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Review of drivers and threats to coastal groundwater quality in China

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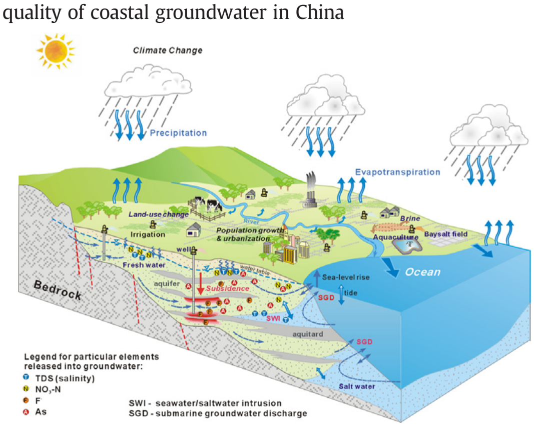
HIGHLIGHTS

- First review of groundwater quality issues and their extent in coastal China
- Analysis of data reveals primary causes of China's coastal groundwater quality issues.
- High nitrate, salinity, fluoride, and arsenic groundwater are all major challenges.
- Implications and learnings for improving coastal groundwater quality management

GRAPHICAL ABSTRACT

Processes affecting the quality of coastal groundwater in China



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ABSTRACT

With rapid socio-economic development, China's coastal areas are among the fastest growing and most economically dynamic regions in the world. Under the influence of climate change and human activities, protecting the quality of coastal groundwater has emerged as one of the key environmental and resource management issues for these areas. This paper reviews (for the first time) groundwater quality data for the coastal basins of China, where over 600 million people live, focussing on key inorganic indicators/pollutants; groundwater salinity, nitrate, fluoride, and arsenic. These pollutants present major water quality issues and are also valuable as indicators of wider processes and influences impacting coastal groundwater quality – e.g. saltwater intrusion, agricultural pollution and release of geo-genic contaminants. We discuss the major drivers causing water quality problems in different regions and assess future trajectories and challenges for controlling changes in coastal groundwater quality in China. Multiple processes, including modern and palaeo seawater/brine migration, groundwater pumping for agricultural irrigation, pollution from agrochemical application, rapid development of aquaculture, urban growth, and water transfer projects, may all be responsible (to different degrees) for changes observed in coastal groundwater quality, and associated long-term health and ecological effects. We discuss implications for sustainable coastal aquifer management in China, arguing that groundwater monitoring and contamination control measures require urgent improvement. The evolution and treatment of coastal groundwater quality problems in China will serve as an important warning and example for other countries facing similar pressures, due to climate change, coastal development, and intensification of anthropogenic activity in coming decades.

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1. Introduction

Coastal areas are the fastest growing and most economically dynamic regions in the world. As an interface between the land and oceans in the earth system, the coastal zone is abundant in natural resources and plays an important supporting role for social and economic development (Lin and Pussella, 2017). Approximately 2.4 billion people live within 100 km of the coast globally (The Ocean Conference, United Nations, 2017). Groundwater is an important source of water supply in coastal areas (including islands) and provides drinking water for nearly 1 billion people in coastal areas around the world (Small and Nicholls, 2003). Due to the high intensity of human activities and the influence of climate change, environmental issues in coastal areas, such as water pollution, soil salinization, eutrophication, seawater intrusion, land subsidence, and wetland shrinkage, are becoming increasingly serious, threatening ecosystem security (Slott et al., 2006; Zhu et al., 2012; Johnson et al., 2015; Gao and Luo, 2016). Many of these problems are attributable (or in some way linked) with groundwater utilization in the coastal zone. The physical, chemical, and biological processes across the land-ocean interface have been affected by groundwater exploitation and associated development, not only changing the hydrological balance between groundwater, surface water and seawater but also onshore groundwater and seawater quality, and ecosystems dependent on these waters.

Groundwater is vital for agricultural and domestic purposes in arid and semi-arid regions, where it may be the only freshwater source, especially during periods of peak water demand. With increasing understanding of the hydrological cycle, groundwater is considered an important link maintaining the coastal eco-hydrological environment and biogeochemical cycles (Burnett et al., 2001; Moore, 2006, 2010; Luo and Jiao, 2016; David et al., 2019). However, groundwater quality in this zone has received relatively limited attention (Treidel et al., 2012). Terrestrial nutrients, waste (such as plastics) and microbial pathogens entering the sea via surface or subsurface pathways can lead to deterioration of coastal ecosystems, resulting in food safety and human health risks (Robins et al., 2016). Offshore fresh groundwater discharge also plays a key role in biogeochemical fluxes from land to ocean (Wilson, 2003; Michael et al., 2016; Micallef et al., 2020). Coastal areas, because of their low topography, serve as discharge areas for onshore flow systems and may accumulate salts and other pollutants, integrating up-stream water quality impacts from a potentially large area. In China, this appears to have resulted in more severe water quality deterioration compared to most inland groundwater flow systems (Han et al., 2016a; Han and Currell, 2017). The intensive nature of human activities in coastal zones – including both urban development and agriculture further heightens the potential for water quality degradation.

The continental coastline of China is approximately 18,000 km long, starting from the Yalu River estuary in Liaoning at the border of China and North Korea, and stretching to the Beilun River estuary in Guangxi, at the border of China and Vietnam in the south. This coastline is shaped in an arc that bulges to the southeast and across the temperate, subtropical, and tropical zones with numerous estuaries, bays, and islands (Fig. 1). In China, the coastal provinces and cities occupy less than 15% of the country's land area; however, the population accounts for nearly 50% of the national total, and GDP nearly 60% of the national total (Luo, 2016; Peng et al., 2018). Nearly 80% of the coastal population in China is concentrated across three major economic zones (Fig. 1). There are 9 'megacities' (>10 million inhabitants) and 83 large cities (1 to 10 million inhabitants) in these coastal regions (Fig. 2).

In 2019, the total water supply utilization in China's coastal areas (including the continental coast and Hainan Island) was 218.24 billion m^3, accounting for 36.2% of the country's total water supply. Among this, groundwater utilization reached 28.3 billion m^3 (Ministry of Water Resources of China, 2020); however, nearly 90% of this total (25.3 billion m^3) was used in the coastal provinces north of the Yangtze River. The proportion of water used in agriculture exceeds 50% in Liaoning, Hebei, Shandong, Jiangsu, Guangdong, Guangxi, and Hainan provinces, while the proportion used for industry is more than 20% for Jiangsu, Zhejiang, Fujian, and Guangdong provinces. These differences in water use structure have caused different degrees of groundwater-related environmental problems.

Exploitation and utilization of groundwater resources has played an important role in sustaining socio-economic development, particularly in northern China. However, since the 1970s, with large-scale development and utilization of water and land resources, a series of environmental problems have emerged. Navigating the challenges of intensive socio-economic development pressures in coastal regions, while minimizing harm to ecological systems and safeguarding the long-term sustainability of groundwater and other water resource utilization has been (and continues to be) a major policy challenge. We believe a synthesis and review of groundwater quality data for China's coastal regions is overdue and provides an opportunity to gain important insights into the extent of water quality issues and their associated processes, at a scale that is relevant for both national and provincial level policy development. Review and analysis of current coastal groundwater quality problems in China (which has experienced rapid and sustained development pressures in recent decades) and the causative processes, provides an important reference for other coastal regions facing significant climate and development pressures (such as Southeast Asia).

The objectives of this paper are to:

(i) document (for the first time) groundwater quality problems and their extent, encompassing the whole of coastal China, through compilation and summary of the existing literature and data;
(ii) analyse and discuss the driving forces behind these coastal groundwater quality issues, encompassing saltwater intrusion, pollutant mobilization, enrichment, and transport (e.g., via submarine groundwater discharge), and their links to human activities;
(iii) draw out implications and learnings for improving coastal groundwater quality through land and water management initiatives.

This will help to inform better management strategies for groundwater-environmental problems in China and other areas facing similar pressures, which to date have not been given adequate attention in coastal regions facing rapid economic development.

2. China's coastal aquifers

The major coastal river basins of China (from north to south) are the Liao, Luan, Hai, Huang (Yellow), Huai, Yangtze and Pearl River basins (Supplementary Fig. S1). The coastal region straddles several climatic zones, from the tropical zone in Hainan Island, to subtropical south of the Yangtze River, to the temperate zone north of the Yangtze. There is a clear decreasing trend in precipitation from south to north - average annual precipitation varies from 1200 to 2000 mm in Guangxi and 1300 to 2500 mm in Guangdong, to 351 - 843 mm in Hebei, and then somewhat increasing to 800-1200 mm in Liaoning (the northernmost coastal province). Taking Hangzhou Bay as the geographic boundary between south and north, the northern coastal zone is mainly low-lying alluvial plains underlain by sandy aquifers (Fig. 2), while the southern coastal zone is mainly mountainous, with hard rock aquifers. Both regions contain localized karst (limestone) aquifers (Zhang and Li, 2005).

