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Spatial distribution and formation mechanisms of high-iodine groundwater throughout China

Junxia Li [a,d], Shilin Zhao [a], Zhou Jiang [a,*], Xianjun Xie [a,d], Yamin Deng [a,d], Liang Shi [a,b,c,d], Andreas Kappler [e], Philippe Van Cappellen [f], Yanxin Wang [a,b,c,d,*]

[a] MOE Key Laboratory of Groundwater Quality and Health, School of Environmental Studies, China University of Geosciences, Wuhan, Hubei, China
[b] State Key Laboratory of Geomicrobiology and Environmental Changes, China University of Geosciences, Wuhan, Hubei, China
[c] Hubei Key Laboratory of Yangtze Catchment Environmental Aquatic Science, China University of Geosciences, Wuhan, Hubei, China
[d] State Environmental Protection Key Laboratory of Source Apportionment and Control of Aquatic Pollution, Ministry of Ecology and Environment, China University of Geosciences, Wuhan, Hubei, China
[e] Geomicrobiology Group, Department of Geosciences, University of Tübingen, Tübingen, Germany
[f] Ecohydrology Research Group, Department of Earth and Environmental Sciences, University of Waterloo, Waterloo, Canada

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ABSTRACT

High-iodine groundwater poses a severe threat to the health of millions of people worldwide, especially in China. Understanding iodine mobilization in aquifers is crucial for sustainable exploitation of groundwater resources. In this Review, we summarize the spatial distribution characteristics of high-iodine groundwater across China, elucidate the sources and hosts of iodine, and discuss the hydrogeological and biogeochemical processes responsible for iodine enrichment in aquifers. High-iodine groundwater is widely distributed in inland semi-arid/arid basins/plains and coastal areas, occurring in both shallow and deep layers with iodide as the dominant iodine species. Terrestrial/marine-derived sedimentary organic matter and organic matter-bound iron minerals are the primary sources of iodine. The mobilization, transformation and enrichment of iodine in aquifers are controlled by both hydrogeological processes (i.e., evaporation concentration, compaction release and burial dissolution) and biogeochemical processes, including iodate reduction by iodate-reducing bacteria, Fe(III)-reducing bacteria, sulfate-reducing bacteria and anaerobic methane-oxidizing microorganisms, and organic iodine degradation/dehalogenation. Future studies should focus on the investigation of radioiodine-contaminated groundwater, identification and quantification of organic iodine species, characterization of anaerobic oxidation of methane coupled with iodate reduction, iodine oxidation and methylation, and cost-effective remediation of high-iodine groundwater.

Contents

* Corresponding authors at: 388 Lumo Road, Wuhan 430074, China.
E-mail addresses: jiangzhou@cug.edu.cn (Z. Jiang), yx.wang@cug.edu.cn (Y. Wang).

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1. Introduction

Iodine is an essential micronutrient for the thyroid gland to produce thyroxine and triiodothyronine. Insufficient iodine intake causes iodine deficiency disorders (IDD) such as goiter, cretinism and intellectual impairment, and became a major global public health problem in the 20th century (Li and Eastman, 2012; Zimmermann, 2009). To manage the public health problem due to iodine deficiency, the World Health Organization (WHO) recommended universal salt iodization (USI) in 1993, which has remarkably improved the health of the global population (Çaylan and Yılmaz, 2017; Pearce and Zimmermann, 2023). The recommended daily iodized salt (iodine: 15 mg/kg salt) intake of about 10 g/day is adequate for adults to prevent population iodine deficiency. However, the risk of excessive iodine intakes may increase if the cumulative intake of iodized salt exceeds 10 g/day or if iodine-rich food and water, particularly seaweed and groundwater, are consumed daily (Farebrother et al., 2019; Wang et al., 2021). Excessive iodine intake can also lead to thyroid dysfunction, such as hypothyroidism, hyperthyroidism and thyroid autoimmunity (Leung and Braverman, 2014). As the largest and most accessible freshwater stock, groundwater provides almost half of all drinking water and supplies ~40 % of irrigation water and ~ 33 % of the water for industry (Lall et al., 2020). Recent work in China has confirmed that natural occurrence of groundwater with elevated iodine concentration poses a threat to the health of people (Lv et al., 2013; Ma et al., 2022). Additionally, iodine excess disorders caused by drinking iodine-rich groundwater have been reported in other countries such as Argentina, Chile, Denmark and Japan (Farebrother et al., 2019; Ma et al., 2022; Voutchkova et al., 2014). Given the global iodine cycling, terrestrial iodine in aquifers is transported through the water cycle to the ocean, which is the largest reservoir of iodine on Earth. Volatile iodine from the ocean enters the atmosphere, causing ozone destruction, atmospheric mercury depletion, and the formation of ultrafine aerosol particles that help cloud condensation and offset radiative forcing (Carpenter et al., 2021; Fuge and Johnson, 2015; Saiz-Lopez et al., 2012). Atmospheric iodine is then transported to the continents and deposited onto the land. Therefore, understanding the distribution of high-iodine groundwater and iodine mobilization in aquifers is of great significance to mitigating public health risk on the basis of understanding global iodine cycling.

China is the first country in the world to report cases of goiter caused by drinking high-iodine groundwater (Shen et al., 2011). In the 1970s, fishermen in the Bohai Bay, Hebei province were found to have a high prevalence of goiter, which was attributed to drinking iodine-rich groundwater (Zhao et al., 1987). Since then, similar iodine-excess goiter cases have emerged in other regions (Mu et al., 1987; Zhao et al., 2000). In 2005, the Chinese Center for Disease Control and Prevention conducted a survey on iodine content in drinking water and found that high-iodine groundwater (total iodine concentration > 150 μg/L) was mainly located in North China Plain (NCP), Huai River Plain, Datong Basin, Taiyuan Basin and Hetao Basin (Shen et al., 2007). To control excessive iodine intake, the Chinese government implemented a policy in 2010 to replace iodized salt with non-iodized salt in these areas (Lv et al., 2015). Although the coverage rate of non-iodized salt in those areas reaches 96.9 %, the median iodine concentration of children urinary is still higher than 300 μg/L, and the prevalence of goiter of children is greater than 5 %, suggesting the need for careful screening of high-iodine groundwater or the change of the drinking water source (Meng et al., 2017). To date, high-iodine groundwater has been reported in 14 provinces/municipalities in China, including Beijing, Tianjing, Hebei, Henan, Shandong, Shanxi, Shaanxi, Inner Mongolia, Xinjiang, Jiangsu, Anhui, Fujian, Hubei and Guangzhou, which negatively impacts the health of ~31 million people (Liu et al., 2022; Wang et al., 2018; Xue et al., 2022). Thus, mapping the spatial distribution of high-iodine groundwater across China is critical to securing groundwater sustainability and public health for such a densely populated country.

In groundwater systems, iodine mainly exists in the form of iodide (I^-), iodate (IO_3^-), and organic iodine (OI). The pH and redox conditions control the iodine species in groundwater. Due to the low adsorption coefficient, I^- is often dominant in groundwater, while IO_3^- and OI are the main species in the sediments (Kodama et al., 2006; Wang et al., 2019a). Organic matter-rich iron mineral is the primary host of iodine in sediments (Li et al., 2013; Xue et al., 2024b). The iodine concentration in groundwater generally increases from the recharge to discharge area of regional groundwater flow (Li et al., 2022a; Li et al., 2022b). Some hydrogeological processes can elevate the concentration of iodine in groundwater, e.g. the evaporative concentration effect (Jia et al., 2018; Wang et al., 2021), the release of iodine-rich porewater from sediment compaction (Xue et al., 2019), and the reductive dissolution of iodine-bearing iron minerals (Li et al., 2022a; Li et al., 2020a). Microbial functional populations including dissimilatory IO_3^--reducing bacteria (Jiang et al., 2023b), Fe(III)-reducing bacteria (Guo et al., 2022b), sulfate-reducing bacteria (Jiang et al., 2023c) and dehalogenating microorganisms (Yeager et al., 2017), are able to mediate the transformation of solid phase IO_3^- and OI to the more mobile I^-. In addition, some reductants such as Fe(II) and sulfide can chemically reduce IO_3^- to I^- (Councell et al., 1997; Jiang et al., 2023a). These hydrogeological and biogeochemical processes jointly control the iodine levels in groundwater.

