{DAs}^{3+}$ ).

### 3.2. Speciation Transformation of Arsenic

The toxicity, migration potential and biogeochemical behavior of arsenic in groundwater of SLIZs are fundamentally governed by its chemical speciation. Driven by the unique dynamic hydrogeochemical conditions of coastal environments, the transformation processes of arsenic speciation are far more frequent and complex than those in inland aquifers. In groundwater of SLIZs, inorganic arsenic is the predominant species, accounting for more than 90% of the total dissolved arsenic, which mainly exists in the forms of trivalent arsenite ( $\text{As}^{3+}$ ) and pentavalent arsenate ( $\text{As}^{5+}$ ) [28,38,39]. Organic arsenic species, including monomethylarsonic acid (MMA) and dimethylarsinic acid (DMA), are generally detected only at low concentration levels and are mainly produced via microbial methylation of inorganic arsenic [40]. From a toxicological perspective, inorganic arsenic is classified as a Group 1 human carcinogen by the WHO, among which  $\text{As}^{3+}$  exhibits approximately 60-fold higher acute toxicity and stronger carcinogenicity than  $\text{As}^{5+}$  [9,41]. Notably, under the typical neutral pH conditions of coastal groundwater,  $\text{As}^{3+}$  mainly exists in the form of uncharged arsenous acid ( $\text{H}_3\text{AsO}_3$ ), which has significantly higher migration capacity and lower adsorption affinity to aquifer sediments compared with anionic  $\text{As}^{5+}$ . Previous studies have shown that the desorption rate of  $\text{As}^{3+}$  in sandy sediments is 3 to 5 times faster than that of  $\text{As}^{5+}$  [42]. This difference directly leads to a wider contamination plume of  $\text{As}^{3+}$  in groundwater with a higher spatial coefficient of variation, which also poses a greater risk to groundwater security and ecological health.

The speciation transformation of arsenic in SLIZs is co-regulated by four types of coastal-specific coupled driving factors. First, fluctuations in redox potential (Eh) are the core controlling factor. Periodic tidal pumping, seawater intrusion and estuarine organic matter degradation drive drastic fluctuations in Eh, from  $-200$  mV in the wet season to  $+150$  mV in the dry season, forming alternating oxic-anoxic conditions [43]. Oxic environments promote the oxidation of  $\text{As}^{3+}$  to less mobile  $\text{As}^{5+}$ , while anoxic environments enhance arsenic mobility via two pathways, with the reductive dissolution of Fe/Mn oxyhydroxides releasing adsorbed arsenic [42]. Meanwhile, microbially mediated reduction of  $\text{As}^{5+}$  produces highly mobile  $\text{As}^{3+}$ . Experimental evidence confirms that

dissolved arsenic concentration can surge from a background level of  $<10 \mu\text{g/L}$  to  $7650 \mu\text{g/L}$  within 96 h under microbial action, with  $\text{As}^{3+}$  accounting for more than 85% of the total [29,30]. Second, pH fluctuations indirectly regulate the transformation processes by altering mineral surface charge and arsenic protonation states. Under the combined effect of seawater buffering and acid rain, decreased pH enhances the positive surface charge of Fe/Mn oxides and promotes the adsorption of  $\text{As}^{5+}$ , while increased pH weakens adsorption via electrostatic repulsion and drives the proportion of dissolved  $\text{As}^{3+}$  to increase by nearly 50% [30]. Third, microbial activity serves as the core hub of biological driving forces. Salinity gradients shape unique microbial communities, with culturable arsenic-oxidizing bacteria (e.g., *Achromobacter*) dominating in low-salinity areas ( $<5\%$ ), and arsenic-methylating bacteria (e.g., Methanogen) enriched in high-salinity areas ( $>15\%$ ) [44]. These bacterial communities directly catalyze redox and methylation reactions, with their effects being particularly prominent in organic-rich mangrove aquifers. Fourth, salt-freshwater mixing triggers multi-pathway chemical responses. The high ionic strength of seawater weakens arsenic adsorption via the charge shielding effect, leading to a nearly 50% increase in desorption capacity [29]. Meanwhile, sulfate input from seawater drives sulfate reduction, which forms insoluble arsenian pyrite ( $\text{FeS}_{2-x}\text{As}_x$ ) to immobilize arsenic under incompletely reducing conditions, but produces soluble thioarsenates (e.g.,  $\text{AsS}_3^{3-}$ ) in strongly reducing, sulfur-rich environments, with their toxicity and mobility being 2.3 times that of  $\text{As}^{3+}$  [45]. The complexity of this speciation transformation is manifested as drastic concentration fluctuations in the tidal flat transition zone. In the rainy season, arsenic concentrations can nearly double within a single day, while during the dry season, concentrations can increase by a factor of 200 or more, far exceeding the WHO drinking water guideline limit by 45 [8,46]. This underscores the critical impact of dynamic speciation changes on risk assessment.

The transformation of specifically bound arsenic species cannot be ignored either. Exchangeable arsenic in soils can rapidly enter aquifers via leaching during the rainy season, while organically bound arsenic is released, driven by organic matter degradation during microbial mineralization, and this pathway represents a significant source of arsenic release in carbon-rich sediments [47]. Arsenic bound in sulfides exhibits a lag in transformation under oxygenated conditions, with a release rate significantly lower than that of free arsenic. However, when the amplitude of oscillations at the freshwater-saltwater interface increases, fluctuations in dissolved oxygen can lead to the sustained release of this arsenic compound [48]. Although the microbially mediated methylation pathway can produce low-toxicity organic arsenic, demethylation readily occurs under moderate to low salinity conditions (5‰~20‰), which reconverts organic arsenic to highly toxic  $\text{As}^{3+}$  [40]. The spatiotemporal coupling of the above processes makes SLIZs a hotspot for arsenic speciation transformation. Thus, the transformation sensitivity of arsenic needs to be comprehensively considered to accurately predict and control the trend of risk diffusion.