The aquifer systems in the coastal regions of China are comprised of multiple porous, karst and fractured bedrock aquifer systems (Fig. 3). The coastal porous aquifer systems (mainly north of the Huai River) are composed of the alluvial and deltaic sediments of the Huang-Huai-Hai Plain and Yellow River Delta (together comprising the North China Plain), as well as the Yangtze River delta. These Quaternary sediments and the paleo-channels of the rivers constitute a multi-layered aquifer-aquitard system. Several interbedded fine-grained marine

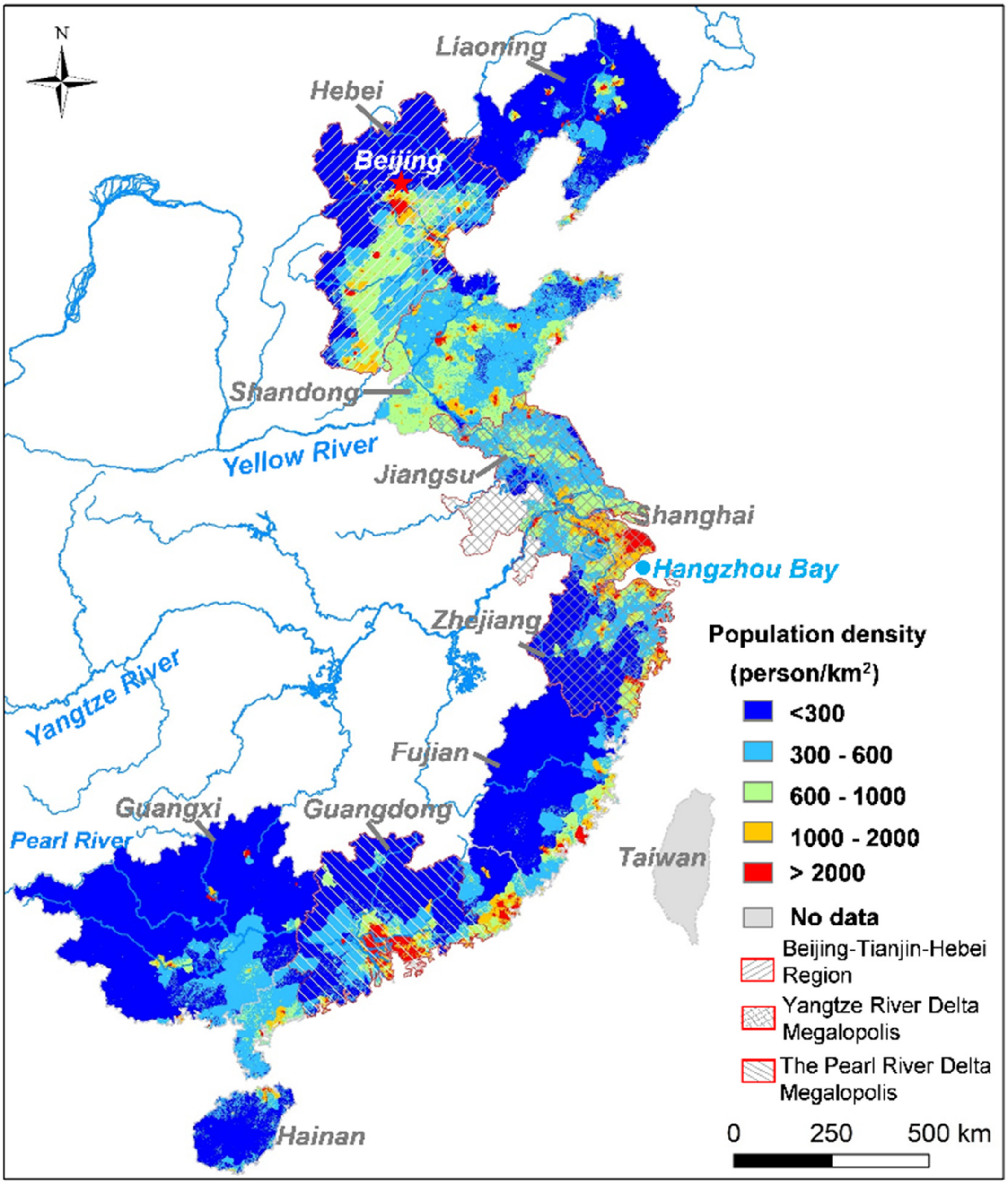

Fig. 1. Population density in coastal China. Data source: Urban Construction Statistical Yearbook, 2019.

sediments can also be found in the coastal region, which were deposited during multiple transgression events since the middle Pleistocene (Yang and Chen, 1985; Han et al., 2020). The shallow groundwater (generally <60 m below the surface) in these aquifers is generally characterized by relatively high salinity (total dissolved solids TDS concentrations >1 g/L) associated with seawater ingress during marine transgression events (Han et al., 2020). The groundwater in deeper confined aquifers generally has TDS concentrations <1 g/L and can be artesian. However, the confined groundwater has been highly developed over past decades and the groundwater levels have shown continuous decline, particularly in the coastal region of the North China Plain (Figs. S2 and S3) (Song, 2006; Cao et al., 2013).

Quaternary unconsolidated aquifers in the south of China are mainly distributed in the Yangtze River Delta and Pearl River Delta (Fig. 3). Coastal carbonate aquifers are mainly distributed in Beihai (Guangxi) in the south, and the Liaodong Peninsula, east of Bohai Bay in the north. Other areas are mostly characterized by semi-consolidated or fractured igneous rock aquifers. Cambrian and Ordovician limestone and dolomitic limestone aquifers also occur along the Shandong Peninsula. Karst development in this region is primarily controlled by tectonic structures and generally becomes less permeable with depth (Zhang and Li, 2005).

The coastal aquifers south of the Huai River are predominantly fractured rock aquifers, in Fujian and Guangdong provinces these are developed in igneous rocks including Mesozoic and Cenozoic granite and volcanics. Groundwater mainly exists in structurally controlled fracture zones and the specific capacity of the aquifer system is generally less than 0.01 m^2/s. Geothermal springs are common in the coastal regions of Fujian Province and the distribution of geothermal water is controlled by tectonic structures. In the coastal region of Guangdong Province, particularly the Leizhou Peninsula, volcanic rocks from the middle Quaternary overlay ~3000 m thick Tertiary sand and gravel deposits. Groundwater is abundant in the shallow fractured basalt aquifer (specific capacity generally >0.2 m^2/s) and the downward leakage of water is the primary recharge source for the deeper porous confined aquifer system (Zhang and Li, 2005).

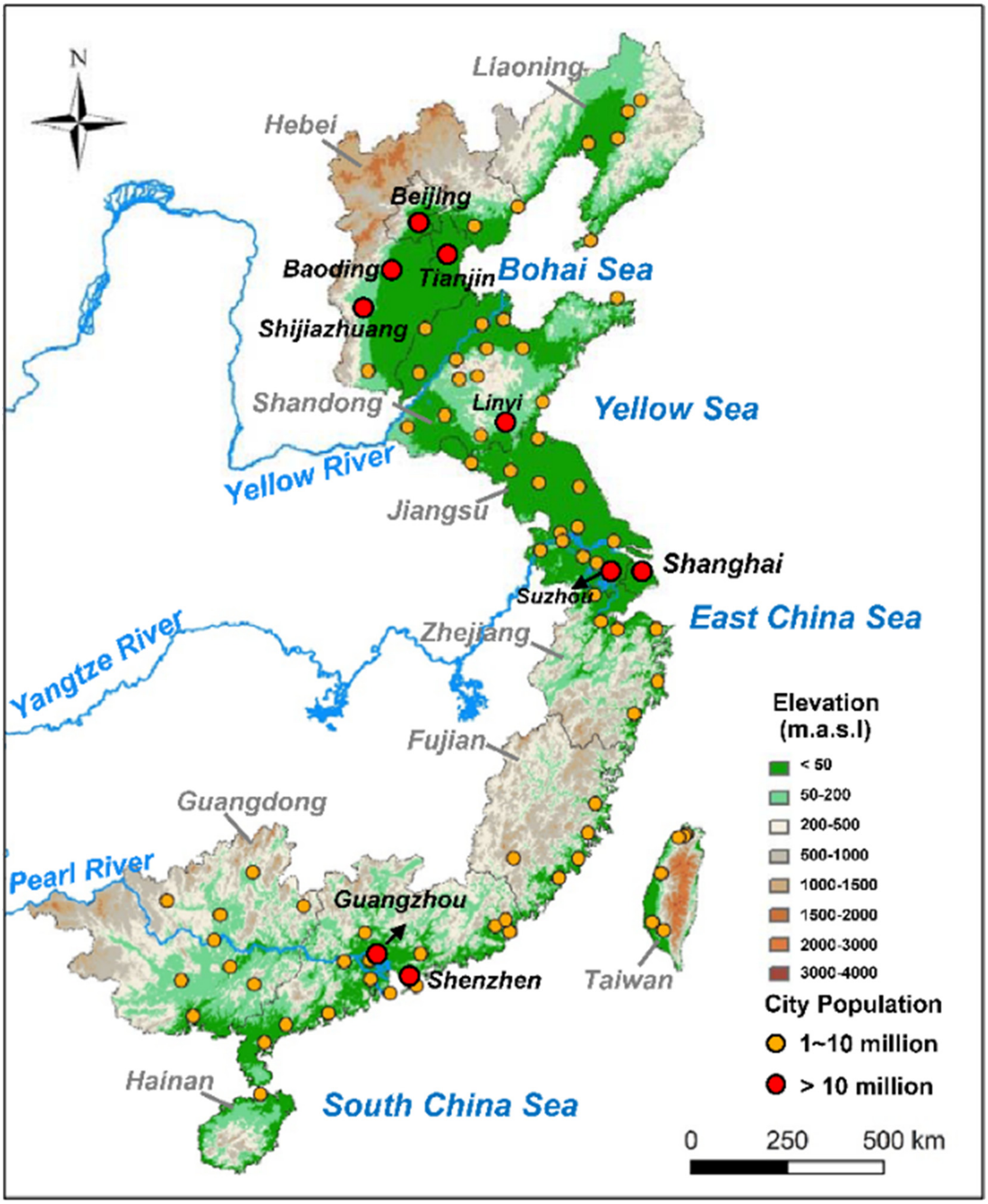

Fig. 2. Coastal topographic map of China and distribution of major cities. Data sources: Urban Construction Statistical Yearbook, 2019 and Jarvis et al. (2008).

3. Materials and methods

Groundwater quality data were collected from published literature, covering the last decade or slightly longer, for the coastal areas of China. The major groundwater quality indicators examined in the review included groundwater salinity (as TDS), nitrate, and reported occurrences of high fluoride groundwater (HFG) and high arsenic groundwater (HAG). Organic pollutants were excluded from the analysis as the data for groundwater (as opposed to surface water – see Han and Currell, 2017) are generally limited, sparse and difficult to statistically process and extrapolate in a meaningful way. Salinity, nitrate, fluoride and arsenic are considered key indicator pollutants which are indicative of the major sources and process resulting in quality degradation – i.e., fluoride and arsenic are geogenic elements, typically mobilized in response to other geochemical or water quality changes taking place (Currell et al., 2011; Rodríguez-Lado et al., 2013; Coomar and Mukherjee, 2021); nitrate is known to be overwhelmingly input to groundwater from anthropogenic sources – e.g. wastewater effluents and synthetic and natural fertilizers, while TDS is most sensitive to saltwater intrusion.