In addition to earlier work on goiter related to drinking high-iodine groundwater, systematic research on iodine behavior in groundwater systems has been carried out only in the past decade, with predominant efforts made in China. This review will first describe the spatial distribution of geogenic high-iodine groundwater throughout China and the sources and hosts of iodine, then discuss hydro-biogeochemical processes controlling the mobilization and enrichment of iodine in groundwater, and finally provide the future directions of studies on high-iodine groundwater.

2. Spatial distribution of high-iodine groundwater in China

2.1. Investigated areas of high-iodine groundwater

In China, high-iodine groundwater (>100 μg/L) is widely distributed in inland basins/plains such as in the Tarim Basin (Sun et al., 2022), Hetao Basin (Li et al., 2022b), Datong Basin (Li et al., 2013), Taiyuan Basin (Tang et al., 2013), Guanzhong Basin (Duan et al., 2016), Wei River Basin (Duan et al., 2020) and Jianghan Plain (Xu et al., 2023), and coastal areas such as the North China Plain (NCP) (Li et al., 2017), Luxi Plain (Zhi et al., 2023), Yishusi Plain (Wei et al., 2021), Yangtze River Delta (Zhao et al., 2017), Pearl River Delta (Huang et al., 2020) and the Yuehai Plain (Wang et al., 2018) (Fig. 1A). Although high-iodine groundwater was reported in the areas with humid (e.g., Jianghan Plain) and subhumid (e.g., Luxi Plain) climate, it mainly occurs in arid/semi-arid basin/plains with high densities of population, where

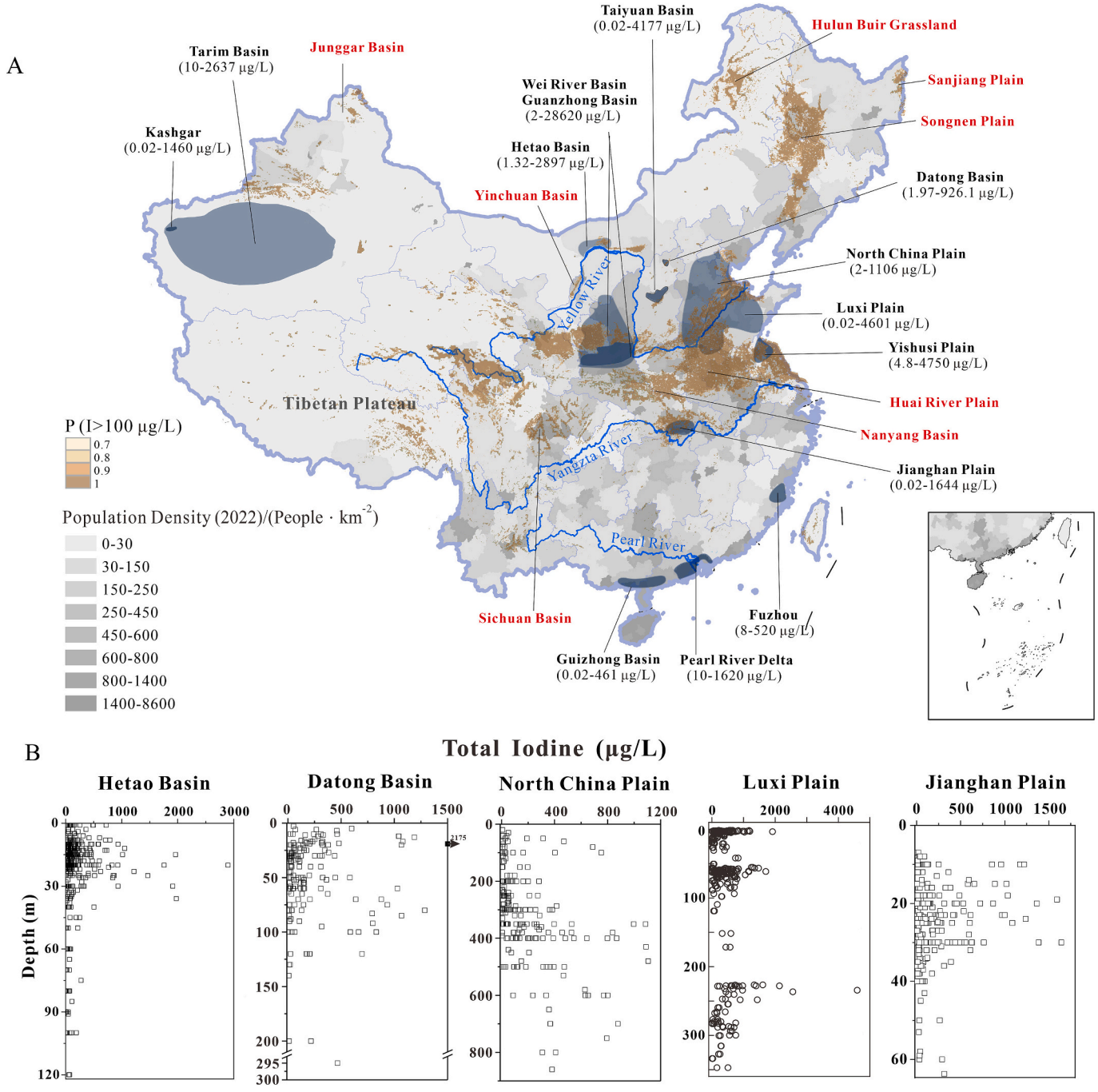

Fig. 1. Distribution of investigated (blue patches, black fonts) and predicted (brown patches, red fonts) areas of high-iodine groundwater (>100 μg/L) throughout China (A), and depth profiles of groundwater iodine at the Hetao Basin, Datong Basin, North China Plain (NCP), Luxi Plain and Jianghan Plain (B). The distribution of high-iodine groundwater was overlapped on the map of China's population density. The data of investigated high-iodine groundwater was collected from publications (Duan et al., 2016; Duan et al., 2020; Huang et al., 2020; Li et al., 2022a; Lin et al., 2023; Sun et al., 2022; Tang et al., 2013; Wang et al., 2019b; Wang et al., 2018; Wei et al., 2021; Wu et al., 2023; Xu et al., 2023; Zhi et al., 2023). The data of predicted high-iodine groundwater was from our previous work on predicting the distribution of high-iodine groundwater (Liu et al., 2022). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

groundwater serves as the main source for drinking, irrigation and industry purposes. The highest iodine concentration detected in groundwater was 28,620 μg/L in the arid Guanzhong Basin (Duan et al., 2016). As shown in Fig. 1B, high-iodine groundwater was observed in both shallow and deep confined aquifers, especially for coastal plains, such as the NCP, Luxi Plain and Yishusi Plain. In the coastal areas, shallow and deep groundwater are affected by marine-derived iodine during modern seawater intrusion and historical marine transgression, with iodine concentrations up to 4750 μg/L in the Yishusi Plain (Wei et al., 2021) and 4601 μg/L in the Luxi Plain (Wu et al., 2023), respectively. The high-iodine groundwater was mainly developed from thick Quaternary aquifers with alluvial-lacustrine, fluvial-lacustrine or alluvial-marine deposit, and sediment iodine derived from terrestrial and marine facies serves as the source of groundwater iodine, which will be presented in the "Sources and hosts of iodine in aquifers" section below.