### 3.3. Migration Mechanisms of Arsenic

In SLIZs, the spatiotemporal distribution and long-distance transport of arsenic in groundwater are dominated by coupled hydrodynamic and geochemical processes. Affected by the dynamic salt-freshwater interaction and highly heterogeneous hydrogeological conditions of coastal systems, this process exhibits characteristics fundamentally distinct from those in inland aquifers. The hydrodynamic migration of arsenic is primarily driven by regional groundwater flow, which controls the lateral and vertical transport of dissolved arsenic along hydraulic gradients in estuarine and deltaic aquifers [11]. The most characteristic driving factors, such as periodic tidal fluctuations and seawater intrusion induced by storm surges, can significantly enhance the migration and diffusion of arsenic by altering the groundwater flow field. The drastic swing of the salt-freshwater interface forms a hydraulic pumping effect, which drives the rapid transport of deep high-arsenic groundwater to shallow aquifers [49,50]. Meanwhile, precipitation recharge in the rainy season causes a rapid increase in groundwater flow velocity, accelerates the diffusion of contamination plumes, and leads to a highly heterogeneous horizontal distribution of highly labile arsenic [51]. Furthermore, the abrupt nature of this migration is particularly prominent, and colloid-bound transport is a unique pathway in the terrestrial-marine transition zone. Iron oxyhydroxide colloids and natural organic matter colloids can strongly adsorb arsenic via surface complexation, while salinity fluctuations regulate their transport distance through a switch mechanism of dispersion and flocculation [52]. During the low-salinity period ( $\text{Cl}^- < 0.1 \text{ mol/L}$ ), the zeta potential of colloids can rise above  $|-30| \text{ mV}$ , with the dispersion degree increasing by nearly half, extending the horizontal transport distance of colloid-bound arsenic to 35 times that of dissolved-phase arsenic [30,35]. In contrast, the high-salinity environment induced by seawater intrusion promotes the flocculation and sedimentation of colloids, which in turn forms a secondary arsenic enrichment zone in the saltwater front [29]. Microbial activity indirectly regulates this process by altering colloid stability. For example, the secretion of bacterial extracellular polymeric substances (EPS) can significantly increase colloid transport flux, and the contribution of this mechanism to total arsenic transport can reach 40% in organic-rich tidal flat aquifers [6,53].

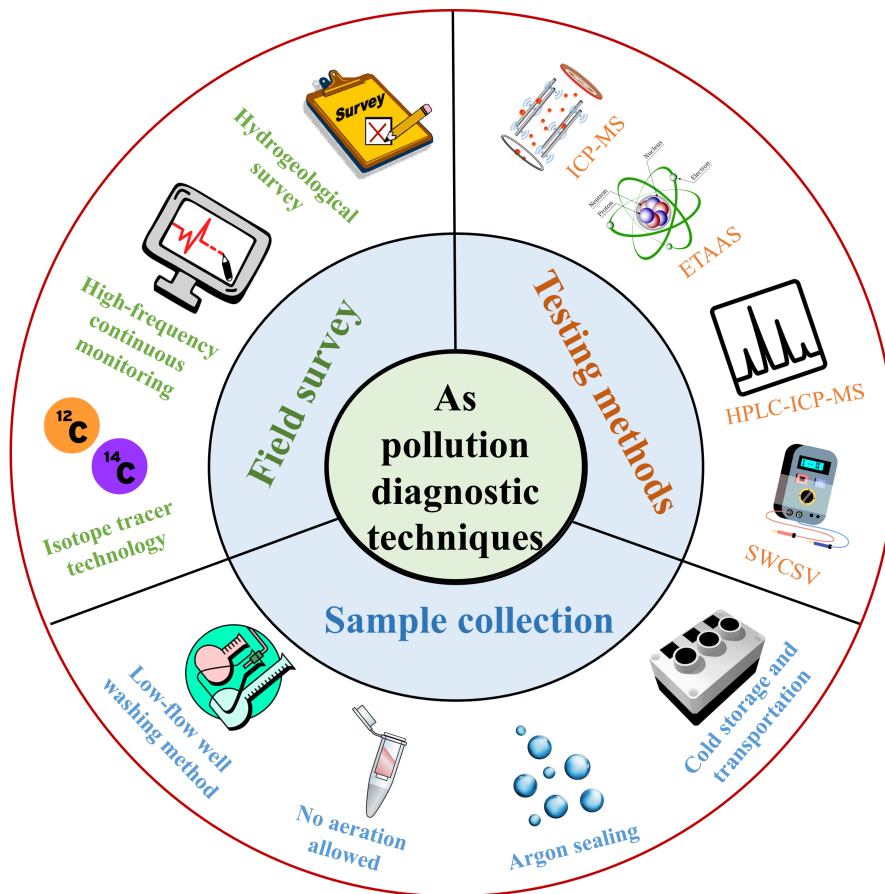
The dynamic equilibrium of adsorption and desorption constitutes the core of the geochemical regulation of arsenic. Under normal conditions, arsenic-bearing ferrihydrite has an arsenic adsorption capacity of up to 3.5 mg/g, while seawater intrusion can trigger a dual desorption effect [54]. On the one hand, high ionic strength weakens electrostatic adsorption via charge shielding, increasing the desorption rate by nearly one third. On the other hand, competitive anions ( $\text{HCO}_3^-$ ,  $\text{SO}_4^{2-}$ ) occupy mineral surface sites, which can increase the amount of desorbed arsenic by nearly one third [55]. More critically, microbially mediated reductive dissolution of iron oxides under anaerobic conditions can directly destroy the host carriers of arsenic, and tide-driven redox oscillations cause this process to occur repeatedly in the intertidal zone. The spatial differentiation characteristics of precipitation and dissolution transformations further regulate the environmental fate of arsenic. In the sulfate reduction zone ( $\text{SO}_4^{2-} > 100 \text{ mg/L}$ ), approximately 35% of dissolved arsenic is immobilized in sediments via the formation of arsenic sulfide phases (e.g., orpiment  $\text{As}_2\text{S}_3$ , realgar  $\text{As}_4\text{S}_4$ ) and arsenian pyrite [25]. However, local sulfur-excess conditions in the transition zone can generate soluble thioarsenates ( $\text{AsS}_3^{3-}$ ), whose mobility is more than 2 times higher than that of  $\text{As}^{3+}$  [56]. Notably, anthropogenic disturbances complicate this process. Engineering activities such as bank slope excavation expose deep reducing sediments, and oxygen intrusion triggers the oxidative dissolution of arsenic-bearing sulfides, resulting in a significant increase in arsenic flux released from secondary contamination [57]. The combined effects of hydrodynamic, colloidal, adsorption and precipitation processes make SLIZs a high-speed channel for arsenic migration, and also determine that arsenic contamination remediation requires the synergistic blocking of multi-interface processes.

Climate change is emerging as a critical amplifying factor that exacerbates arsenic mobilization in SLIZs by intensifying saltwater intrusion, enhancing surface leaching, and disrupting redox equilibrium. However, precise quantitative analysis of the individual contributions of specific climate change factors (e.g., sea-level rise, extreme rainfall, storm surges) remains extremely challenging at present. This difficulty stems from multiple interconnected confounding factors. First, the extreme spatial heterogeneity of coastal hydrogeological conditions leads to highly variable responses of arsenic cycling to climate perturbations even within the same SLIZ. Second, climate change drivers interact nonlinearly with existing natural hydrogeochemical processes and intensive human activities such as groundwater overexploitation and coastal reclamation, making it impossible to isolate the pure effect of any single climate factor. Third, there is a severe global shortage of long-term continuous high-resolution monitoring data that simultaneously records climate parameters, hydrogeochemical conditions, and arsenic concentrations, which is essential for establishing quantitative relationships. While existing regional studies have observed general trends of increased arsenic mobilization associated with climate change events, the magnitude of these impacts varies widely across different sites and cannot be reliably extrapolated to global scales.