The data include numerous sites where groundwater salinization caused by seawater intrusion (SWI), resulting from groundwater overexploitation, has been identified. Fig. 3 is a location map showing identified SWI localities, with the ranges of intrusion distances included where these are reported (State Oceanic Administration, 2018). To investigate the distribution of coastal groundwater salinity, box-plots of groundwater TDS concentrations were generated from 2224 groundwater samples, from 13 investigated coastal areas, collected from literature mostly published within the last 10 years (see Section 4.1). Considering the complexity of land-sea interactions, submarine groundwater discharge (SGD) plays an important role in the coastal ecological environment. We performed a detailed analysis of SGD for 19 bay and estuary regions, using data from 46 sources (see Section 5.1).

Raw NO_3^- concentrations were compiled from the literature, totaling 690 surface water and 1217 groundwater samples, in 14 different areas/

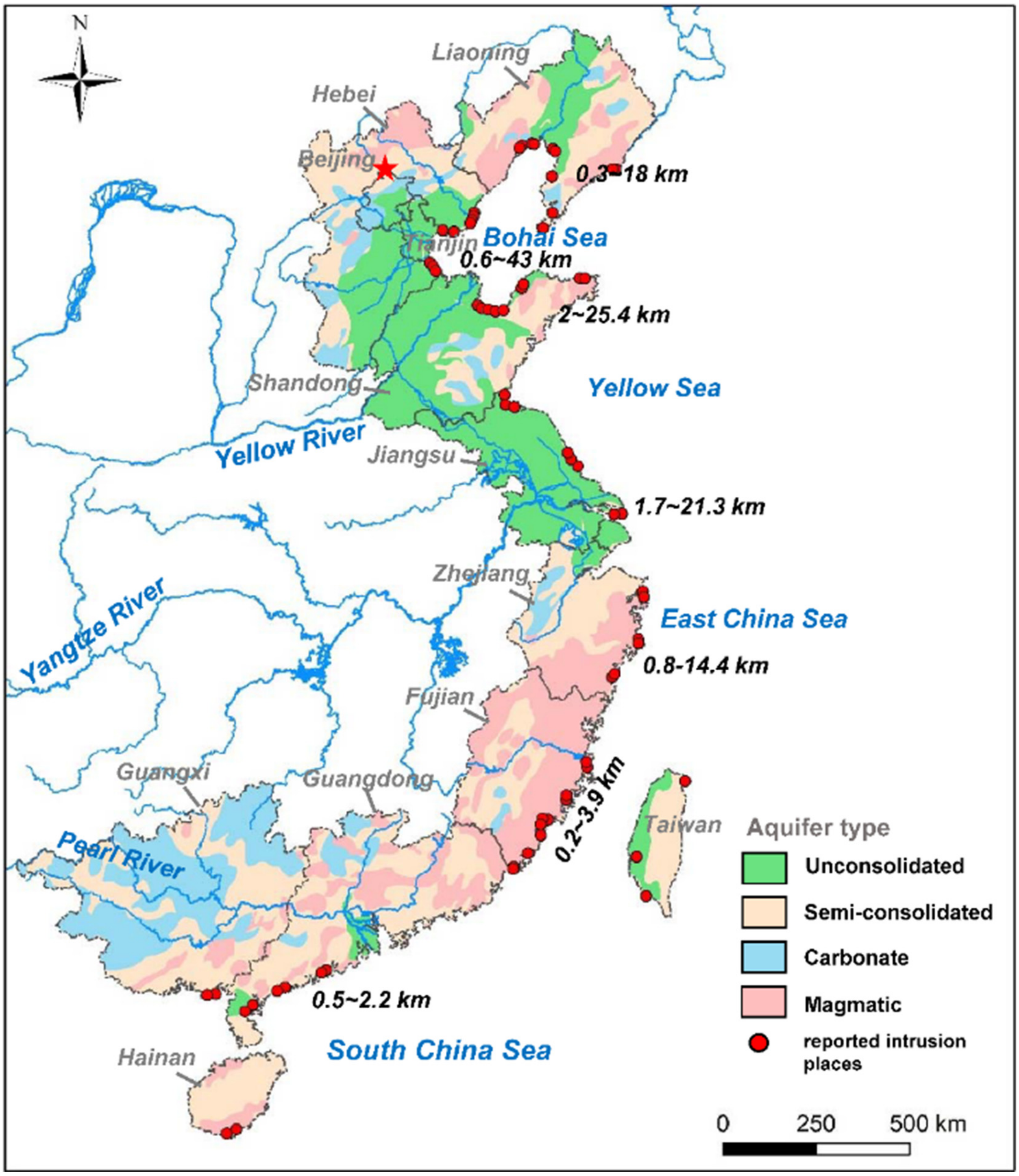

Fig. 3. Map of coastal aquifer types and seawater intrusion localities reported in China. Data sources: Institute of Hydrogeology and Environmental Geology, Chinese Academy of Geological Sciences (CAGS), Hydrogeological map of China, 1979 (available at: http://www.geoscience.cn/swdz/swdzt/index.htm), and State Oceanic Administration, 2018.

coastal provinces. These were converted to NO_3-N. The NO_3-N data were aggregated in statistical box-plots with calculated median, 25th and 75th percentiles and inter-quartile ranges indicated (see Section 4.2). Data were compared to the United States Environmental Protection Agency (USEPA) maximum contaminant level (MCL 10 mg/L) and an approximate background concentration of 1 mg/L, according to Burow et al. (2010). These boxplots provide a graphical summary of the degree of nitrate pollution of the major coastal groundwater systems. Nitrate isotope data were also compiled from 69 literature sources for surface water and groundwater across coastal regions (see Section 5.2). These data were compiled and plotted on a N vs. O plot showing typical nitrate source isotopic compositions.

Fluoride (F^-) concentrations from 2226 groundwater samples in 10 major coastal areas in China were collected from published literature, and similarly compiled into box-plots with calculated median, 25th and 75th percentiles and inter-quartile ranges (see Section 4.3). A distribution map of high arsenic groundwater (HAG) in the coastal areas of China is presented, showing groundwater with arsenic concentrations higher than 10 μg/L (guideline for drinking water recommended by the world health organization (WHO)) and 50 μg/L (severely exceeding the guideline), respectively, building on previous arsenic in groundwater mapping and vulnerability analysis presented by Rodríguez-Lado et al. (2013) (see Section 4.4). Box-plots of As concentrations in groundwater were also developed based on 4892 groundwater samples from 21 investigated areas, collected from literature mostly published within the last 10 years (see Section 4.4).

4. Results: distribution of groundwater quality indicators and associated issues in coastal China

4.1. Salinity and saltwater intrusion

Groundwater salinity is a particularly important aspect of groundwater quality in coastal areas. There are several potential causes of coastal groundwater salinization, including invasion of modern or ancient seawater induced by groundwater exploitation (e.g. Han et al., 2011; Han and Currell, 2018), evapotranspiration and dissolution and infiltration of chemical fertilizers in areas of farmland irrigation (Currell et al., 2012; Han et al., 2015a), influx of highly mineralized geothermal water into aquifers (e.g. Cao et al., 2020), and water-rock interaction (mineral dissolution and ion exchange), particularly in carbonate aquifers (Han et al., 2016b). Among these, seawater intrusion (SWI) is the most common identified issue in the compiled case studies. Groundwater salinization caused by seawater intrusion is mainly identified where Cl concentrations in groundwater exceed 250 mg/L or TDS concentrations in groundwater exceed 1000 mg/L. Box-plots of groundwater TDS concentrations for the coastal areas of China (Fig. 4) show obvious groundwater salinization in coastal aquifers of Liaodong Bay, the North China Plain, northern Shandong, Jiangsu, and the Pearl River Delta.

SWI in aquifers around the Bohai Sea in northern China is the most serious salinization issue identified in China, with the intrusion area estimated to have increased from 1552 km^2 at the end of 1980s to 2674 km^2 in 2002 (Sun et al., 2007). Due to higher rates of groundwater over-exploitation in northern China (see Section 2), SWI is mainly distributed in the coastal areas north of the Yangtze River mouth (Fig. 3). The most severely impacted areas are mainly distributed in the coastal areas of Liaoning and Shandong provinces – the largest expansion of SWI has occurred at Laizhou Bay (Han et al., 2011). Fig. 3 shows variation in the estimated SWI distances along the continental coastal region in 2017. The range of intrusion distances is 0.3-18 km for the Liaodong Peninsula, 0.6-43 km for the region around Bohai Bay, 2-25.4 km for the Shandong Peninsula, 1.7-21.3 km for Jiangsu, 0.8-14.4 km for Zhejiang, 0.2-3.9 km for Fujian, and 0.5-2.2 km for Guangdong. The 'SWI' (as represented by very high Cl or TDS concentrations) occurring locally up to 43 km from the present coastline, may be related to both present-day seawater encroachment and the effect of paleo-seawater, that has remained in the sediments but progressively freshened after extensive marine transgressions since the late Pleistocene (Han et al., 2020).

4.2. Nitrate

Nitrate concentrations ($n = 690$ for surface water and 1217 for groundwater) from 36 coastal areas of China (survey areas and references shown in Supplementary Fig. S5 and Table S2) are summarized in Fig. 5. Except for the Liaohe River Basin and Taihu Lake Basin in southern Jiangsu Province, the extent of nitrogen pollution in groundwater is generally more severe than surface water (as shown in Fig. 5). Nitrate pollution in groundwater is particularly serious in the northern coastal areas. The mean nitrate concentration in groundwater in Dalian, the Yangdai River Plain of Qinhuangdao, and eastern Shandong are 49.2 mg/L, 24.7 mg/L and 35.2 mg/L, respectively, far higher than the USEPA or WHO drinking water standards (10 mg/L). For southern China, the average nitrate concentration in groundwater is 15.3 mg/L in the Pearl River Delta, while in other areas groundwater pollution with nitrate is less severe.