In terms of hydrogeological setting, high-iodine groundwater was mainly detected in the discharge area of these aquifers, with Na-HCO_3 and Na-Cl(•SO_4) type groundwater with elevated total dissolved solids (TDS) under reducing conditions. I^-, IO_3^- and OI coexists in the groundwater, with I^- as the dominant iodine species. It was found that the percentage of I^- to total iodine in groundwater was commonly higher than 60 % in the Datong Basin, Hetao Basin, NCP and Jianghan Plain (Li et al., 2022a; Xue et al., 2024a; Ye et al., 2024). Notably, high levels of IO_3^- were detected in the groundwater of several regions, which may be attributed to variation of redox conditions (Li et al., 2022a). The OI concentrations in groundwater are relatively low, which originates from the migration of OI from aquifers sediments and the iodination of dissolved organic matter (DOM) by reactive iodine species during microbial IO_3^- reduction (Xue et al., 2024a).

2.2. Predicted areas of high-iodine groundwater

Although high-iodine groundwater has been reported in some basin/plains, its distribution and the relevant public health risks in most areas of China remains unknown. Based on ~4600 data of groundwater iodine reported above and the 22 environment factors, our previous study used the machine learning method (artificial neural networks) to predict the nationwide occurrence of high-iodine groundwater in China with a resolution of 1-km (Liu et al., 2022). The highly uncertainty areas of prediction map with the standard deviation of 0.4–0.5 covered ~3.7 % of China, mainly located in the Songnen Plain, Yangtze River Delta and Sichuan Basin, where less groundwater data was available for the model training. The predicted results showed that in addition to previously investigated basin/plains, high-iodine groundwater probably occurs in Hulun Buir, Sanjiang Plain, Songnen Plain, Nanyang Basin, Junggar Basin, and Sichuan Basin, the sedimentary of which is commonly characterized by alluvial/fuvial/pluvial deposits (Fig. 1A). Coincidentally, the updated data of groundwater iodine concentration measurement subsequently confirmed the presence of high-iodine groundwater in Hulun Buir, Songnen Plain and Junggar Basin (Ma et al., 2022). The 12 factors related to climate, groundwater and sedimentary environments extracted from the 22 environment factors dominantly control the occurrence of iodine-rich in groundwater, and it is consistent with the field observation that high iodine groundwater generally occurs in the Late Pleistocene-Holocene sedimentary aquifers with reducing conditions under the arid/semi-arid climate. As shown in Fig. 1A, high-iodine groundwater mostly occurs in the eastern part of China, with the elevation less than 500 m and the 94 % of Chinese population, highlighting the urgent need of nationwide investigation on high-iodine groundwater to mitigate negative health effects of potential exposure to excessive iodine uptake. Our previous work predicted that approximately 31 million population in China is under high risk of exposure to high-iodine groundwater (Liu et al., 2022).

3. Sources and hosts of iodine in aquifers

Iodine is an ultra-trace element in the Earth's crust and its mean concentration is 0.25–0.30 mg/kg (Fuge, 1988; Muramatsu and Hans Wedepohl, 1998). Unlike the lithophile elements such as arsenic and fluorine whose primary sources in aquifers come from rock weathering,

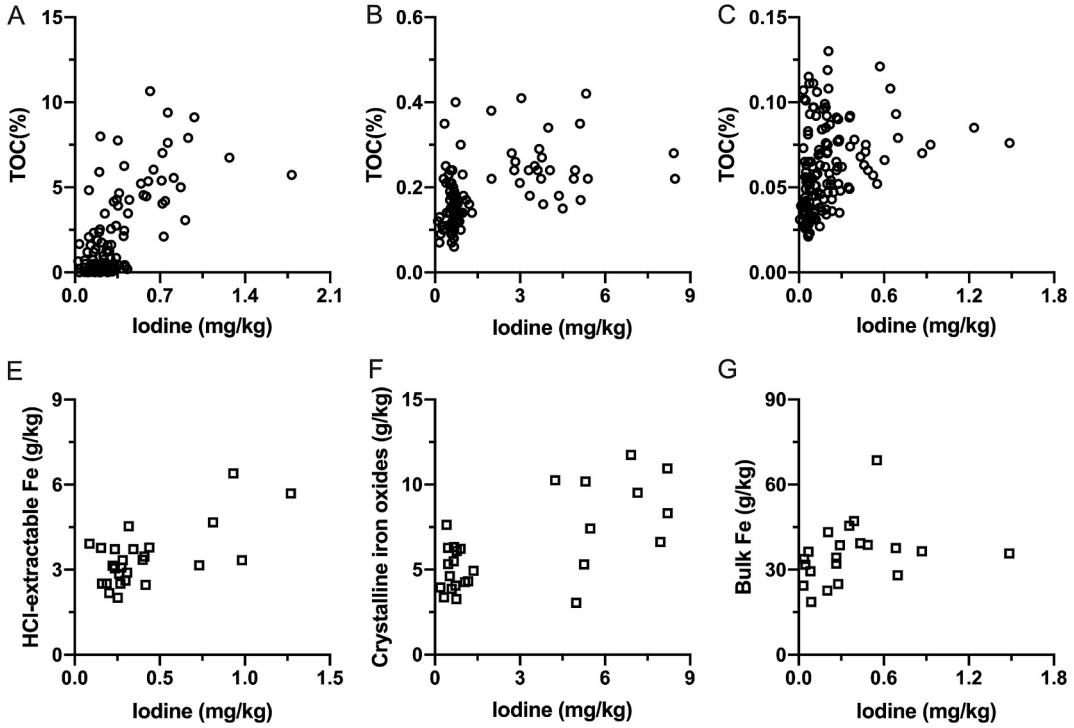

Fig. 2. Relation between iodine and TOC or different types of Fe concentrations in the borehole sediment samples from the Datong Basin (A, E), Hetao Basin (B, F) and NCP (C, G). Data in this figure are collected from Li et al. (2022a) and Ye et al. (2024). Note that only a small number of representative sediment samples from the boreholes were selected to determine iron concentrations.