## 4. Diagnostic Technologies for Arsenic Contamination in SLIZs Groundwater

### 4.1. Field Investigation and Sample Collection

Hydrogeological field investigation and standardized sample collection constitute the fundamental cornerstone for the accurate diagnosis of groundwater arsenic contamination in SLIZs (Figure 3). Its technical system must be fully adapted to the unique environmental characteristics of SLIZs, including the strong heterogeneity of aquifers, tide-driven dynamic fluctuations of hydrodynamic conditions, and frequent migration of the saltwater-freshwater interface. These efforts aim to provide reliable basic data for delineating the distribution range of arsenic contamination, revealing its migration mechanisms, and conducting subsequent risk assessment and remediation design. For typical estuarine, deltaic and coastal aquifer systems within SLIZs, hydrogeological surveys adopt a comprehensive approach integrating borehole drilling, in-situ geophysical exploration, multi-point long-term monitoring network, geospatial mapping and piezometric monitoring, to systematically identify the spatial occurrence characteristics of aquifers, including lithological structure, stratum thickness, permeability coefficient and the distribution of aquitards [58,59]. Meanwhile, high-frequency continuous monitoring of groundwater level, hydraulic gradient and groundwater flow direction is conducted to eliminate monitoring deviations caused by periodic tidal fluctuations [60]. This method can also accurately characterize the spatial extent of seawater intrusion and the dynamic migration law of the saltwater-freshwater interface, thereby identifying the recharge, runoff and discharge pathways of the regional groundwater flow system. Specific attention should be paid to the unique hydrodynamic processes of estuaries in the study, such as tide-driven oscillatory flow and SGD. In addition, isotope tracing technology is combined with permeability coefficient profiles to identify the main migration pathways of seawater intrusion [32]. Areas with a conductivity gradient exceeding 10 mS/cm can effectively delineate the hotspots of arsenic migration.



**Figure 3.** Diagnostic techniques for arsenic contamination in groundwater in the sea-land interaction zone, including field surveys, sample collection, and analytical testing.

In the sample collection process, strict full-process operating protocols must be implemented to prevent the transformation of arsenic speciation, targeting the characteristics of SLIZs groundwater, including high salinity, variable redox conditions, and high sensitivity of arsenic speciation to environmental interference. Groundwater samples are strictly collected using the low-flow well purging method, with a dedicated flow-through cell equipped to maintain in-situ redox conditions and control their fluctuations within 50 mV. Sampling can only be initiated after the in-situ parameters, including pH, Eh, dissolved oxygen, conductivity and water temperature, are stabilized. Aeration is strictly prohibited throughout the sampling process to avoid oxidative transformation of highly toxic trivalent arsenic ( $\text{As}^{3+}$ ) [61]. Undisturbed sediment cores are collected using a nitrogen-purged push-core sampler to minimize air contact and oxidation of the samples, which are immediately sealed in amber vials under an argon atmosphere. Immediately after collection, all samples are filtered on-site through 0.45  $\mu\text{m}$  polyethersulfone membranes. Subsequently, total arsenic samples are acidified to  $\text{pH} < 2$  on-site with ultra-pure nitric acid or hydrochloric acid to inhibit the adsorption of arsenic on container walls, precipitation and microbial methylation. Samples for arsenic speciation analysis are aliquoted, sealed, and immediately stored and transported under low-temperature ( $\leq 4\text{ }^{\circ}\text{C}$ ) and dark conditions to terminate biotic transformation processes. Field blanks, transport blanks and parallel samples are set simultaneously during the sampling process to ensure the accuracy, reliability and repeatability of the test data.

#### 4.2. Laboratory Detection Methods

Laboratory detection methods are the core technical support for the quantitative characterization of groundwater arsenic contamination in SLIZs, which must overcome three key challenges: high-salinity matrix, trace arsenic species, and complex geochemical interfacial interactions. For the quantification of total arsenic, inductively coupled plasma mass spectrometry (ICP-MS), relying on helium collision mode (KED) or dynamic reaction cell (DRC) technology, can effectively eliminate polyatomic interferences caused by high salinity and achieve an ultra-trace detection limit of 0.01  $\mu\text{g/L}$  [36,50]. In practical applications, this method presents recovery rates of 90–98% with a coefficient of variation (CV)  $\leq 5\%$ , and maintains a linear response even for heavily contaminated samples with locally high arsenic concentrations [8,50]. Electrothermal atomic absorption

spectrometry (ETAAS), despite its slightly lower sensitivity (detection limit of 1 µg/L), can significantly inhibit volatile interferences from chlorides in the seawater matrix by optimizing the pyrolysis curve with a matrix modifier, thus providing critical validation for ICP-MS data [62,63]. Atomic fluorescence spectrometry (AFS) is characterized by coupling with hydride generation (HG). It converts  $\text{As}^{5+}$  to  $\text{As}^{3+}$  using a thiourea-ascorbic acid pre-reduction system, generates arsine ( $\text{AsH}_3$ ) via online reaction with potassium borohydride, and achieves a detection limit of 0.02 µg/L, making it a cost-effective and efficient approach for large-scale screening [64].

Arsenic speciation analysis requires higher resolution, and high-performance liquid chromatography coupled with ICP-MS (HPLC-ICP-MS) can achieve baseline separation (resolution >1.5) of  $\text{As}^{3+}$ ,  $\text{As}^{5+}$ , MMA and DMA within 12 min, with a limit of quantitation (LOQ) of 0.1 µg/L, through an anion exchange column and gradient elution with phosphate buffer [50]. Most critically, this technology has an elevated salinity tolerance up to 35 g/L NaCl, which is perfectly suitable for samples from seawater intrusion areas, and can correct signal drift via an online internal standard (e.g.,  $^{74}\text{Ge}$ ) [8]. Among complementary techniques, square wave cathodic stripping voltammetry (SWCSV), with a gold-nanoparticle modified electrode, enables direct determination of  $\text{As}^{3+}$  without pre-separation and reaches a detection limit of 0.2 µg/L, which has unique advantages for the study of in-situ redox dynamics such as tide-driven Eh fluctuations [65]. The analysis of arsenic occurrence phases in sediments relies on an improved sequential extraction procedure, and the Tessier five-step sequential extraction method divides arsenic into exchangeable fraction (extracted by  $\text{NH}_4\text{Cl}$ ), Al-bound fraction (extracted by  $\text{NH}_4\text{F}$ ), Fe-bound fraction (extracted by NaOH), Ca-bound fraction (extracted by  $\text{H}_2\text{SO}_4$ ) and residual fraction (digested by aqua regia) [66]. Among these fractions, Fe-bound arsenic and exchangeable arsenic collectively account for more than half of the total arsenic contamination flux to groundwater [67]. Supporting molecular biological techniques such as quantitative real-time polymerase chain reaction (qPCR) can elucidate microbially driven arsenic mobilization mechanisms by quantifying the abundance of arsenic reduction gene (*arsC*) and respiratory arsenic reduction gene (*arrA*), thereby deepening the understanding of the underlying mechanisms [27,68].