4.3. Fluoride

Groundwater fluoride concentrations ($n = 2226$) from 24 surveys in 10 coastal regions (see Fig. S6 and Table S3 for investigation areas and

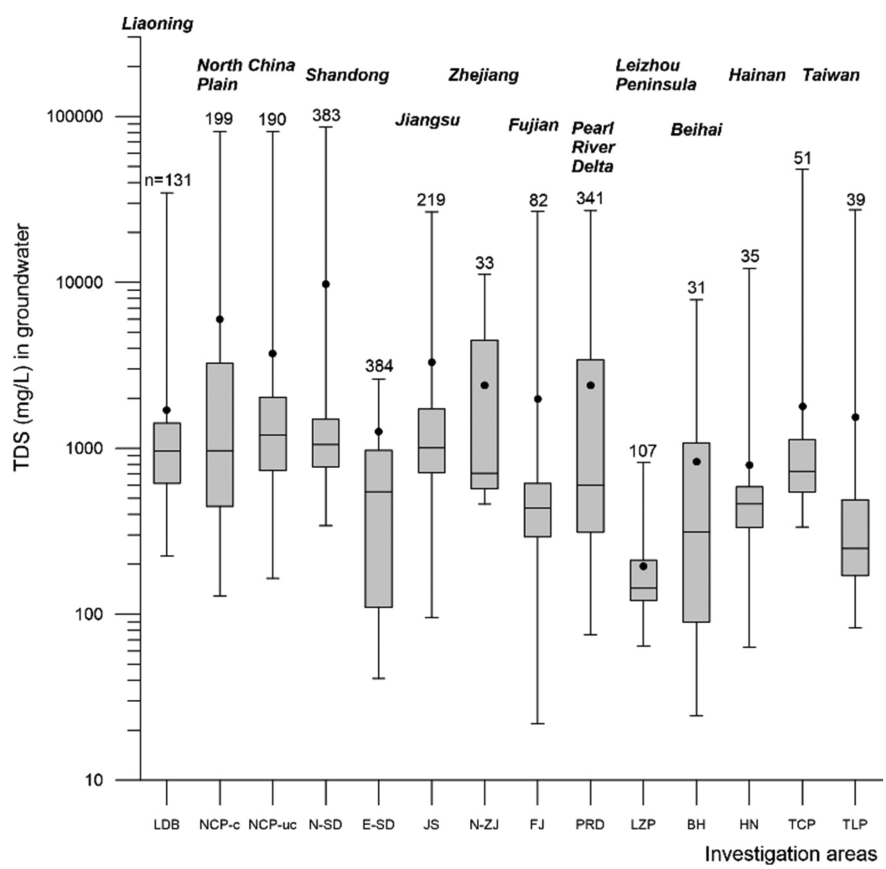

Fig. 4. Boxplot distributions of TDS concentrations in groundwater in coastal areas of China. LDB for LiaoDong Bay; NCP-c and NCP-uc for confined and unconfined groundwater of the North China Plain, respectively; N-SD for North of Shandong; *E*-SD for Eastern Shandong; JS for coastal region of Jiangsu Prov.; N-ZJ for Hangzhou-Jiaxing-Yuhang Plain, Zhejiang Prov.; FJ for the Changle District and Pingtan Island, Fujian; PRD for the Pearl River Delta; LZP for the Leizhou Peninsula; BH for Beihai, Guangxi Prov.; HN for Southeast Hainan; TCP for the Chia-Nan plain, southwestern Taiwan; TLP for the Lanyang Plain, Taiwan. Boxplots show median, inter-quartile range and 25th and 75th percentile values, with mean values in black dots. TDS > 1000 mg/L is generally used as the criterion for groundwater salinization. For full key to the location map and all data sources, refer to Supplementary Fig. S4, Supplementary Table S1 and accompanying references.

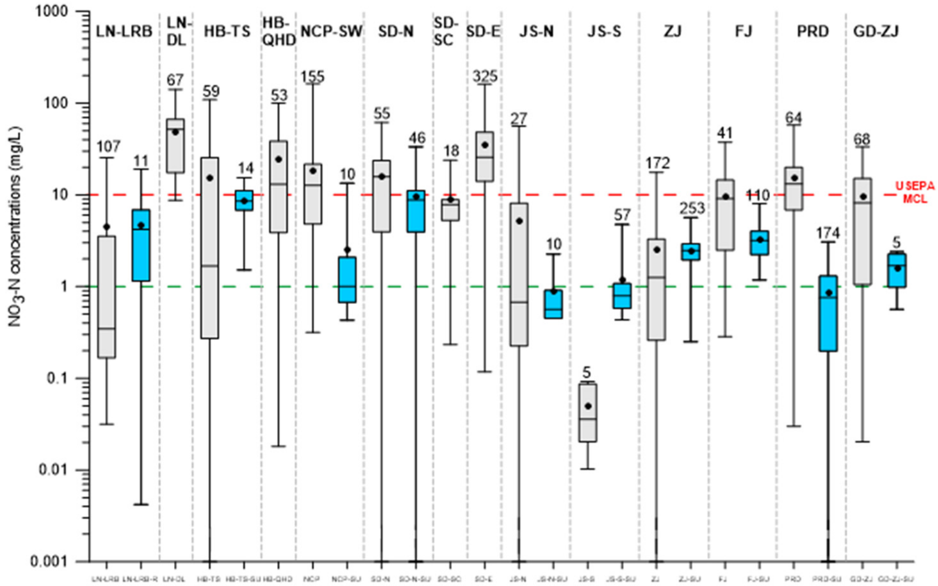

Fig. 5. Boxplot distributions of NO_3-N concentrations in groundwater (grey box) and surface water (blue box) in coastal areas of China. LN-LRB: the Liao River Basin of Liaoning Province; LN-DL: Dalian area of Liaoning Province; HB-TS: Tangshan in Hebei Province; HB-QHD: Qinhuangdao in Hebei Province; NCP-SW: southwest North China Plain; SD-N: northern Shandong Province; SD-SC: southcentral Shandong Province; SD-E: eastern Shandong Province; JS-N: northern Jiangsu Provinces; JS-S: southern Jiangsu Province; ZJ: Zhejiang Province; FJ: Fujian Province; PRD: Pearl River Delta; GD-ZJ: Zhanjiang of Guangdong Province. Boxplots show median, inter-quartile range and 25th and 75th percentile values, with mean values in black dots. Data is compared to the United States Environmental Protection Agency maximum contaminant level (USAEPA MCL 10 mg/L) and a background concentration of 1 mg/L (Burow et al., 2010). For full key to the location map and all data sources, refer to Supplementary Fig. S5 and Table S2 and accompanying references. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

references) are summarized in Fig. 6. High fluoride groundwater (HFG) in coastal areas of China is mainly distributed in the North China Plain, including Cangzhou and Tianjin, where the average concentration of fluoride (F) reaches 2.3 mg/L, higher than the upper drinking water safety limit set by the World Health Organization (WHO) (1.5 mg/L). Severe HFG also occurs in the coastal areas of the Shandong Peninsula, where a wide range of concentrations (0.1–15.7 mg/L) and the highest mean concentration (2.75 mg/L) was reported (Fig. 6). South-central Shandong Province, southern Jiangsu Province, and the Pearl River Delta exhibit mean values lower than 0.3 mg/L and have few samples exceeding 1.5 mg/L. In the most serious HFG regions (Cangzhou and northern Shandong), fluoride concentrations are generally higher in deep groundwater compared to shallow groundwater (Fig. S7).

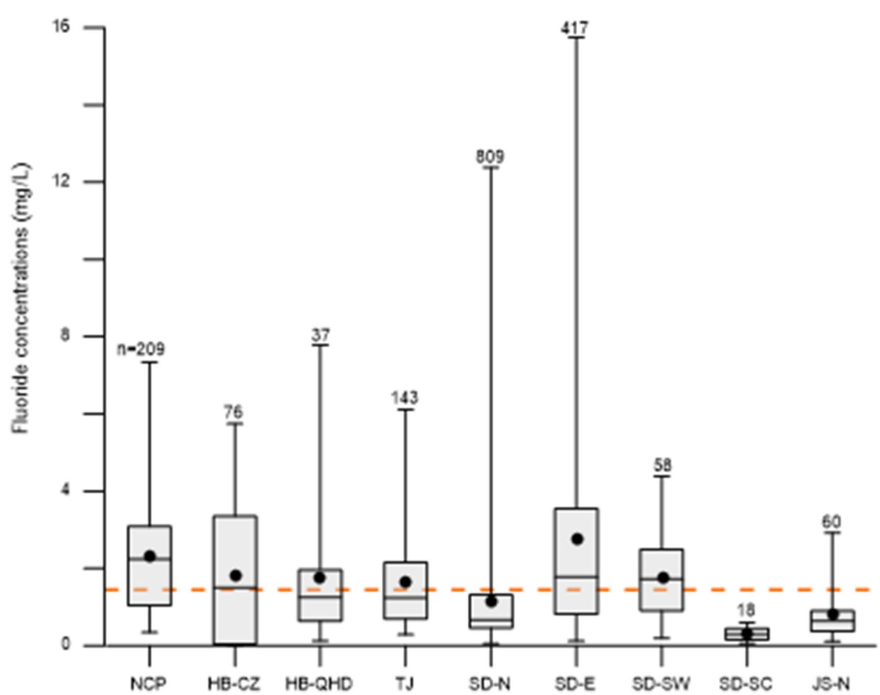

Fig. 6. Boxplot distributions of fluoride concentrations in groundwater from coastal areas in China. NCP: the North China Plain; HB-CZ: Cangzhou in Hebei Province; HB-QHD: Qinhuangdao in Hebei Province; TJ- Tianjin City; SD-N: north Shandong Province; SD-E: east of Shandong Province; SD-SW: southwest of Shandong Province; SD-SC: southcentral Shandong Province; JS-N: north of Jiangsu Provinces. Boxplots show median, inter-quartile range and 25th and 75th percentile values, with mean values in black dots. The red line is the WHO maximum contaminant level (1.5 mg/L) for drinking water. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

4.4. Arsenic

As shown in Fig. 7, high arsenic groundwater (HAG) is also prevalent in coastal areas, with concentrations exceeding the WHO guideline of 10 μg/L distributed in Hebei Province (Langfang and northern Chengde), the Shandong Peninsula, north and southwest Shandong Province, northern Jiangsu and some areas of Guangdong. Groundwater with arsenic concentrations higher than 50 μg/L is mainly distributed in these same regions, north of the Yangtze, as well as southwest and northeast Taiwan, with localized zones in the Pearl River Delta and Leizhou Peninsula, along the southern coastline.