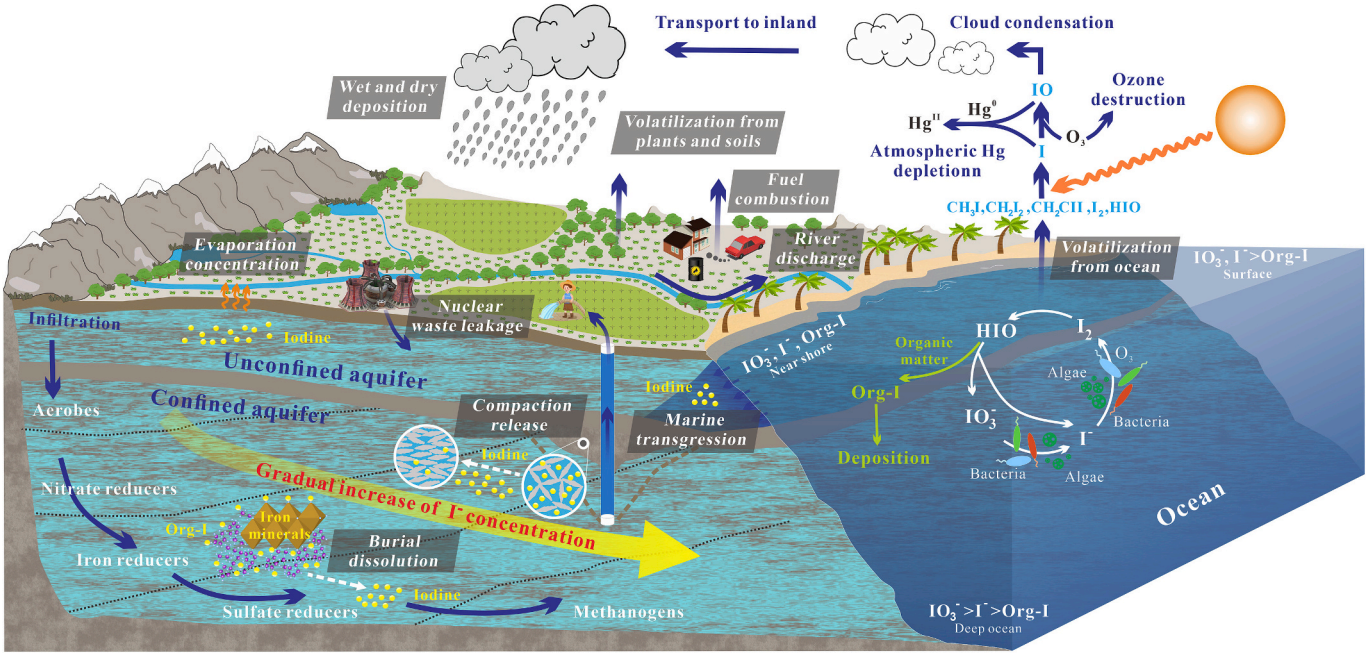

Fig. 3. Global iodine cycling and three hydrogeological processes (evaporation concentration, compaction release and burial dissolution) proposed for iodine enrichment in groundwater.

river transport and downstream deposition (Fendorf et al., 2010; Fuge, 2019), iodine is a biophile element and concentrated in sedimentary organic matter derived from marine plankton and terrestrial plants (Kennedy and Elderfield, 1987). Oceanic sediment and continental sediment rocks are the major reservoirs of iodine, accounting for 68.2 and 27.7 % of iodine in the Earth's crust (Muramatsu et al., 2004). In natural environments, organic matter interacts physically and chemically with iron minerals to form iron mineral-organic matter associations (Dong et al., 2023). Thus, organic matter-rich iron minerals are the primary hosts of iodine in aquifers, which was supported for example by the observed positive relation between iodine and TOC or different types of iron concentrations in borehole sediment samples from the Datong Basin, Hetao Basin and NCP (Fig. 2). It was found that the iodine content of sandy aquifer sediments is less than 1.0 μg/g, while the iodine content of organic matter-rich clayey sediments is as high as 1.78 μg/g at the Datong Basin and 8.46 μg/g at the Hetao Basin (Li et al., 2022a; Ye et al., 2024). To date, all reported high-iodine groundwater throughout China is of geogenic origin, with no reports about radioactive iodine contaminated groundwater in areas of nuclear waste disposal such as Hanford and Savannah River sites in the United States (Kaplan et al., 2014; Neeway et al., 2019).

Different from inland aquifers, coastal aquifers can be affected by marine-derived iodine. With an averaged iodine concentration of 60 μg/L, seawater is the most active iodine pool (Fig. 3). The bioaccumulation of iodine by marine plankton and subsequent sedimentation form the largest iodine reservoir, i.e., oceanic sediments (Kennedy and Elderfield, 1987). The volatilization of iodine from seawater and its transport through the atmosphere onto the continents are the most important pathways of global iodine cycling, which not only cause ozone destruction, atmospheric mercury depletion and cloud condensation, but also lead to the heterogeneity of iodine distribution in soils (Carpenter et al., 2021; Fuge and Johnson, 2015; Saiz-Lopez et al., 2012). The national-wide investigation of iodine content in the surface soil (depth < 0.25 m) indicates that the soils near the coastal areas (> 2.33 μg/g) commonly had the higher iodine content than those from the Northwest regions such as the Xinjiang and western Qinghai-Gansu (Wang et al., 2023a). More importantly, in geologic history, sea-level changes triggered several events of marine transgression and regression, resulting in iodine from the ocean being retained in coastal aquifers. As a result, the higher iodine content up to 28.3 μg/g was mainly observed in the sediment along the Chinese coastline (Wang et al., 2023a). In the NCP, the sediments of marine facies had higher iodine content (up to 3.82 μg/g) than those of terrestrial deposits with iodine content of less than 1 μg/g (Xue et al., 2022). Therefore, the sediments deposited during marine transgression have been considered as the main source of groundwater iodine in coastal aquifers (Wei et al., 2021; Xue et al., 2019). Additionally, due to groundwater over-exploitation in coastal areas with high population densities, seawater intrusion directly carries marine iodine into the shallow groundwater (Zhang et al., 2013).

4. Hydrogeological processes responsible for iodine enrichment in groundwater

Natural hydrogeological processes in aquifers, such as intense evaporation and progressive development of anoxic conditions, promote the enrichment of iodine in groundwater through the concentration and desorption of iodine and the reductive dissolution of iodine-rich iron minerals. The slow groundwater flow rates in basins/plains allow a long time of water-rock interaction, which is favorable for the release and accumulation of iodine in groundwater. Groundwater over-pumping for domestic drinking and agricultural irrigation cause the compaction of interbedded clays and thus expel pore water containing iodine or iodine-mobilizing solutes to adjacent aquifers (Mcmahon et al., 2011; Xue et al., 2019). Based on the characteristic occurrence patterns, we proposed three main hydrogeological processes for the genesis of high-iodine groundwater in China (Fig. 3) (Wang et al., 2021).

4.1. Evaporation-concentration

Under semi-arid/arid climatic conditions, evaporation is generally at least 10 times higher than precipitation (Jia et al., 2018). The shallow groundwater in these climatic zones undergoes strong evaporation and thus dissolved iodine can be enriched in the groundwater (Fig. 3). Furthermore, strong evaporation leads to higher salinity and concentrations of other competing ions in groundwater, which promote the

desorption of iodine from the sediments and rocks (Jia et al., 2018; Nagata and Fukushi, 2010). Under the strong evapotranspiration, the high-iodine groundwater in shallow aquifers is commonly characterized by Na—Cl and/or Na-SO_4 type water, and groundwater iodine had a positive correlation with groundwater TDS or Cl concentrations (Duan et al., 2016; Li et al., 2016a; Tang et al., 2013). It was found that the slope of the δ^2H_{H2O}-$\delta^{18}O_{H2O}$ plot in high-iodine groundwater of the Hetao Basin and the Datong Basin was lower than that in low-iodine groundwater, suggesting the evident effects of evaporation on iodine enrichment in groundwater (Li et al., 2016b; Ye et al., 2024). The evaporation-concentration role in iodine enrichment prevalently occurs in shallow groundwater systems of inland basins, China, such as Datong Basin, Hetao Basin, Taiyuan Basin and Guanzhong Basin, and have also been proposed to contribute to the enrichment of iodine in groundwater of the La Pampa aquifer, Argentina and Tindouf Basin, Algeria (Fuge and Johnson, 2015).

4.2. Compaction-release

Excessive groundwater extraction from aquifer systems causes pore water pressure to decrease and effective stress to increase (Mahmoudpour et al., 2016). The increase in effective stress results in the compaction of both the aquifer matrices and adjacent clayey layers. Compared to aquifer units consisting primarily of sand, aquitard units consisting primarily of clay and silty clay have higher compressibility and greater compaction (Calderhead et al., 2011). The compaction of aquifer systems is often manifested by regional land subsidence, which has been reported in the NCP (Gong et al., 2018) and Datong Basin (Zhao et al., 2011) as well as many other areas of the world (Galloway and Burbey, 2011; Mahmoudpour et al., 2016). It should be noted that the decrease in pore water pressure in coastal aquifers can lead to seawater intrusion and directly introduce marine-derived iodine into aquifers (Zhang et al., 2013).