Current diagnostic technologies for arsenic contamination in SLIZs have made significant progress, but they still face three core interconnected limitations that prevent accurate capture of real-time arsenic dynamics. Firstly, the insufficient temporal resolution of static sampling remains the most critical bottleneck, as approximately 85% of global coastal arsenic surveys adopt static single-point or monthly sampling modes [15], which completely fail to resolve the >40% diurnal arsenic concentration fluctuations driven by tidal pumping [4], and quantitative analysis shows that this approach can only detect less than 30% of peak arsenic concentrations, leading to systematic underestimation of contamination risks by 2–5 times. Furthermore, commercial in-situ arsenic sensors exhibit poor performance under high-salinity conditions, with a drift rate of >20% per month in SLIZs with salinity >15‰ that requires weekly calibration and maintenance, and most sensors can only measure total arsenic concentrations and cannot distinguish between highly toxic  $\text{As}^{3+}$  and less toxic  $\text{As}^{5+}$ , which is essential for accurate human health risk assessment. Finally, there is a widespread lack of multi-parameter synchronous monitoring, as over 70% of existing monitoring networks only measure arsenic concentrations without synchronous recording of redox potential (Eh), salinity, dissolved oxygen, and pH, the hydrogeochemical parameters that directly regulate arsenic speciation transformation, and their absence makes it impossible to establish causal relationships between environmental changes and arsenic dynamics.

#### 4.3. Numerical Simulation and Risk Assessment

Numerical simulation and risk assessment constitute the advanced core of the arsenic contamination diagnostic system for SLIZs. This not only bridges the full workflow from contamination characterization to the implementation of targeted remediation, but also provides quantitative support for pollution trend prediction and risk zoning regulation. For the simulation of arsenic migration, the core of model construction for SLIZs lies in the coupled characterization of density-dependent hydrodynamic processes and multi-field coupled biogeochemical reactions. The mainstream simulation frameworks include MODFLOW-MT3DMS for solute transport simulation, SEAWAT for variably saturated density flow simulation, as well as PHREEQC and TOUGHREACT for coupled hydrogeochemical simulation [60,69]. The above models fully integrate the unique driving factors of SLIZs, including periodic tidal fluctuations, salt-freshwater interaction, seawater intrusion and tide-driven oscillatory flow, while embedding core processes such as adsorption-desorption of arsenic on Fe/Mn oxides, redox speciation transformation, and microbially mediated reactions [30]. The models are calibrated via field-measured adsorption isotherms and validated against datasets of arsenic mobility within tidal cycles, which can accurately characterize the spatiotemporal migration laws of arsenic in heterogeneous coastal aquifers, identify contamination hotspots, and predict the long-term evolution trend of contamination under sea-level rise stress.

In terms of risk assessment, this system covers human health risk assessment and ecological risk assessment, supported by standardized grading criteria. Among them, the human health assessment adopts the models recommended by the United States Environmental Protection Agency (USEPA) Integrated Risk Information System (IRIS) and the WHO, focusing on the drinking water ingestion exposure pathway to calculate carcinogenic risk values and non-carcinogenic hazard quotients [70,71]. The assessment follows universal standards, where a carcinogenic risk value lower than  $10^{-6}$  is defined as a negligible level, a value between  $10^{-6}$  and  $10^{-4}$  is an acceptable level, and emergency control measures are required when the value exceeds  $10^{-4}$  (or the hazard quotient is greater than one) [72]. The ecological risk assessment focuses on coastal ecosystems dependent on groundwater. It employs sediment quality guidelines, the risk quotient method, and species sensitivity distribution models, and applies the theory of equilibrium distribution to predict benthic toxicity [73]. By incorporating the bioaccumulation characteristics of arsenic and utilizing isotope tracing technology to identify pollution sources, the assessment achieves a risk classification based on liability.

## 5. Remediation Technologies for Arsenic Contamination in SLIZs Groundwater

### 5.1. Physicochemical Remediation Technologies

Physicochemical remediation technologies are the core approaches for the remediation of groundwater arsenic contamination in SLIZs. The main advantages and challenges of arsenic remediation technologies are summarized in Table 1. Besides, Table 2 analyzes the limitations of these remediation technologies from the aspects of arsenic removal efficiency and life cycle cost. In particular, physical remediation of arsenic contamination primarily relies on adsorption. Among adsorption technologies, iron-based materials have become the preferred option owing to their high-efficiency sequestration capacity for both  $\text{As}^{3+}$  and  $\text{As}^{5+}$  (typically  $\geq 150$  mg/g) [74,75]. Surface hydroxyl modification and phosphate esterification can significantly enhance the resistance to competitive adsorption from  $\text{Cl}^-$  and  $\text{SO}_4^{2-}$ . For example, in backfilled aquifers, modified nanoscale hydrous ferric oxide (HFO) exhibits an adsorption capacity more than 4 times higher than that of unmodified activated carbon, with a salinity tolerance threshold exceeding 25 g/L [76]. However, conventional adsorbents such as clay and biochar show significant performance degradation under high-salinity conditions, requiring regular chemical regeneration to maintain their efficacy in practical applications. Among membrane separation technologies, nanofiltration (NF) can simultaneously retain more than 90% of  $\text{As}^{5+}$  and more than 50% of salinity, owing to its charge effect and pore size sieving properties (1–5 nm) [77]. However, colloids formed by humic acid and divalent cations ( $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ) in coastal groundwater can cause irreversible membrane fouling, with a monthly flux decay rate as high as 40% [78]. Meanwhile, although reverse osmosis (RO) achieves a comprehensive removal rate higher than 95%, its high energy consumption and operational cost restrict its large-scale application.