Groundwater arsenic concentrations ($n = 4892$) from 48 surveys in 23 regions of China (see Fig. S8 and Table S4 for investigation areas and references) are summarized in Fig. 8. HAG in coastal areas of China is mainly distributed in the confined aquifer of the Shanghai-Nantong area (Yangtze River Delta), the Chia-Nan Plain of southwestern Taiwan, and the Lanyang Plain of Taiwan, where the average concentrations of arsenic (As) reach 115 μg/L, 166 μg/L, and 91 μg/L, respectively, far greater than the upper drinking water safety limit set by the WHO (10 μg/L). For the most part, arsenic concentrations in groundwater in the investigated areas have very wide ranges. Comparison of As concentrations across inland and coastal basins of China is illustrated by Fig. 8; severe HAG occurs in the Kuitun-Shihezi area of Xinjiang (mean 60 μg/L), the confined aquifer of the Guide Basin (mean 111 μg/L), the Yinchuan Plain (mean 65 μg/L), Hohhot Basin (mean 82 μg/L), Hetao Plain (mean 53 μg/L), Datong Basin (mean 180 μg/L), Songnen Plain (mean 94 μg/L), and the middle reach of Yangtze River (mean 75 μg/L). In some serious HAG regions (e.g. the Guide Basin, Shanghai-Nantong area), As concentrations are generally higher in deep confined groundwater compared to shallow unconfined groundwater (Fig. 8).

5. Discussion: major drivers of China's coastal groundwater problems

5.1. Seawater intrusion and submarine groundwater discharge in coastal China

The interaction between groundwater and seawater in coastal aquifers is an important part of the hydrological cycle, comprising two complementary processes: seawater/saltwater intrusion (SWI), and submarine groundwater discharge (SGD) (Taniguchi et al., 2002; Kaleris, 2006). Understanding these two processes and their dynamics is very significant for coastal groundwater resources management. SWI and SGD processes can not only affect the fluxes of water, salt, and nutrients crossing the land-sea interface, but also contribute to the coastal ecosystem. Groundwater quality in coastal aquifers depends on SWI, while SGD causes groundwater discharge into the ocean, carrying and transporting possible pollutants and affecting the water quality near the coast (Moore, 1999; Burnett et al., 2003; Sawyer et al., 2016; Zhou et al., 2019).

5.1.1. Seawater/saltwater intrusion (SWI)

Groundwater salinization caused by SWI, including both the lateral intrusion of modern seawater, the vertical invasion of saline tidal water, and intrusion (vertically and/or horizontally) of ancient seawater or evaporated brines into freshwater aquifers, has become a major global resource and environmental problem (Post, 2005; Han et al., 2014; Sawyer et al., 2016). As the climate warms and sea levels rise, SWI into coastal aquifers is expected to become more severe (Oude Essink et al., 2010; Cary, 2015), with modeling indicating the increasing demand for groundwater under a changing climate (and ongoing

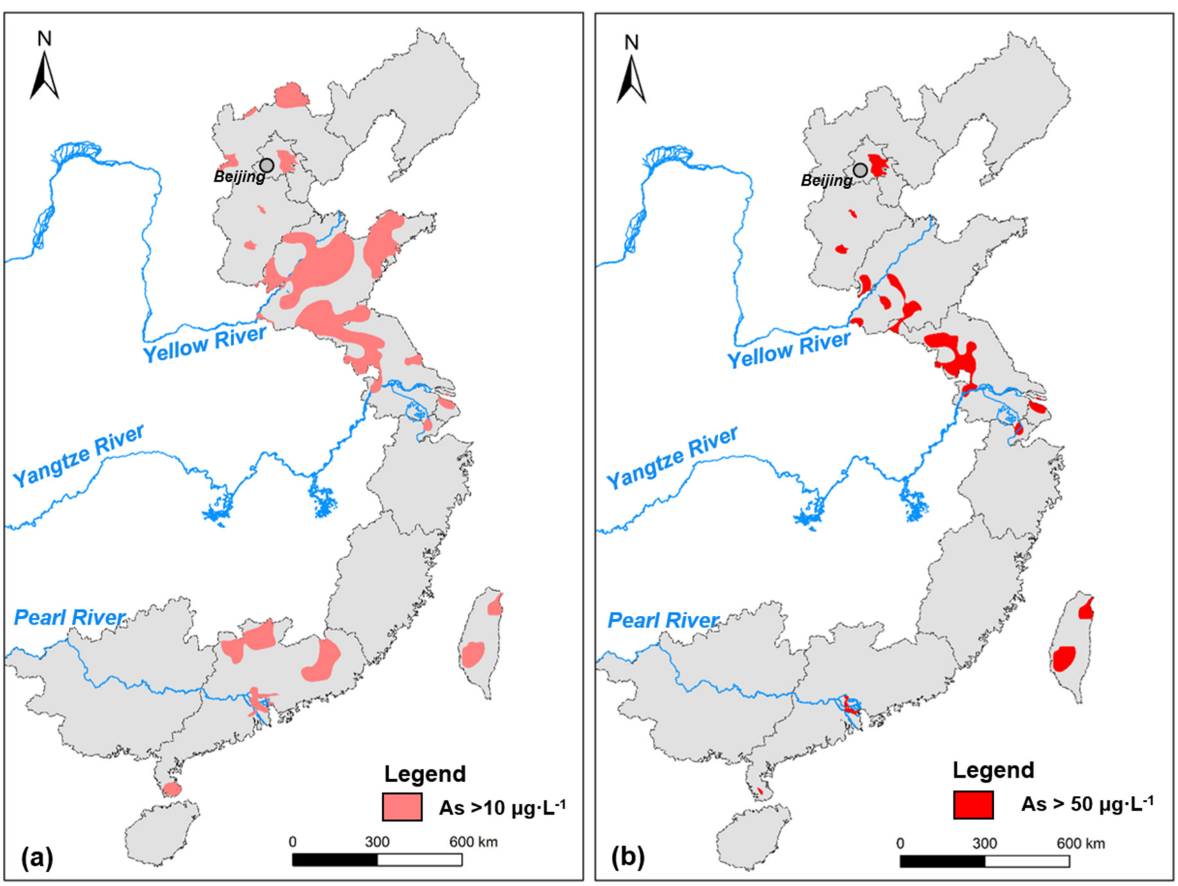

Fig. 7. Distribution maps of HAG in coastal China. Map (a) for showing areas with groundwater As concentrations higher than 10 μg/L; map (b) for areas with As concentrations higher than 50 μg/L. Data source from Wang et al. (2012), Guo et al. (2013), and Zhang et al., 2018b.

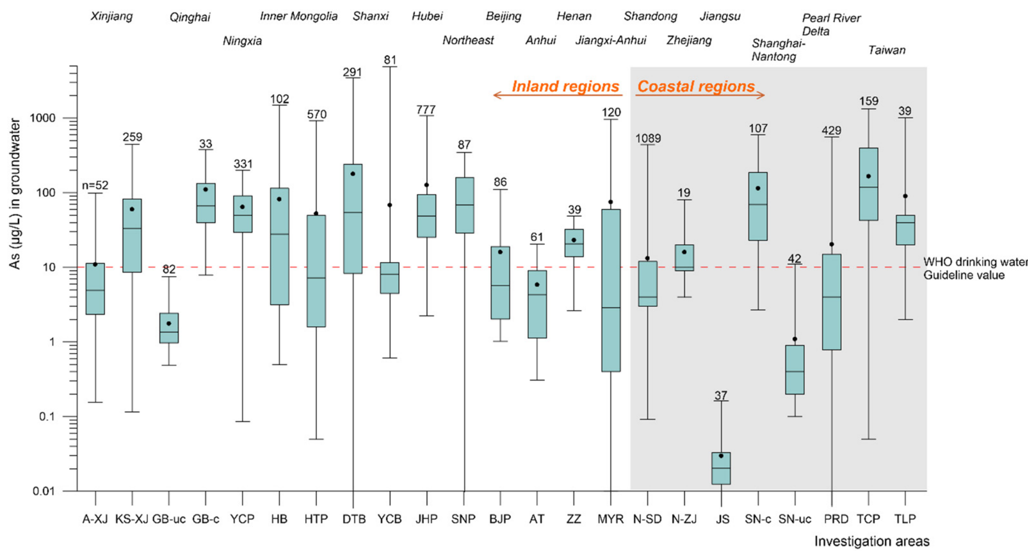

Fig. 8. Boxplot distributions of As concentrations in groundwater in investigated areas of China. A-XJ for Aksu area, Xinjiang; KS-XJ for Kuitun-Shihezi area, Xinjiang; GB-c and GB-uc for confined and unconfined groundwater of the Guide Basin, respectively; YCP for Yinchuan Plain; HB for Hohhot Basin; HTP for Hetao Plain; DTB for Datong Basin; YCB for the Yuncheng Basin; JHP for the Jianghan Plain; SNP for Songnen Plain; BJP for north of the Beijing Plain; AT for Antai, Anhui Prov.; ZZ for north of Zhengzhou City, Henan Prov.; MYR for the Middle Reach of Yangtze River; N-SD for North of Shandong Prov.; N-ZJ for North of Zhejiang Prov.; JS for Lianyungang-Yancheng, Jiangsu Prov.; SN-c and SN-uc for confined and unconfined groundwater of the Shanghai-Nantong region, respectively; PRD for the Pearl River Delta; TCP for the Chia-Nan plain, Southwestern Taiwan; TLP for the Lanyang Plain, Taiwan. Boxplots show median, inter-quartile range and 25th and 75th percentile values, with mean values in black dots. Data is compared to the WHO guideline value for drinking water (10 μg/L). For full key to the location map and all data sources, refer to Supplementary Fig. S8, Table S4 and accompanying references.

coastal development) likely to play a greater role than global sea level rise itself (Ferguson and Gleeson, 2012).

Groundwater over-exploitation to meet increasing water demands has led the regional groundwater levels to decline substantially in many coastal areas of China (Wang et al., 2018a), and the consumption of a tremendous amount of groundwater has increased the SWI extent in China's coastal regions (Shi and Jiao, 2014). The SWI area in China's coastal areas exceeds 16,000 km^2 (State Oceanic Administration, 2012), which is mainly distributed along the coast of the Bohai Sea (Fig. 3). SWI has occurred along more than 90% of the coastline around Bohai Bay, with the maximum intrusion distance more than 43 km inland from coast, at Laizhou Bay (Yu et al., 2021). Additionally, SWI problems have been reported in the PRD (Liu et al., 2019), the coastal area in Beibu Gulf (Zhong et al., 2020), and Hainan Island (Xu et al., 2009).