Natural sea transgression and regression as well as anthropogenic seawater intrusion allow marine iodine to be persevered or tapped in aquifers. During the compaction of aquifers, the pore water containing iodine or other iodine-mobilizing solutes (such as dissolved organic carbon and competing ions) is squeezed out from the confining clays to adjacent aquifers (Fig. 3). Our previous compaction experiments of clayey and silty sediments of the NCP showed the pore solution has an iodine concentration up to 830 μg/L (Xue et al., 2019). Based on groundwater δ^2H, $^{87}Sr/^{86}Sr$ and Cl concentration, the mixing model calculation results further indicated that the compaction-released pore solution in the NCP contributed approximately 49.1–68.9 % iodine to high iodine groundwater (Xue et al., 2019). In addition to the direct release of iodine residing within pore water of clay strata, dissolved organic carbon and competing ions expulsed from aquitard units can facilitate the reductive dissolution of iodine-bearing iron minerals and the desorption of mineral-bound iodine in aquifers, respectively, thereby indirectly promoting iodine enrichment in groundwater (Mihajlov et al., 2020). Similar to iodine, this compaction-release scenario was also reported to contribute to the enrichment of other geogenic hazardous elements such as arsenic and fluoride in aquifers where groundwater was overexploited (Erban et al., 2013; Li et al., 2020b; Smith et al., 2018; Xiao et al., 2021).

4.3. Burial-dissolution

As the main host of iodine in aquifers, marine/terrestrial-derived organic matter or organic matter-bound iron minerals were deposited as sediments over geological time. The clayey sediments transported by rivers serve as a cover layer and facilitate the development of reducing conditions in the underlying aquifers (Wei et al., 2021). Under favorable conditions, microbial-mediated reductive dissolution of iron minerals and decomposition of organic matter can release the iodine from fine-grained sediments into groundwater (Fig. 3) (Wang et al., 2018). The release of adsorbed iodine into the solution along with microbial iron (III) reduction had been documented in microcosm experiments with in-situ sediments from the Datong Basin (Li et al., 2020a) and NCP (Li et al., 2022a) as well as with radioiodine-contaminated sediments (Guido-Garcia et al., 2015). As redox conditions evolve in the aquifer (Fig. 3), various types of microbial functional populations can participate in the reductive dissolution of iodine-bearing iron(III) minerals as well as the conversion of solid phase IO_3^- and OI to more mobile I^-, which will be summarized in detail in the "Biogeochemical processes responsible for iodine enrichment in groundwater" section below. The organic matter as well as its degradation intermediates deposited in the aquifer act as electron donors and carbon sources for these microbial functional populations to promote the release of adsorbed iodine through reductive dissolution of iron minerals. High-iodine groundwater is mainly distributed in semi-closed/closed sedimentary environments with flat terrain, small hydraulic gradient and sluggish underground runoff (Fig. 1). The slow movement of water allow for a relatively long time of water-rock interaction and low migration rate of iodine, making it favorable for iodine to be liberated from sediments and enriched in groundwater (Zheng et al., 2024). Thus, burial-dissolution plays an important role in iodine enrichment in both shallow and deep groundwater systems and in both inland basins and coastal plains in aquifers of China and beyond (Wang et al., 2021; Xu et al., 2023).

5. Biogeochemical processes responsible for iodine enrichment in groundwater

Microbial transformation of iodine species is crucial for I^- production (Yeager et al., 2017). Given that IO_3^- and OI are the dominant species of iodine in aquifer sediments, IO_3^- reduction and OI degradation play vital roles in the generation and enrichment of I^- in groundwater. To date, various biogeochemical processes in aquifers could participate in the reduction of IO_3^- to I^-, such as dissimilatory IO_3^- reduction, iron (III) reduction, sulfate reduction and anaerobic oxidation of methane (Fig. 4). The iodine in OI may also be liberated into groundwater through microbial degradation or dehalogenation. The gradual increase of I^- concentration in groundwater along groundwater flow direction and redox potential gradient in the Datong Basin, Hetao Basin and NCP (Li et al., 2022a; Li et al., 2022b) should result from the combined effect of these biogeochemical processes (Fig. 3). It should be pointed out that, similar to the proposed hydrogeological processes, the biogeochemical processes summarized below play important roles in the mobilization and enrichment of iodine not only in iodine-rich groundwater in China but also around the world.

5.1. Dissimilatory IO_3^- reduction

Dissimilatory IO_3^-−reducing microorganisms (DIRM) use the periplasmic IO_3^- reductase (Idr), which consists of subunits of IdrA, IdrB, IdrP1 and IdrP2, to reduce IO_3^- to I^- (Fig. 4)(Reyes-Umana et al., 2022a; Yamazaki et al., 2020). We isolated a dissimilatory IO_3^--reducing bacterium *Azonexus hydrophilus* strain NCP973 from high-iodine groundwater of NCP for the first time (Li et al., 2024). Microcosm experiments showed that the addition of strain NCP973 to in-situ sediments could facilitate the release of I^- to the aqueous phase. Furthermore, the IdrABP1P2 was located on conjugative plasmid, implying that its IdrABP1P2 might be transmitted by horizontal gene transfer and allow recipient microorganisms to participate in the enrichment of I^- in aquifers (Reyes-Umana et al., 2022b). Another dissimilatory IO_3^--reducing bacterium, *Azoarcus* sp. DN11, was also isolated from a gasoline-contaminated aquifer in Kumamoto, Japan (Sasamura et al., 2023). Groundwater metagenomic analyses further indicated that *idrA* was widely detected in geogenic high-iodine groundwater of Hetao Basin, Datong Basin, Yingchuan Basin and NCP (Jiang et al., 2023b; Li et al., 2024). These results suggest that high-iodine groundwater systems are important habitats for DIRM, and their activities in aquifers promote