Chemical precipitation methods achieve remarkable performance in freshwater systems, but coexisting ions in SLIZs severely inhibit floc formation. Sulfate competes with arsenic for  $\text{Fe}^{3+}$  binding sites, leading to a nearly 50% sharp reduction in the precipitation efficiency of Fe-As complexes [79]. When salinity exceeds 10 g/L, aluminum-based coagulants are more prone to form soluble chloroaluminum complexes, resulting in residual aluminum concentrations exceeding 200  $\mu\text{g/L}$  [35]. The generated arsenic-bearing sludge is prone to secondary release of exchangeable arsenic in sediments under tide-driven redox fluctuations, requiring subsequent solidification treatment. However, iron-anode electrocoagulation technology exhibits unique advantages. High salinity (conductivity  $> 5$  mS/cm) significantly reduces energy consumption, and *in situ*-generated goethite ( $\text{FeOOH}$ ) efficiently immobilizes arsenic via ligand exchange, yielding a stable effluent arsenic concentration below 5  $\mu\text{g/L}$  that is barely affected by Eh fluctuations [80,81]. In engineering applications, electrodialysis achieves an arsenic removal rate of up to 85% for brackish water (salinity 3–10 g/L), but its retention rate for organic colloids with a particle size less than 0.1  $\mu\text{m}$  is below 50% [82]. In contrast, the coupled electrocoagulation-RO system decomposes organic matter through pre-positioned electrochemical oxidation, extending the service life of RO membranes by 60%, and achieved 300 days of continuous scaling-free operation in a pilot test in the saltwater intrusion area of the Yangtze River Delta [83]. It should be emphasized that post-remediation environmental risks cannot be ignored. Under long-term seawater erosion, the leaching toxicity of associated heavy metals such as lead and cadmium in solidified sludge may far exceed the regulatory limits, and a long-term monitoring mechanism is urgently required to support decision-making.

**Table 1.** Main advantages and challenges of groundwater arsenic remediation technologies in the sea-land interaction zone, including physicochemical remediation, bioremediation, and combined remediation technologies.

Remediation Technology	Main Advantages and Characteristics	Main Challenges and Limitations
Physical remediation	<ul style="list-style-type: none"> <li>High technical maturity: Membrane separation (RO/NF), extraction technologies widely applied</li> <li>No chemical agents required: Avoids secondary chemical pollution</li> <li>High removal efficiency: Reverse osmosis can achieve &gt;90% arsenic removal</li> <li>High automation: Easy operation and monitoring</li> <li>Small footprint: Suitable for small centralized water supply systems</li> </ul>	<ul style="list-style-type: none"> <li>High energy consumption: Especially membrane separation technology has high operating costs</li> <li>Membrane fouling: Requires regular cleaning and membrane replacement</li> <li>Incomplete pollutant elimination: Only transfers pollutants rather than degrading them</li> <li>High salinity environment impact: Saline-freshwater interface affects treatment efficiency</li> <li>Concentrated waste generation: Requires further treatment and disposal</li> </ul>
Chemical remediation	<ul style="list-style-type: none"> <li>Short remediation time: Quick results</li> <li>Clear targeting: Strong specificity, precise control possible</li> <li>Mature technology: Chemical precipitation, redox methods are well-developed</li> <li>Significant arsenic removal: Iron salts form stable precipitates with arsenate</li> <li>Wide applicability: Can treat high-concentration arsenic contamination</li> </ul>	<ul style="list-style-type: none"> <li>Secondary pollution risk: Chemical reagent residues may affect water quality</li> <li>High reagent costs: Large economic burden for long-term operation</li> <li>Alters water chemistry: May affect original groundwater balance</li> <li>Limited by high salinity: Saline-freshwater interface affects chemical reaction efficiency</li> <li>Requires continuous dosing: Cannot achieve one-time permanent remediation</li> </ul>
Microbial remediation (Bioremediation)	<ul style="list-style-type: none"> <li>Environmentally sustainable: Utilizes natural microbial metabolic processes</li> <li>Low cost: Low operation and maintenance expenses</li> <li>No secondary pollution: Pollutants are transformed or immobilized</li> <li><i>In-situ</i> remediation possible: No need to extract groundwater</li> <li>Compatible with other technologies: Bio-stimulation or bioaugmentation</li> </ul>	<ul style="list-style-type: none"> <li>Long remediation period: Requires months to years to show effects</li> <li>Sensitive to environmental conditions: Affected by temperature, pH, Eh</li> <li>High salinity inhibits activity: Microbial activity is limited in seawater intrusion areas</li> <li>Arsenic speciation transformation risk: May produce more toxic trivalent arsenic</li> <li>Strain selection difficulties: Need to adapt to the special SLIZs environment</li> </ul>
Phytoremediation	<ul style="list-style-type: none"> <li>Ecologically friendly: Green purification improves the ecological environment</li> <li>Lowest cost: Low initial investment and operating expenses</li> <li>Harvestable removal: Hyperaccumulator plants can harvest and remove arsenic</li> <li>Environmental beautification: Dual landscape function</li> <li>Rhizosphere synergy: Plant-microbe combined remediation</li> </ul>	<ul style="list-style-type: none"> <li>Longest remediation period: Usually requires years or even longer</li> <li>Plant growth limitations: High salinity and waterlogging conditions affect growth</li> <li>Limited remediation depth: Only suitable for shallow groundwater</li> <li>Post-treatment required: Arsenic-enriched plant biomass needs safe disposal</li> <li>Seasonal limitations: Plant growth is affected by seasons</li> </ul>
Combined remediation	<ul style="list-style-type: none"> <li>Comprehensive advantages: Integrates multiple technology benefits</li> <li>High remediation efficiency: Synergistic effects accelerate the remediation process</li> <li>Wide application scope: Can handle complex contamination scenarios</li> <li>High flexibility: Can adjust combinations based on actual conditions</li> <li>Good sustainability: Aligns with green remediation concepts</li> </ul>	<ul style="list-style-type: none"> <li>High technical complexity: Requires professional design and optimization</li> <li>High initial cost: Large investment for multi-technology integration</li> <li>High management difficulty: Need to coordinate multiple technical parameters</li> <li>Compatibility challenges: Potential conflicts between different technologies</li> <li>Lack of standard specifications: Combination schemes require case-by-case customization</li> </ul>

**Table 2.** Comparative evaluation of groundwater arsenic remediation technologies in sea-land interaction zones.

Technology Category	Typical Technologies	Typical Arsenic Removal Efficiency	Full Life Cycle Cost	Key Limitations in SLIZs
Physical remediation	Reverse osmosis (RO), nanofiltration (NF), and adsorption	RO: >95% Modified iron-based adsorption: 90–99%	High (RO: \$1.2–2.5/m <sup>3</sup> ) Medium (adsorption: \$0.5–1.2/m <sup>3</sup> )	Membrane fouling (40% monthly flux decay), high energy consumption, and concentrated waste disposal
Chemical remediation	Electrocoagulation, chemical precipitation, and ion exchange	Electrocoagulation: 90–98%, Chemical precipitation: 70–85%	Medium (electrocoagulation: \$0.6–1.5/m <sup>3</sup> ) High (ion exchange: \$1.0–2.0/m <sup>3</sup> )	High salinity reduces precipitation efficiency, chemical sludge secondary release risk, and electrode passivation
Microbial remediation	<i>In-situ</i> biostimulation, immobilized functional bacteria	70–85%	Low (\$0.2–0.6/m <sup>3</sup> )	Long remediation period (3–12 months); salinity inhibits microbial activity; biomass loss from tidal fluctuations
Plant remediation	Salt-tolerant hyperaccumulators, constructed wetlands	50–75%	Very low (\$0.1–0.3/m <sup>3</sup> )	Limited to shallow groundwater (<2 m); seasonal growth variation; post-harvest biomass disposal
Combined remediation	Adsorption-membrane, electro-microbial, phytoremediation-electrokinetic	95–99.5%	Medium-high (\$0.8–1.8/m <sup>3</sup> )	High initial investment; complex system operation; lack of standardized design protocols