SWI caused by groundwater over-exploitation, resulting in kilometer-scale intrusions, has been recognized as a serious threat to groundwater quality in many coastal aquifers globally (Vengosh et al., 1999, 2002; Jørgensen et al., 2008; Ranjan et al., 2009; Petelet-Giraud et al., 2016; Mao et al., 2020). Groundwaters impacted by salinization are documented to exceed 500 mg/L of chloride (Han and Currell, 2018) and > 110 g/L of TDS (Han et al., 2014) in Northern China, while TDS of shallow waters affected by SWI is generally lower (< 6 g/L) in South China (Zhang et al., 2015). As illustrated by Fig. 4, TDS concentrations in coastal groundwater have a wide range, from 128.3 mg/L to 81.3 g/L with a mean of 4887.9 mg/L and 79.3 mg/L to 140.0 g/L (mean value of 9748.2 mg/L) for the coastal North China Plain and northern Shandong, respectively. A combination of modern SWI and palaeo-SWI is thought to be responsible for the high groundwater salinity depending on the region (e.g. Han et al., 2014).

In the past 5 decades, coastal groundwater salinization in the sandy coastal aquifer of the Yang-Dai River Plain has become increasingly serious due to the anthropogenic activities and climatic change. Intensive groundwater pumping is the primary process driving SWI, either by vertical infiltration along riverbeds, which convey saline surface water inland, and/or direct subsurface lateral inflow (Han and Currell, 2018). Due to other local sources of pollution, such as fertilizers and/or domestic wastewater, more severe and widespread nitrate contamination has also occurred in this region and other analogous settings (e.g. surrounding Bohai Bay), further degrading water quality in these aquifers (Han et al., 2016a). Geothermal water has also been documented to contribute to groundwater salinization in the Yang-Dai River Plain (Cao et al., 2020). Large-scale extraction of groundwater has ceased in the coastal carbonate aquifer in the Daweijia area of Dalian, northeast China since 2001, after salinization of the main well field. Historical monitoring there has demonstrated the reversibility of SWI in the carbonate aquifer setting, as evidenced by a decrease of the Cl^- concentrations in groundwater following restrictions on groundwater abstraction (Han et al., 2015a). This can be attributed to rapid flushing in this system, where flow occurs preferentially along karst conduits, fractures, and fault zones. Present-day elevated salinity in shallow groundwater – which is highly associated with pollutants like nitrate and sulfate, is more likely due to intensification of agricultural activities than SWI (Han et al., 2015b).

Hydrochemical-isotopic investigations in the Laizhou Bay Quaternary aquifer provided new insights into palaeo-SWI and brine migration as a cause of coastal salinization (Han et al., 2011; Han et al., 2014). In this setting, brines evolved during different phases of palaeo-regression and transgression during the Holocene. The brine water, with TDS concentrations up to 184 g/L (Xue et al., 2000) have corrected ^{14}C mean residence times of approximately 2.3–7.0 ka BP, accounting for mixing effects (Han et al., 2011, 2014). Brines are thought to have formed during periods of endorheic evaporation within coastal lagoon/palaeo-lake environments, which have been preserved in the sedimentary profile. Subsequent migration of these brines and mixing with fresher groundwater (particularly through vertical mixing) is evident in trace element (e.g. Cl/Br ratios) and stable isotopic compositions of saline and brackish waters of this region (Han et al., 2014).

At present, countermeasures against SWI in China's coastal areas mainly include management initiatives prohibiting or limiting groundwater abstraction, and engineering measures such as constructing tidal dams, underground dams, and artificial recharge schemes (Shi and Jiao, 2014; Zhu et al., 2019). However, there is still a lack of quantitative and systematic evaluation of the effects of these measures on the mitigation of SWI and related groundwater quality change (Wu et al., 2018). Such analysis is imperative to inform the management of coastal groundwater resources and make science-informed decisions on the most effective strategies, noting that engineered solutions require significant capital expenditure and ongoing maintenance.

5.1.2. Submarine groundwater discharge (SGD)

Between 90 and 99% of the total water discharged from the continents to the ocean is discharged via rivers (Church, 1996), with the remaining 1-10% discharged directly from aquifers to coastal wetlands, beaches, and continental shelves (Burnett et al., 2003). While SGD flows are small compared to rivers, they provide the oceans with a large amount of nutrients and solutes that sustain (and can potentially contaminate) coastal ecosystems (Taniguchi et al., 2002; Slomp and Van Cappellen, 2004; Moore, 2010; Amato et al., 2016). On a global scale, SGD-driven dissolved organic nitrogen (DIN) and dissolved inorganic phosphorus (DIP) fluxes are thought to be 1.4 and 1.6 times global ocean and river inputs, respectively (Cho et al., 2018). SGD nutrient fluxes can thus dramatically affect nutrient budgets and biogeochemical cycles, and drive water quality deterioration through coastal eutrophication (Moore, 2006; Wang et al., 2018c; Adolf et al., 2019). SGD can also transport heavy metals and organic pollutants (Gonneea et al., 2014; Johannesson et al., 2017; Wang et al., 2018b; Zhang et al., 2020b). SGD comprises submarine fresh groundwater discharge (SFGD) and re-circulated saline groundwater discharge (RSGD). RSGD includes re-circulating saline water driven by waves, tides, storm surges, and density currents (Li et al., 1999; Taniguchi et al., 2002; Burnett et al., 2003).

The magnitude of SGD in China's coastal regions is estimated at 5.4-10.2 $\times$ 10^{12} m^3/a, accounting for 5 - 9% of global SGD flux. SGD accounts for 50% of total inputs of DIN, DIP, and dissolved inorganic silicate (DSi) in China's coastal waters (Zhang et al., 2020a). Based on 53 regional SGD studies, variations of SGD flux and SGD-driven nutrients are compiled in Fig. 9. In recent years, radium (Ra) and radon (Rn) isotope mass balance models have been created for SGD flux estimation for Bohai Bay, Laizhou Bay, Daya Bay, and Jiaozhou Bay (e.g., Wang et al., 2015; Wang et al., 2018c; Zhang et al., 2020a). These suggest SGD in Bohai Bay is 3.11-7.43 $\times$ 10^8 m^3/d in the dry season and 4.47-11.98 $\times$ 10^8 m^3/d in the wet season. This is an order of magnitude larger than the average annual runoff from the Yellow River to the sea, ranging from 7.23 $\times$ 10^7 m^3/d for 2012 (Wang et al., 2015) to 2.45 $\times$ 10^7 m^3/d in 2017 (Wang, 2020). The SFGD along Laizhou Bay is estimated to range from 4.12 $\times$ 10^7 to 6.36 $\times$ 10^7 m^3/d, and the total groundwater discharge between 5.3 $\times$ 10^8 m^3/d to 6.2 $\times$ 10^8 m^3/d. For Daya Bay, SGD ranges from 2.8 $\times$ 10^6 to 1.0 $\times$ 10^7 m^3/d, while the Danao River discharge it is only estimated at 3.1 $\times$ 10^3 m^3/d (Wang et al., 2017b; Wang et al., 2018a). Upstream water harvesting schemes on northern China's rivers (which divert substantial flows to irrigated agriculture) have had a major effect on these river flow volumes.

The areas with highest rates (per area) of SGD flux are Laizhou Bay, the Dongshan Bay, the Jiulong River Estuary, and the Daya Bay, with the estimated SGD rate exceeding 100 cm/d. SGD fluxes of porous aquifers, composed of loose and semi-consolidated sediments, such as the Laizhou Bay and the Dongshan Bay, appear to be relatively high. The spatial and seasonal variations of SGD flux are significant - SGD rates in a given area may vary by a factor of ten or greater at different times. SGD usually has a lag effect in response to seasonal hydrological conditions, in contrast to river discharges (Luijendijk et al., 2020), which leads to SGD being several times larger than river flux during drought periods; conversely SGD flux is several times smaller than the river flux during the wet season.

In terms of nutrient flux and associated water quality, DIN and DSi fluxes are generally 1-2 orders of magnitude larger than DIP fluxes. Based on the available datasets, areas with high SGD flux tend to be accompanied by high nutrient flux. The highest DIN, DIP, and DSi fluxes occurred in the Jiulong River Estuary, the Dongshan Bay, and the Yangtze River Estuary, respectively. There is a potential correlation between the three nutrients, i.e., in coastal areas with high DIN fluxes, DIP and DSi also tend to be higher. Porous aquifers composed of loose sediments contribute higher nutrient fluxes than bedrock coastal zones (as well as higher SGD flux rates). The SGD-driven nutrient fluxes in the Pearl River Delta and the Yangtze River Delta are relatively high, which is related to the high density of population and (likely) the degree of urbanization in these regions. The SGD-driven nutrient flux in large coastal cities is generally higher than coastal agricultural areas (Fig. 9).