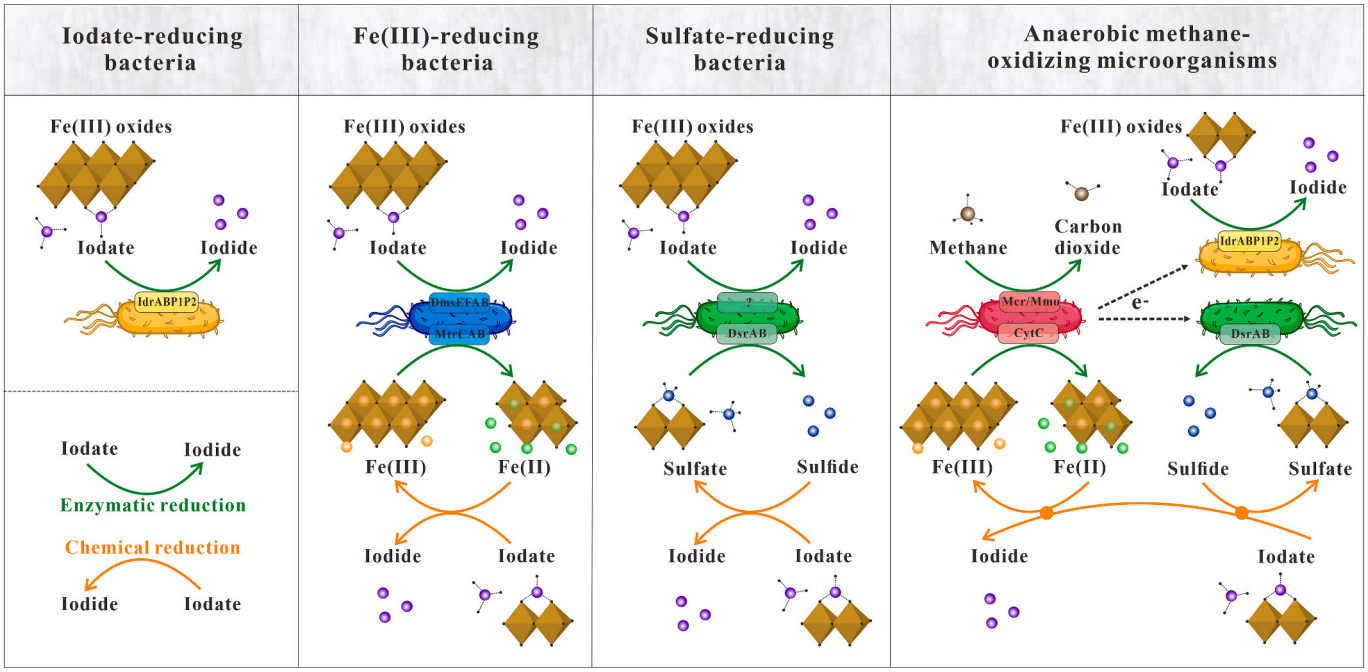

Fig. 4. Enzymatic or chemical reduction of mineral-bound or dissolved IO_3^- to I^- mediated by different types of microbial functional populations.

the mobilization and enrichment of iodine in groundwater.

The standard redox potential of IO_3^-/I^- (+0.67 V) is in between that of the O_2/H_2O (+0.82 V) and NO_3^-/NO_2^- (+0.42 V), suggesting that IO_3^- reduction probably takes place in nitrate reduction zone in aquifers (Fig. 3) (Lam and Kuypers, 2011; Reyes-Umana et al., 2022a). It was inferred that IO_3^- can be reduced to I^- by microorganisms having a NO_3^- reductase (Hung et al., 2005; Tsunogai and Sase, 1969). Subsequent studies documented that IO_3^- reduction in these organisms did not involve a NO_3^- reductase but rather other enzymatic mechanism (Bluhm et al., 2010; Mok et al., 2018). However, the strain DVZ35 isolated from radioiodine-contaminated groundwater at Hanford, USA could reduce IO_3^- in the presence of NO_3^- but not in the absence of NO_3^- (Lee et al., 2018), suggesting that NO_3^--reducing bacteria may also contribute to IO_3^- reduction, but the molecular mechanism needs further investigation.

5.2. Iron(III) reduction

Fe(III)-reducing bacteria not only promote the release of adsorbed iodine through the reductive dissolution of iodine-rich iron minerals, but also reduce IO_3^- to I^- via abiotic reactions of IO_3^- with biogenic Fe(II) or via enzymatic reactions (Fig. 4). Previous microcosm experiments on high-iodine sediments from the Datong Basin and NCP (Li et al., 2022a) and on radioiodine-contaminated sediments near the Sellafield nuclear facility in northwest England (Guido-Garcia et al., 2015) showed that solid-phase iodine was released into the solution as I^- along with microbial Fe(III) reduction. The formation of I^- in solution should be attributed to the abiotic/biotic reduction of adsorbed or free IO_3^- triggered by Fe(III)-reducing bacteria. It was found that Fe(II) reacts with IO_3^- to form Fe(III) and I^-, with HIO as the intermediate (Councell et al., 1997; Jiang et al., 2023a). All types of biogenic Fe(II), such as dissolved Fe^{2+}, surface-adsorbed Fe(II) and mineral structural Fe(II) produced by the reduction of Fe(III)-citrate, Fe(III) oxides and clay minerals, were able to chemically reduced IO_3^- (Jiang et al., 2023a). Meanwhile, the dissimilatory Fe(III)-reducing bacterium *Shewanella oneidensis* MR-1 can directly employ the dimethylsulfoxide reductase DmsEFAB and metal reductase MtrCAB for biological IO_3^- reduction (Guo et al., 2022b; Shin et al., 2022). Comparative genomic analysis showed that *dmsEFAB* and *mtrCAB* gene clusters coexist in many other Fe(III)-reducing bacteria, some of which have been demonstrated to perform IO_3^- reduction (Farrenkopf et al., 1997; Guo et al., 2022a). Incubation experiments of IO_3^- reduction of MR-1 wild type and mutants in the presence of different amounts of Fe(III)-citrate, Fe(III) oxides, or clay minerals further revealed that chemical IO_3^- reduction by biogenic Fe(II) predominated under iron-rich conditions, while biotic IO_3^- reduction by DmsEFAB played a more dominant role under iron-poor conditions (Jiang et al., 2023a). Natural and artificial processes such as surface water-groundwater interaction, riverbank filtration treatment, and over-pumping disturb the redox conditions of aquifers. Under redox fluctuation conditions, biogenic Fe(II) reacts with O_2 to generate hydrogen peroxide (H_2O_2) and hydroxyl radicals (•OH), which can inversely oxidize I^- to I_2 and further to HIO, followed by disproportionation of HIO to IO_3^- and I^- (Li et al., 2012; Luther, 2023). Recent incubation experiments with MR-1 and Fe(III)-containing minerals under altered redox conditions showed that I^- oxidation by reactive oxygen species (ROS) under oxic conditions did not lead to significant production of IO_3^- compared with the amounts of I^- formed under anoxic conditions (Zhu et al., 2024). Consequently, the iron redox processes under redox fluctuating conditions drive the successive reduction of IO_3^- to I^-, thus highlighting the vital role of Fe(III)-reducing bacteria in I^- generation and mobilization in aquifers.

5.3. Sulfate reduction

Similar to Fe(III)-reducing bacteria, sulfate-reducing bacteria (SRB) also mediate the release of adsorbed iodine through the reductive dissolution of iodine-rich iron minerals and the reduction of IO_3^- to I^- via biogenic sulfide or via specific enzymatic reactions (Fig. 4). Previous studies showed that iron oxides and clay minerals were directly reduced by SRB, such as *Desulfovibrio vulgaris, Desulfovibrio putealis* and *Desulfovibrio desulfuricans* strain G-20, or indirectly reduced by biogenic sulfide (Hansel et al., 2015; Li et al., 2007; Liu et al., 2012). Moreover, the reduced sulfur species, such as sulfide and cysteine, were found to chemically reduce IO_3^- to I^- (Hird and Yates, 1961; Zhang and Michael, 1986). In addition, some SRB directly reduce IO_3^- to I^- using an unknown enzyme that probably belong to the DMSO reductase family (Councell et al., 1997; Jiang et al., 2023c). The concentrations of I^- and total iodine in deep groundwater of NCP were positively correlated with

groundwater $\delta^{34}S_{SO4}$, *dsrB* gene abundance and SRB abundance, respectively, indicating that SRB could promote iodine mobilization and I^- enrichment in the high-iodine groundwater of NCP (Jiang et al., 2023c). Given the similar roles in iodine mobilization and their ubiquity in aquifers (Bethke et al., 2011), microbial iron reduction and sulfate reduction should be the main driver for the enrichment of I^- in aquifers in China and other countries. Given the thermodynamic ladder, groundwater passes through a series of redox zones in which electron acceptance yields progressively less energy. Thus, the spatially successive distribution of nitrate reduction, Fe(III) reduction, sulfate reduction, and methanogenesis zones in aquifers suggested that different microbial functional populations alternatively contribute to the production and accumulation of I^- in groundwater, which was consistent with the observation that I^- concentration in groundwater gradually increased along the groundwater flow direction (Li et al., 2022a; Li et al., 2022b). However, under some circumstances, Fe(III) reducer and sulfate reducer do not compete for energy but enter into a tightly balanced mutualistic relationship (Bethke et al., 2011), suggesting the enrichment of I^- by the coupled biogeochemical processes of multiple elements.