## 5.2. Bioremediation Technologies

Bioremediation technologies exhibit unique advantages in the remediation of groundwater arsenic contamination in SLIZs by virtue of their environmental friendliness, cost-effectiveness, and high compatibility with coastal ecosystems. This technical system addresses the challenges of high salinity, tide-driven hydrodynamic fluctuations, and complex hydrochemical conditions through dual strategies. Microbial remediation relies on arsenic-metabolizing functional microbial consortia to achieve the speciation transformation and immobilization of arsenic, and salt-tolerant arsenic-oxidizing bacteria (e.g., *Thiomonas* spp.) can oxidize more than 90% of  $\text{As}^{3+}$  to readily adsorbable  $\text{As}^{5+}$  at a salinity of 15 g/L via expressing arsenite oxidase encoded by the *aox* gene [84,85]. In reducing aquifers, sulfate-reducing bacteria (e.g., *Desulfovibrio* spp.) and arsenate-reducing bacteria (e.g., *Sulfurospirillum* spp.) form a synergistic metabolic network, which promotes the conversion of dissolved arsenic into insoluble arsenic sulfide ( $\text{As}_2\text{S}_3$ ) or its co-precipitation with pyrite ( $\text{FeS}_2$ ), achieving an arsenic retention efficiency of 80% in tidal flat sediments [68]. To address biomass loss caused by tidal fluctuations, microbial immobilization technology encapsulates functional consortia in a composite carrier of sodium alginate and biochar, which not only enhances salt stress resistance but also maintains biological activity through a sustained-release effect, reducing the arsenic concentration in the effluent to below 5  $\mu\text{g/L}$  [86,87]. However, the competitive inhibition of inoculated strains by indigenous microbial communities may reduce remediation efficiency, and in-situ biostimulation is required to activate native arsenic-transforming functional microorganisms.

The core of phytoremediation lies in the screening of salt-tolerant arsenic hyperaccumulators adapted to high-salinity environments. For example, smooth cordgrass (*Spartina alterniflora*) can still accumulate arsenic at a salinity of 20 g/L, and blocks arsenic translocation to aerial tissues via vacuolar sequestration [88,89]. The mangrove-associated plant grey mangrove (*Avicennia marina*) dissolves Fe/Mn oxide-bound arsenic by secreting root organic acids, which significantly improves the bioavailability of arsenic [90]. This rhizosphere synergistic effect among plants, microorganisms and fungi is the key to the efficiency enhancement of phytoremediation technologies. The hyphal network of mycorrhizal fungi (e.g., *Glomus* spp.) expands the root absorption area, while rhizosphere functional bacteria (e.g., *Pseudomonas* spp.) promote Fe-As co-precipitation by producing siderophores, increasing the arsenic immobilization rate in micro-domain soil by nearly half [91,92]. Constructed wetland systems integrate the above mechanisms and can enhance arsenic removal efficiency by establishing artificially selected plant species and enriched functional microbial communities while simultaneously reducing tide-borne organic pollution loads [93]. Notably, gene editing technology is breaking through natural limitations. Studies have found that transgenic feather cockscomb (*Celosia argentea*) with overexpressed *ACR3* gene achieves a 2-fold increase in arsenic translocation factor (TC) and a nearly 40% increase in biomass, providing new germplasm resources for phytoremediation in high-salinity areas [85]. Nevertheless, bioremediation has a relatively long cycle (usually 2–5 years), and secondary metabolic risks still need to be mitigated by coupling with passivation materials such as hydroxyapatite.

## 5.3. Combined Remediation Technologies

Aiming at the complexity of high salinity and dynamic hydrological conditions in SLIZs, combined remediation technologies break through the efficiency bottlenecks of single technologies via multi-stage synergistic mechanisms. In the adsorption-membrane separation hybrid system, hydrous ferric oxide-reduced graphene oxide (HFO-rGO) nanocomposites, with their high specific surface area and sulfonic acid group modification, can effectively bind arsenic in water with salinity higher than 10 g/L [94,95]. Meanwhile, they can remove more than 90% of humic acid and 80% of divalent cations, reducing the flux decay rate of the subsequent NF membrane to 8% per month, which is far lower than 35% per month without pretreatment [74,95]. This system achieved a stable effluent arsenic concentration of  $<2 \mu\text{g/L}$  (removal efficiency  $>99.5\%$ ) in a pilot-scale test in the saltwater intrusion area of the Pearl River Estuary, with the water treatment cost per cubic meter being only 67% of that of the single RO process [96,97]. The microbial-chemical precipitation synergistic mechanism activates indigenous arsenic-oxidizing microbial communities via sulfate-reducing bacteria (*Desulfovibrio desulfuricans*), converting 98% of  $\text{As}^{3+}$  to  $\text{As}^{5+}$  within one day and generating ferrous sulfide (FeS) colloidal nuclei in an environment with  $\text{Eh} < -150 \text{ mV}$  [48,98]. The addition of low-dose  $\text{FeCl}_3$  (10 mg/L) can form  $\text{FeOOH-AsO}_4^{3-}$ -FeS ternary flocs, reducing the sludge yield by one third compared with the conventional coagulation process, and increasing the proportion of residual fraction arsenic in the precipitates to more than 70%, which significantly inhibits the secondary release risk under tidal fluctuations [28,93]. The electrochemical-adsorption hybrid system can fully exploit the inherent advantage of high conductivity ( $>5 \text{ mS/cm}$ ) in SLIZs. Nascent  $\text{Fe}^{2+}/\text{Fe}^{3+}$  generated by iron-anode electrocoagulation forms a dynamic adsorption interface with pre-packed thiol-functionalized mesoporous silica, which enhances  $\text{As}^{3+}$  sequestration via Fe-S coordination bonds and simultaneously mitigates

electrode passivation [99]. This system achieves an 85% removal rate for organic colloids with a particle size  $<0.1$   $\mu\text{m}$ , and successfully resisted salinity fluctuations caused by offshore rainstorms in the coastal aquifer of the Yellow Sea, maintaining the effluent arsenic concentration below  $5$   $\mu\text{g/L}$  [9]. Notably, the emerging phytoremediation-electrokinetic synergistic system realizes the directional migration of deep arsenic to the shallow rhizosphere enrichment zone through the guiding effect of electro-osmotic flow around the root system of arsenic hyperaccumulators [100,101]. After the simultaneous application of a  $0.5$   $\text{V/cm}$  direct current electric field, the arsenic accumulation capacity of the plant increases to  $1800$   $\text{mg/kg}$ , and the remediation cycle is shortened by nearly  $40\%$  [90,102]. However, permeability loss caused by mineral precipitation during long-term operation remains a key bottleneck. The recently developed UV/ultrasound-nanoscale zero-valent iron (nZVI) in-situ regeneration device can extend the service life of PRBs to more than 5 years. Through the modular combination of functional units, combined technologies provide tailored solutions for different contamination scenarios, with a significantly reduced comprehensive cost in multi-stress environments.