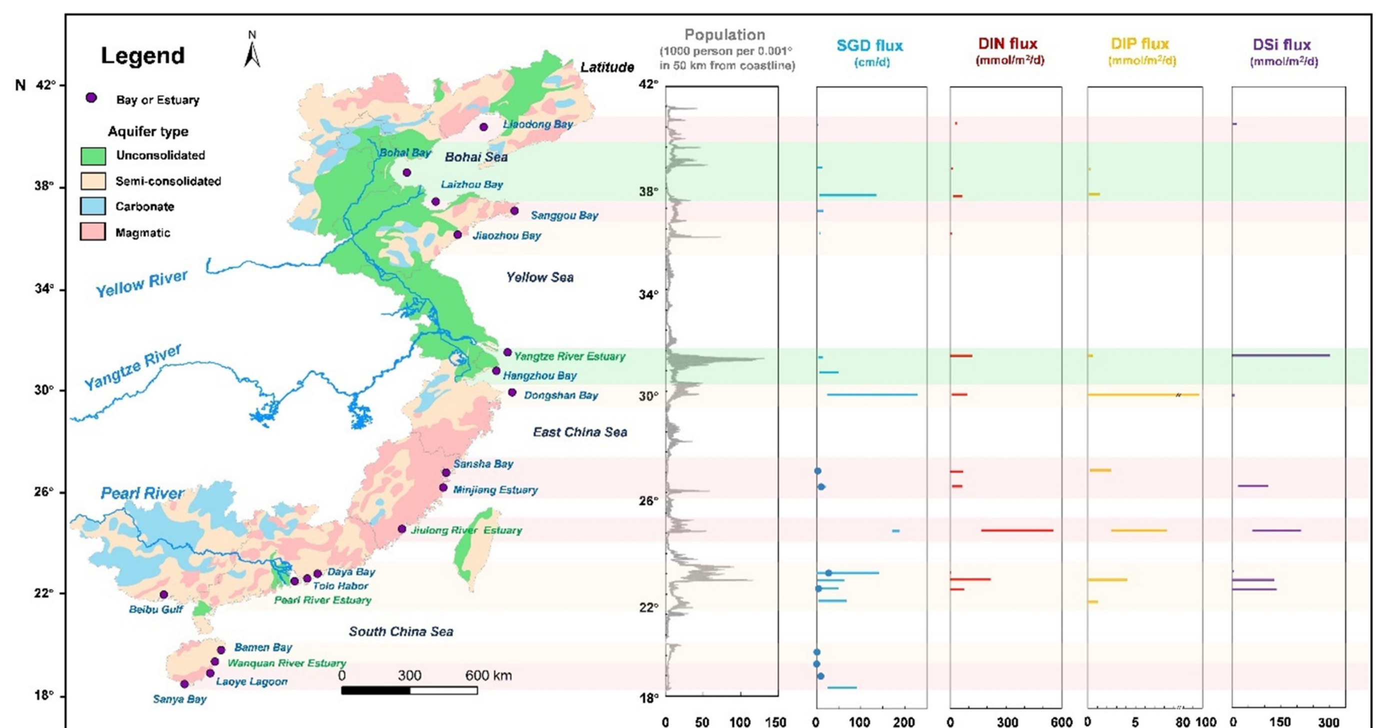

Fig. 9. Variations of population density, SGD flux, SGD-driven DIN, DIP, and DSi flux along the China coastline. Statistical data and data sources are shown in Table S5.

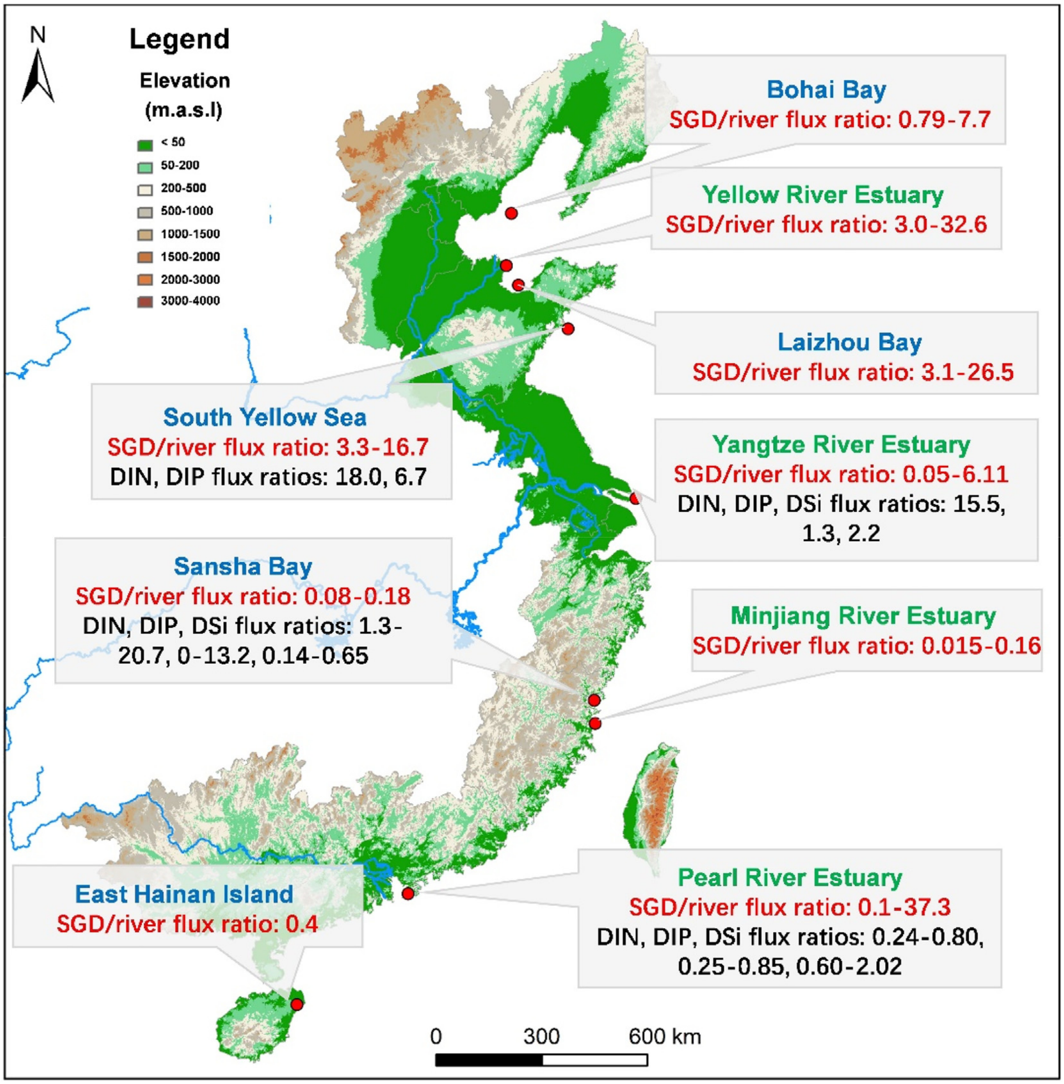

Fig. 10. Flux ratio of SGD and river discharge and corresponding nutrient inputs ratio along the Chinese coastline. DIN, DIP, and DSi flux ratios refer the ratio of SGD-driven and river-discharged DIN, DIP, and DSi, respectively. The numerical ranges of the ratio result from different estimation methods or seasonal change. Data sources: the South Yellow Sea (Liu et al., 2017a); the Sansha Bay (Wang et al., 2018b); the East Hainan Island (Su et al., 2011); the Bohai Bay (Tang et al., 2015; Yi et al., 2019; Wang et al., 2020); the Yellow River Estuary (Xu et al., 2013); the Laizhou Bay (Wang et al., 2015; Zhang et al., 2018b); the Yangtze River Estuary (Gu et al., 2012; Liu et al., 2018; Wang et al., 2018d); the Minjiang River Estuary (Liu et al., 2016); the Pearl River Estuary (Liu et al., 2012; Gao et al., 2018; Liu et al., 2018; Xiao et al., 2019). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

It is important to note uncertainties in the absolute and relative rates of SGD and river flux at the coastline (Fig. 10). It is difficult to determine the spatial range of SGD in estuary areas, and so far, there is no long-term SGD monitoring network. Sedimentary characteristics and geological structure make the spatial distribution of permeability highly heterogeneous, imparting a significant scale effect on the estimated rates, and uncertainty regarding their wider applicability across large areas of coastline.

5.2. Agricultural irrigation and agro-chemical (pesticide and fertilizer) usage

China is the world's largest consumer of nitrogen fertilizers, accounting for around one-third of total global consumption (Kahrl et al., 2010; FAO, 2014). Agricultural systems, including croplands and livestock, are responsible for 59% of current nitrogen discharge. Domestic waste (including urban sewage, rural sewage, and organic waste) contributes 39%, and industrial waste the remaining 2% (Yu et al., 2019). Thus, while the application of nitrogen fertilizers has played a significant role in maintaining food security in China, the resulting loss of excess nitrogen from fertilizers to the environment is huge. Up to 43% of applied N fertilizer is estimated to have been lost through the processes of ammonia volatilization, nitrate leaching (i.e., to groundwater), and nitrification / denitrification in the winter wheat-summer maize crop rotation system, which dominates agricultural land in northern China (Zhang et al., 2012; Ju and Zhang, 2017).

The highest nitrogen inputs are mainly distributed between latitudes 27 and 35 N, within which the coastal provinces and cities include Zhejiang, Shanghai, Jiangsu, Shandong, Hebei, and Tianjin. In these areas, nitrogen input reaches up to 250 $kg\cdot ha^{-1}\cdot yr^{-1}$, while in other coastal provinces, it is generally between 50 and 100 $kg\cdot ha^{-1}\cdot yr^{-1}$ (Gu et al., 2013). The areas with high nitrogen input are generally alluvial and diluvial plains or basins with flat topography. The long-term dependence of agriculture on chemical fertilizer is the primary reason for the high nitrogen input to groundwater in these areas. After the year 2000, nitrogen entering groundwater in the form of landfill leakage showed a rapid increase, while the nitrogen input from cultivated land tended to be stable (Gu et al., 2013).

Spatial distribution of irrigation is directly related to crop type and precipitation (Gao and Mu, 2003; Dong, 2009). In 2010, the total area of irrigated arable land in China was 607,977.1 km^2, approximately half the total (Liu et al., 2017b). China's agriculture can be divided by the line from the northernmost part of Hebei to the southernmost part of Guangxi. The eastern part of this zone is dominated by irrigated agriculture while the western part is mainly rain-fed (with some notable exceptions) (Fig. 11). Irrigated cultivated land in China is most concentrated in the Huang-Huai-Hai Plain and the middle and lower reaches of the Yangtze River; the provinces Henan, Hebei, and Shandong have the largest irrigated area, each exceeding 45,000 km^2. In the southern coastal areas, such as Guangdong, Fujian and Zhejiang, annual rainfall is more than 1000 mm, and irrigation is mostly utilized on scattered and limited paddies (Liu et al., 2017b). The distribution of irrigated agricultural land clearly corresponds with the high nitrate concentrations in groundwater in the northern coastal regions (see Fig. 5).