5.4. Anaerobic oxidation of methane

Groundwater with high level of iodine is generally found in the discharge area where methane generation and anaerobic oxidation of methane (AOM) occur (Glodowska et al., 2020; Li et al., 2022a; Tian et al., 2023). It has been reported that AOM can be coupled to the reduction of various electron acceptors including sulfate, NO_3^-, NO_2^-, Fe (III)/Mn(IV) oxides, SeO_4^{2-}, ClO_4^-, BrO_3^-, $Cr_2O_7^{2-}$ and $HAsO_4^{2-}$ (Bhattarai et al., 2019; Cai et al., 2021). Considering the similar redox potentials of IO_3^-/I^- and NO_3^-/NO_2^- and energetics of reactions (1), we propose that AOM in aquifers may also be directly coupled to the reduction of IO_3^- to the more mobile I^- (Fig. 4), which is similar to reduction of ClO_4^- and BrO_3^- (Luo et al., 2017; Luo et al., 2015). The genome of Candidatus *Methylomirabilis iodofonti* obtained from underground iodine-rich cavern water of Sulzbrunn in Germany was shown to contain the key genes of methane oxidation (*mcrA*) and IO_3^- reduction (*idrA*), ascertaining the potential of methane oxidation coupled to IO_3^- reduction (Zhu et al., 2022).

$$CH_4 + 4/3\ IO_3^- = CO_2 + 4/3\ I^- + 2\ H_2O, \triangle G^o = -716\ kJ/mol\ CH_4 \quad (1)$$

In addition, AOM coupled to the reduction of Fe(III) or sulfate in aquifers have been demonstrated by in-situ sediment microcosms (Pienkowska et al., 2021; Timmers et al., 2016). Their activities in aquifers not only facilitate the release of adsorbed iodine through the reductive dissolution of iodine-rich iron minerals, but also the reduced products, such as sulfide and Fe(II), can chemically reduce dissolved or mineral-bound IO_3^- to I^- (Fig. 4). The characteristic microbial populations of AOM were detected widely in aquifers (Cai et al., 2021), including geogenic high-iodine groundwater (Tian et al., 2023) and high-arsenic groundwater (Glodowska et al., 2020). Therefore, IO_3^--, Fe (III)- or sulfate-dependent methane oxidation in aquifers may be an important pathway for iodine mobilization in the methanogenesis zone, highlighting the role of methane as a driver of iodine mobilization in aquifers.

5.5. Organic-iodine degradation

As another major iodine species in aquifer sediments, OI degradation or dehalogenation can liberate bound iodine into groundwater. It was found that during organic matter degradation in marine and estuarine sediments, iodine is released into the solution as I^- (Kennedy and Elderfield, 1987; Scholz et al., 2024; Upstill-Goddard and Elderfield, 1988). Sulfate-reducing bacteria, such as *Desulfitobacterium* spp. and *Desulfoluna spongiiphila* AA1, were reported to deiodinate 5-amino-2,4,6-triiodoisophthalic acid, 2-iodophenol or 3-iodophenol to I^- (Ahn et al., 2009; Lecouturier et al., 2003). Given that many SRB strains from the genera *Desulfitobacterium*, *Desulfomonile*, *Desulfuromonas* and *Desulfovibro* are well-known dehalogenating bacteria, and have been detected in high-iodine groundwater of the NCP, we propose that SRB is involved not only in IO_3^- reduction but also in OI dehalogenation (Jiang et al., 2023c; Mohn and Tiedje, 1992; Villemur et al., 2006). Bond strength decreases markedly with increasing molecular weight: C-F > C-Cl > C-Br > C-I, which suggest that other organohalide-respiring bacteria, especially organochlorine-respiring bacteria, may also play a role in the dehalogenation of OI in aquifers (Adrian and Löffler, 2016; Smidt and de Vos, 2004). To date, no specific enzymes responsible for OI dehalogenation was reported. The OI degradation in aquifers may proceed via the reductive deiodination, as revealed in *Vallitalea* sp. strain TIP-1 isolated from the marine sponge (Oba et al., 2014) and sediment enrichments collected from an iodine-producing industry, Chiba, Japan (Kawamura et al., 2021). In addition to direct released of I^- from OI dehalogenation, low-molecular weight organic compounds derived from OI degradation in aquifers can serve as carbon sources and electron donors to fuel specific functional microorganisms to mediate the reductive dissolution of iodine-bearing iron minerals and the reduction of IO_3^- to I^- (Xue et al., 2022; Zhao et al., 2023).

6. Perspectives

Despite substantial progresses made over past decades in the research of high-iodine groundwater in China, crucial knowledge gaps remain and the great efforts on the following five aspects (Fig. 5) are needed to improve our understanding of iodine behavior in the subsurface, to find science-based solutions for sustainable safe supply of groundwater resources and to better understand the global iodine cycling.

6.1. Investigation of radioiodine-contaminated groundwater

To date, all known high-iodine groundwater in China is of geogenic origin. Anthropogenic high-iodine groundwater, such as groundwater contaminated by radioactive iodine from nuclear wastes, has been observed in the Hanford and Savannah River sites of USA (Kaplan et al., 2014; Neeway et al., 2019). ^{131}I and ^{129}I are the main radioactive iodine, both of which could negatively affect human health and groundwater quality (Jiang et al., 2024; Wei et al., 2024a, 2024b; Yeager et al., 2017). The anthropogenic sources of radioiodine include nuclear weapons program, nuclear weapons testing, nuclear power plants, uranium mining and milling, commercial fuel reprocessing, geological repository of nuclear wastes and nuclear accidents (Hu et al., 2010; Wang et al., 2024). With the perspective of reducing the reliance on fossil fuels and the emissions of greenhouse gases, nuclear power is undergoing rebirth. It is estimated that more than 100 nuclear power plants will be built and in operation by 2030 in China and the nuclear power generation is expected to reach 8–10 % of the total power generation capacity in China (Xia et al., 2021). A large amount of nuclear waste will need to be disposed. Therefore, to ensure water security, it is necessary to conduct a nationwide hydrogeochemical investigation and long-term monitoring of groundwater iodine at these sites of nuclear-related activities and nuclear waste disposal repositories. Furthermore, the migration, transformation and attenuation of radioiodine in groundwater at those sites and its long-term impact on human health as a drinking or irrigation water resource should be evaluated.

6.2. Identification and quantification of OI species

I^- and dissolved organic matter in drinking water react with disinfectants to form OI disinfection byproduct, whose cytotoxicity and genotoxicity are more than 100 times higher than those of organochlorines (Sharma et al., 2023; Yang et al., 2014). In high-iodine