In this review, we quantitatively compare their effectiveness under varying salinity conditions using three key indicators: (1) baseline arsenic removal efficiency under freshwater conditions (salinity  $<1\%$ ); (2) performance retention rate at moderate salinity ( $5\%$ – $15\%$ ); and (3) relative efficiency under high salinity ( $20\%$ – $35\%$ ) typical of SLIZs. Conventional physical adsorption and microbial remediation show a  $20$ – $40\%$  reduction in removal efficiency with increasing salinity, mainly due to competitive ion interference and inhibited microbial activity. In contrast, electrocoagulation and salinity-tolerant modified materials maintain over  $80\%$  of their original remediation efficiency across the full salinity gradient. Combined remediation systems further reduce performance loss to below  $15\%$ , demonstrating superior stability under dynamic saltwater-freshwater mixing conditions.

#### 5.4. Technology Evaluation Criteria

The establishment of a multi-dimensional evaluation system applicable to remediation technologies for groundwater arsenic contamination in SLIZs requires focused consideration of the extreme dynamic characteristics and ecological vulnerability of high-salinity tidal environments. In terms of the remediation efficiency dimension, in addition to focusing on the effluent arsenic concentration, the critical impacts of anion competition and arsenic speciation transformation must be evaluated. When the concentration of  $\text{HCO}_3^-$  or  $\text{SO}_4^{2-}$  exceeds  $25$   $\text{mmol/L}$ , the arsenic adsorption capacity of conventional iron-based adsorbents may decrease by nearly half, while the microbial-electrochemical coupled system can maintain a removal rate higher than  $90\%$  through the in-situ generated  $\text{Fe}^{2+}/\text{Fe}^{3+}$  redox cycle [103,104]. Full life cycle cost analysis must cover salinity-driven implicit costs. For example, the doubled regeneration frequency of sodium-type ion exchange resins caused by tide-borne  $\text{Mg}^{2+}$  input leads to a significant increase in annual operating costs [105]. In contrast, although bioremediation has a low initial investment, additional costs for salt-tolerant microbial inoculant amendment are required under conditions with  $\text{Cl}^-$  concentration exceeding  $15,000$   $\text{mg/L}$  [104]. Second, environmental impact assessment requires the quantification of secondary risks. The release potential of residual arsenic in chemical sludge under tidal wet-dry cycles and the highly enriched arsenic concentration in membrane concentrate must be matched with supporting negative pressure evaporation and solidification treatment to avoid marine ecological toxicity [9]. The core of the long-term stability of the remediation system lies in the stress resistance of materials and functions. Studies have confirmed that for nZVI-doped biochar reactive barriers under periodic salt-freshwater scouring, for example, with a salinity fluctuation range of  $4$ – $30$   $\text{g/L}$ , the iron dissolution rate is less than  $8\%$  after 5 years, while the arsenic immobilization rate remains higher than  $86\%$  [74,94]. In contrast, the permeability coefficient of pure iron scrap reactive barriers decreases by  $50\%$  within 2 years due to chloride ion corrosion [106,107].

As an exclusive index for SLIZs, salt-freshwater interaction adaptability needs to be combined with hydrochemical dynamic modeling. In the salt tolerance limit test, the arsenic adsorption capacity of the modified silica gel adsorbent only decreases by  $12\%$  at a  $\text{Cl}^-$  concentration of  $35,000$   $\text{mg/L}$ , which is significantly better than that of resins with a capacity decay of more than  $65\%$  [94,108]. Tide-induced hydrodynamic disturbance can increase the risk of short-circuiting flow in fixed bed reactors by  $35\%$ , while the fluidized bed-electrosorption system has strong adaptability to flux fluctuations through electric field-enhanced mass transfer [109]. Furthermore, the enhanced reducibility of aquifers caused by sea level rise will activate the release of sediment-bound arsenic, which requires the remediation technologies to have redox buffering capacity [110]. For example, bioremediation systems can inhibit the secondary release rate of arsenic through the enzyme-controlled  $\text{As}^{3+}/\text{As}^{5+}$  redox cycle [111]. In the future, it is necessary to develop decision-making tools integrated with multivariate models to realize the quantification and standardization of technology comparison and selection based on parameters such as arsenic speciation distribution, tidal intensity and ecological sensitivity.

## 6. Current Challenges and Future Perspectives

### 6.1. Key Challenges

The accurate diagnosis and efficient remediation of groundwater arsenic contamination in SLIZs still face multi-dimensional challenges, which are fundamentally rooted in the inherent extreme dynamic characteristics of coastal environments and the insufficient adaptability of existing technical systems. The highly non-linear nature of tide-driven hydrogeochemical processes is particularly prominent. The periodic migration of the saltwater-freshwater interface induces continuous redox oscillations, leading to the frequent mobilization of Fe/Mn oxide-bound arsenic in sediments. Studies have found that the secondary release flux of arsenic in the silty clay layer can surge by 48% under conditions where the  $\text{SO}_4^{2-}$  concentration exceeds 25 mmol/L [93,112]. Meanwhile, the superposition of storm surges and sea level rise forms a salinity piston effect, which can sharply increase the  $\text{Cl}^-$  concentration in coastal aquifers to dozens of times the background level, exacerbate competitive interference from anions, and result in a more than 50% decrease in the saturated arsenic adsorption capacity of conventional adsorbents [113]. Such combined stresses significantly undermine the reliability of conventional remediation technologies. For example, in the saltwater intrusion area of the Pearl River Estuary, the abundance of functional microbial communities in the bioremediation system decreased by 60% due to salinity fluctuations, and the remediation cycle was prolonged by 2-fold [12,96]. The core technical bottlenecks are mainly manifested in the imbalance between salt tolerance performance and long-term operational stability. For example, the arsenic adsorption capacity of conventional activated alumina decreases by 35% within 90 days at a  $\text{Cl}^-$  concentration exceeding 10,000 mg/L [23,87].

For ion exchange resins, the regeneration frequency must be increased to once per month due to competitive interference from  $\text{Mg}^{2+}$ , leading to an at least 40% increase in full life cycle cost [114]. Although electrochemical systems have strong adaptive capacity for energy consumption, electrode passivation leads to at least 50% decrease in current efficiency after 2 years of operation [87]. A more critical challenge is the difficulty in controlling secondary pollution. The re-release of highly labile arsenic from chemical sludge under alternating tidal wet-dry conditions, and the highly enriched arsenic concentration in membrane concentrate, pose a direct threat to the coastal wetland ecosystem. In addition, the monitoring and evaluation system is also seriously lagging. Discrete sampling fails to capture the transient changes in arsenic speciation during tidal windows, resulting in discrepancies between the design parameters of remediation systems and actual on-site requirements. Furthermore, the lack of region-specific evaluation criteria leads to inaccurate assessments of remediation project performance. For example, after adopting inland evaluation standards along the coast of the Bay of Bengal, carcinogenic risks were still detected in 32% of the areas that met the compliance criteria [46,72]. The coupling effect of these challenges forms a vicious cycle in heavily contaminated areas, and the cost increase of existing technologies exceeds the affordability of economically underdeveloped regions, which ultimately restricts the large-scale implementation of remediation initiatives.

### 6.2. Future Research Directions

In response to the comprehensive challenges of groundwater arsenic contamination in SLIZs, there is an urgent need to establish an interdisciplinary innovative research system, with a focus on breaking through the integrated bottlenecks in basic theory, material systems, intelligent regulation, and ecological synergy. At the basic mechanism level, in-depth research on multi-field coupling at the dynamic saltwater-freshwater interface should be conducted. It is necessary to accurately quantify the mobilization law of Fe/Mn oxide-bound arsenic driven by tide-induced redox oscillations, and combine gene sequencing technology to decipher the expression characteristics of the ars operon in salt-tolerant microbial communities under osmotic stress, such as the dose-effect relationship between the abundance of Proteobacteria and  $\text{As}^{3+}$  oxidation efficiency. Particular attention should be paid to the kinetic model of secondary arsenic release at the sediment-water interface under competitive conditions with  $\text{HCO}_3^-/\text{SO}_4^{2-} > 25$  mmol/L, to provide molecular-level theoretical support for targeted blocking technologies. Material innovation must balance salt tolerance and environmental friendliness. For environments with high  $\text{Cl}^-$  concentrations, graphene oxide-Fe/Mn bimetallic nanomaterials can be designed, which are expected to achieve a significantly higher arsenic adsorption capacity than conventional activated carbon. Meanwhile, organic fouling-resistant seawater RO membranes coupled with photo-Fenton pre-oxidation units should be developed to simultaneously control the membrane fouling rate. In addition, biochar-based microbial immobilization technology should be optimized to achieve a significant breakthrough in the salinity tolerance threshold of functional bacteria through encapsulation with microbial EPS. The establishment of an intelligent system should integrate multi-source sensing and predictive models. In-situ microelectrode arrays should be deployed to capture the transient changes of arsenic speciation during tidal window periods in real time. A multi-

field coupled numerical model should be integrated to warn of the secondary release risk of arsenic contamination within a certain range by inputting real-time  $\text{Cl}^-$ ,  $\text{SO}_4^{2-}$  concentration and hydrological data, and drive the adaptive adjustment of remediation system parameters. Engineering-oriented research must focus on low cost and ecological compatibility.

Solar-driven combined electroadsorption and NF processes should be developed. The design of pre-oxidation coupled bioreactors should be optimized to improve bioaccumulation efficiency through the enzymatic directional conversion of  $\text{As}^{3+}$ , with the target of significantly reducing the cost per cubic meter of water to ensure economic feasibility. Synergistic remediation technologies coupling mangroves and constructed wetlands should be explored, which use organic acids secreted by the roots of halophytes to complex dissolved arsenic, while simultaneously enhancing the carbon sink function of coastal wetlands. Supporting policies should improve risk classification and regulation. Dynamic risk thresholds based on bioavailability should be established, and a full life cycle digital supervision platform should be implemented to track the migration path of highly labile arsenic in remediation sludge. A cross-administrative land-sea coordination mechanism should also be established to align with climate change adaptation planning. Through the three-in-one collaborative innovation of theory, technology and management, the dual goals of arsenic pollution control and resilience enhancement of coastal ecosystems can be achieved.

## 7. Conclusions

This review systematically synthesizes the sources, speciation transformation and migration characteristics of groundwater arsenic contamination in SLIZs, evaluates the merits and limitations of corresponding diagnostic and remediation technologies, and identifies key research priorities for future work. Arsenic contamination in SLIZs is dominated by the superposition of natural geogenic sources and anthropogenic inputs, deeply modulated by tide-driven dynamic hydrogeochemical processes. A full-chain diagnostic system covering field investigation, laboratory detection and numerical simulation has been established, yet it is limited by insufficient dynamic monitoring and the lack of region-specific evaluation standards. For remediation, physicochemical and bioremediation technologies have their respective advantages, while combined remediation technologies are the core development direction to adapt to the high-salinity and dynamic hydrological conditions of SLIZs. The core challenge lies in the mismatch between existing technologies and the extreme dynamics of coastal environments.

This review provides clear practical implications for policymakers and environmental engineers. First, coastal groundwater management should prioritize tidal zones and salt–freshwater mixing interfaces as key control areas, where high-frequency dynamic monitoring is strongly recommended to capture rapid arsenic fluctuations. Second, engineers should prefer salinity-tolerant iron-based adsorption, electrocoagulation, and combined remediation systems for field applications, as these technologies show the highest stability and efficiency under SLIZ conditions. Third, routine monitoring networks should integrate in-situ sensors for redox, salinity, and arsenic speciation to support real-time risk warning. For policymakers, this review recommends establishing region-specific arsenic risk thresholds tailored to dynamic coastal environments, strengthening cross-administrative land–sea coordination mechanisms, and promoting the development of standardized technical guidelines for diagnosis and remediation in SLIZs. Future research should focus on basic mechanism research, salt-tolerant material innovation, intelligent system construction, and ecological synergy, to provide solid theoretical support and practical technical pathways for arsenic contamination control, water security protection, and ecological health maintenance in SLIZs groundwater worldwide.

## Author Contributions

H.W.: conceptualization, methodology, investigation, writing—original draft preparation; S.O.: investigation, methodology; Q.Z.: supervision, writing—reviewing and editing. All authors have read and agreed to the published version of the manuscript.

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### Data Availability Statement

Data will be made available on request.

### Conflicts of Interest

The authors declare no conflict of interest.

### Use of AI and AI-Assisted Technologies

No AI tools were utilized for this paper.

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