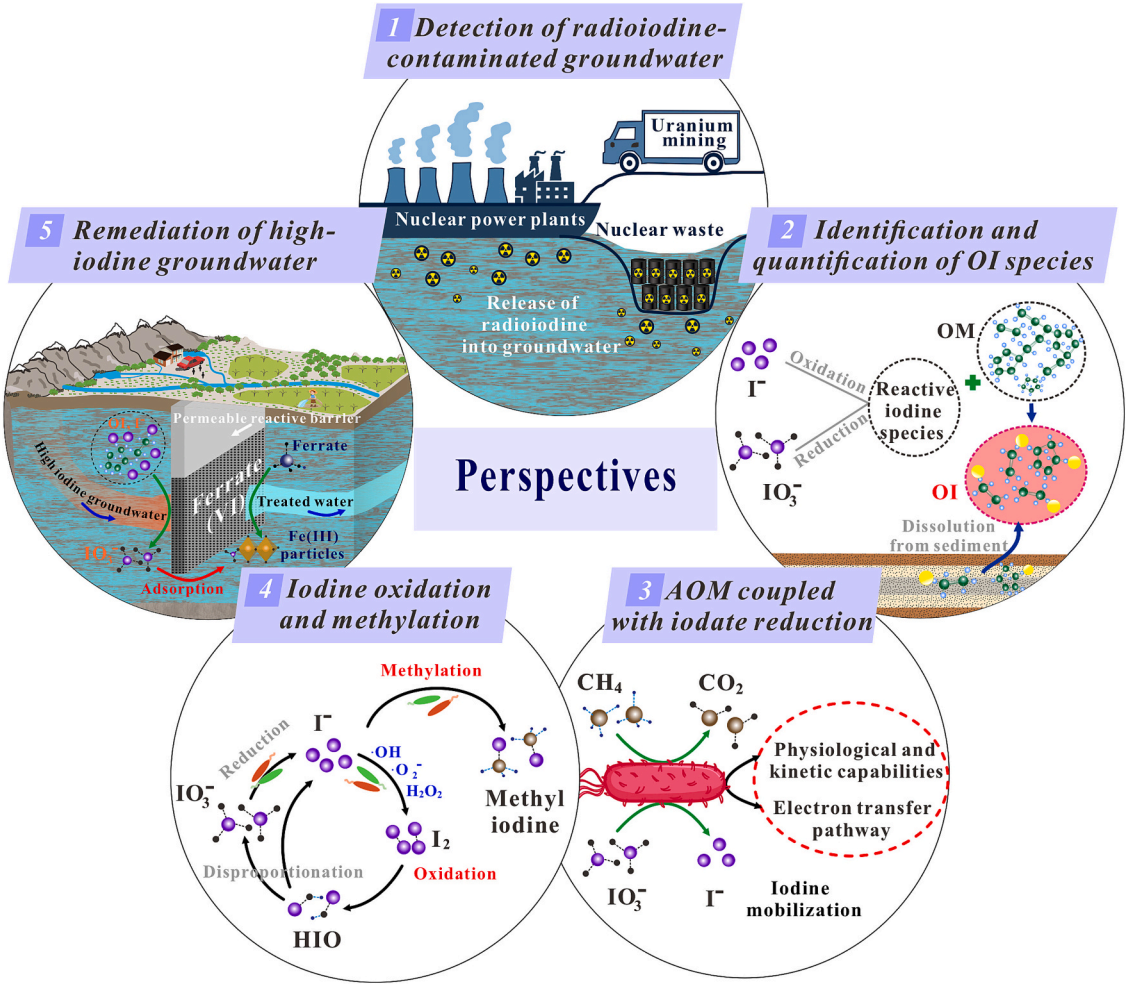

Fig. 5. Five perspectives regarding future research on high-iodine groundwater.

groundwater of China, OI coexists with I^- and IO_3^-, with the highest and mean concentrations of OI of ~300 μg/L and ~ 20 μg/L, respectively (Li et al., 2014; Li et al., 2017; Xue et al., 2024a). The OI in groundwater is generated from the migration from sediment-bound OI and the iodination of dissolved organic matter by reactive iodine species formed during IO_3^- reduction and I^- oxidation (Xue et al., 2024a; Yeager et al., 2017). Recently, we identified a total of 939 OI compounds in groundwater of Jianghan Plain using Fourier transform ion cyclotron resonance mass spectrometry (FT-ICR MS) (Xue et al., 2024a). In light of cytotoxicity and genotoxicity of OI, the identification and quantification of OI species should be as important as the detection of total iodine and inorganic iodine species in hydrogeochemical studies on high-iodine groundwater.

6.3. Characterization of AOM coupled with IO_3^- reduction

AOM plays a crucial role in mitigating methane emissions from anoxic subsurface environments and the biogeochemical cycle of coupled elements (Cai et al., 2021; Caldwell et al., 2008). Although it is thermodynamically feasible, the hypothesized of AOM coupled to IO_3^- reduction needs to be demonstrated experimentally using enrichment cultures and isolated microbial strains. Determination of physiological and kinetic capabilities and molecular mechanism of electron transfer pathway can be revealed with the isolation of the type of microorganisms. Furthermore, the relative contribution of AOM coupled to the reduction of different electron acceptors (IO_3^-, Fe(III) and sulfate) to iodine mobilization in aquifers is not clear. Microcosms and field investigations combined with isotopic analysis and multi-omics techniques will allow the identification of AOM populations, the quantification of respective AOM rates, and the contribution of AOM to iodine mobilization. Seawater is enriched in IO_3^- and more than 90 % methane in the ocean is consumed by AOM (Carpenter et al., 2021; Knittel and Boetius, 2009). The characterization of IO_3^--dependent AOM in aquifers will provide new clues to understanding the effects of iodine biogeochemical cycling driven by AOM for global methane emissions.

6.4. Iodine oxidation and methylation

I^- can be oxidized by I^--oxidizing bacteria or reactive oxygen species to I_2, and then I_2 is hydrolyzed to hypoiodous acid (HIO) and I^-, and finally HIO is disproportionated to IO_3^- and I^- (Amachi and Iino, 2022). The oxidation of I^- to IO_3^- was responsible for the dominance of IO_3^- in radioiodine-contaminated groundwater in Hanford and Savannah River sites (Lee et al., 2020; Li et al., 2012). Thus, I^- oxidation may occur in China's geogenic high-iodine groundwater, especially in the groundwater containing high levels of IO_3^- and at the interface between the vadose and saturated zones (Li et al., 2022a; Wei et al., 2024a, 2024b). In addition to the transformation of inorganic iodine species, microbes are capable of methylating I^- to form methyl iodine via SAM-dependent halide methyltransferases, which have been reported in rice paddies, peat bogs, coastal salt marshes and oceans (Amachi, 2008; Yeager et al., 2017). Similar to the significant contribution of methylation by SAM-dependent arsenite methyltransferases to aquifer arsenic cycling (Maguffin et al., 2015; Zeng et al., 2018), iodine methylation may play

an important role in the formation of methyl iodine in groundwater. Because methyl iodine is more volatile and prone to diffusion than other iodine species, iodine methylation can serve as a link between surface and subsurface iodine cycling.

6.5. Remediation of high-iodine groundwater

Knowledge about the hydro-biogeochemical processes controlling iodine mobilization and migration in iodine-contaminated aquifers is fundamental for the design and application of ex-situ/in-situ remediation method. Novel cost-effective treatment technologies are needed in areas where iodine-rich groundwater serves as drinking water source. Compared to various adsorption materials designed for ex-situ treatment (Moore et al., 2020; Patil et al., 2023), in-situ remediation of iodine-contaminated groundwater remains unexplored. Recently, it was reported that ferrate(VI) could oxidize organic and inorganic iodine to IO_3^- and simultaneously remove the resulting IO_3^- through adsorption to in-situ formed Fe(III) particles from ferrate(VI) reduction (Wang et al., 2023b). Similar to our previous work of in-situ arsenic remediation using iron coatings (Xie et al., 2016), the oxidation and adsorption strategy using ferrate (VI) may be a promising method for in-situ remediation of high-iodine groundwater under moderately reducing conditions. However, the stability of IO_3^--adsorbed Fe(III) particles formed by ferrate(VI) reduction should be evaluated before in-situ remediation, especially under prolonged reducing conditions. Additionally, it should pay attention on the detection of contaminants coexisting with I^- in groundwater, such as Cr(III), Se(O), Tc(IV) and U (IV), which may be also oxidized by ferrate (VI) to more mobile species and cause secondary contamination.

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.

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Data availability

Data will be made available on request.

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