Stable nitrogen isotopic ratios have been used as an important means to identify the sources of nitrate pollution in water. Plotting 1030 samples from seven areas (Fig. 12) showed no significant evidence of denitrification (the primary removal process for nitrate from groundwater) for most regions. Typically, soil organic matter is limited in the North China Plain and soils are sand and silt-rich, likely reducing the opportunity for such nitrate removal. As can be seen from Fig. 12, most nitrate pollution in coastal areas appears to be derived from the pollution of NH_4-fertilizer, followed by mixed sources between NH_4-fertilizers and sewage/manure, as described by Martinelli et al. (2018). Utilization of organic manures as fertilizers and mixing or alternation with synthetic fertilizers may result in the overlap of many samples within these two isotopic composition fields. Nitrogen pollution is often accompanied by seasonal effects (Finlay et al., 2007; Ding et al., 2014), e.g., atmospheric deposition and soil —N depend on storm magnitude (Nestler et al., 2011) and these effects may not be captured within the aggregated data; time-series sampling for nitrogen isotopes is rare.

Apart from excess nutrients from fertilizers, pesticides, and other organic contaminants (including emerging and persistent organic pollutants), pathogenic microorganisms derived from agricultural activities may also contribute to the degradation of both ground and surface water quality (Symonds et al., 2018; Xin et al., 2019). Heavy pollution with POPs (including PAHs, OCPs, PCBs) has been found in coastal (surface) water bodies of China, including the Yangtze River Estuary, the Pearl River Estuary/Delta system, the Minjiang River Estuary, the Jiulong River Estuary, the Daya Bay, the Taihu Lake, and Zhejiang Province (Han and Currell, 2017). Agricultural pesticide use and industrial pollution discharge are both important sources of organic pollutants, leading to these becoming nearly ubiquitous in the water cycle. Compared to surface water data, groundwater data for such pollutants are very limited. It is thus imperative that organic pollution in groundwater be investigated for China's coastal regions, particularly in light of the significant persistent organic pollutant loads reported in surface water (Han and Currell, 2017), the known linkages between surface and groundwater, and the significant rates of nutrient and other solute discharges via groundwater, indicated by the review of SGD rates above (Section 5.1.2).

5.3. Fluoride and arsenic contamination of coastal groundwater

5.3.1. High fluoride groundwater (HFG)

HFG occurrence creates a chronic public health burden, and puts significant pressure on other, safer groundwater resources. More than 200 million people in 28 countries are affected by endemic dental and/or skeletal fluorosis, from drinking untreated groundwater with fluoride concentrations exceeding the WHO guideline value (1.5 mg/L) (Li et al., 2014; Rao, 2017; Narsimha and Sudarshan, 2017). The most severe impacts are felt in India and China, where over 66 million and nearly 45 million people are affected, respectively (Wang et al., 2002;

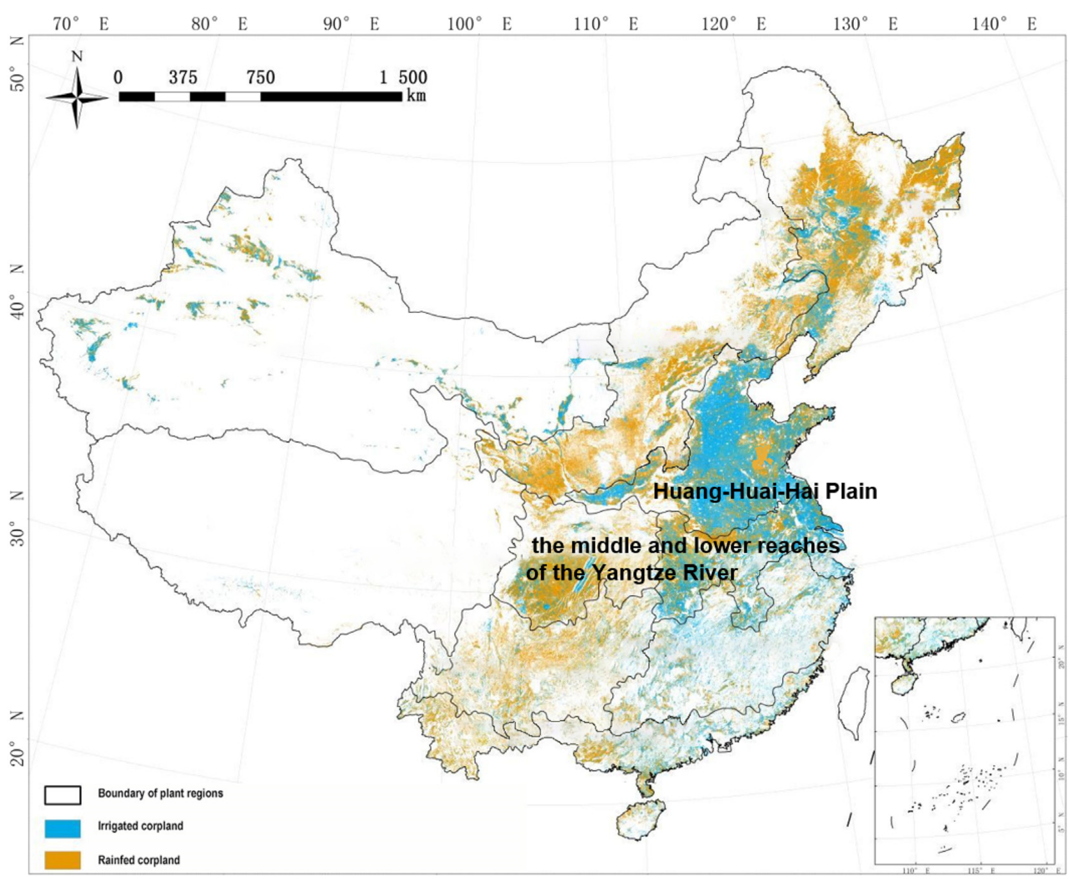

Fig. 11. Spatial distribution of irrigated and rainfed cropland of China in 2010, extracted from time-series NDVI (modified from Liu et al., 2017b).

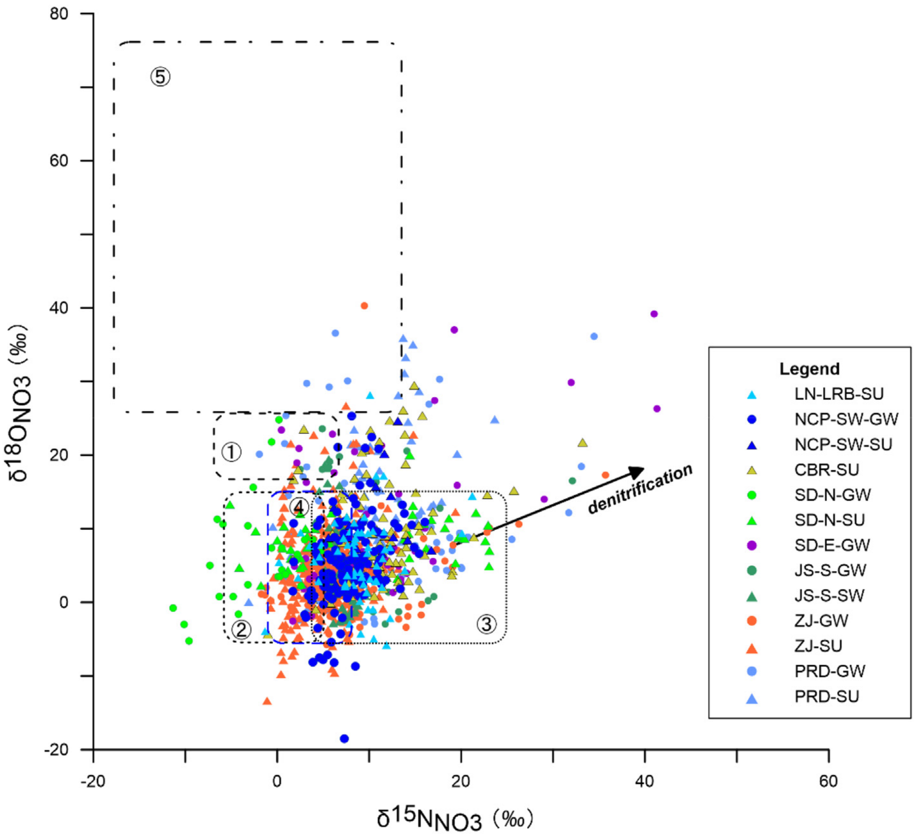

Fig. 12. Variations of $^{15}N_{NO3}$ versus $^{18}O_{NO3}$ values of NO_3^- in groundwater (and locally, surface water) for different coastal areas of China. LN-LRB: the Liao River Basin in Liaoning; NCP-SW: southwest North China Plain; CBR: the Circum-Bohai Sea-Region; SD-N: northern Shandong; SD-E: eastern Shandong; JS-S: southern Jiangsu; ZJ: Zhejiang; PRD: the Pearl River Delta. SU: surface water; GW: groundwater. The ranges of typical nitrate source isotopic compositions are referenced from Kendall and McDonnell (1998), Xue et al., 2009, Ji et al., 2017, Hu et al., 2019, and Huang et al., 2021: ① NO_3-fertilizer; ② NH_4-fertilizer; ③ manure and septic waste; ④ Soil N; ⑤ NO_3 in precipitation. Locations of the surveyed areas and related references can be seen in Fig. S9 and Table S6.

Mukherjee and Singh, 2020). The fluoride mainly originates from geogenic sources (Edmunds and Smedley, 2005; Ozsvath, 2009; Xiao et al., 2015; Mukherjee and Singh, 2020), with the primary origin being fluoride-rich minerals such as fluorite, biotite, fluorapatite, hornblende, amphibole, and mica, contained in fractured rock aquifers or as detrital phases within clastic sedimentary aquifers (Jacks et al., 2005; Kumar et al., 2015; Ali et al., 2016; Thapa et al., 2017; Yousefi et al., 2018).

Fluoride mobility in the aqueous environment is governed by water-rock interaction processes - mineral dissolution (or precipitation), hydrolysis and ion exchange/adsorption reactions, which can be controlled by aquifer and aquitard mineralogy (e.g. presence of F-bearing minerals and oxide phases with exchange sites), hydraulic properties, ambient geochemical environment (e.g. temperature, pH and alkalinity), groundwater residence times, and secondary processes such as evaporation (Apambire et al., 1997; Gosselin et al., 1999; Li et al., 2014; Meenakshi et al., 2004; Kim and Jeong, 2005; Dhiman and Keshari, 2006; Madhnure et al., 2007; Currell et al., 2011; Guo et al., 2012; Su et al., 2013; Kumar et al., 2015; Mukherjee and Singh, 2020; Adimalla and Qian, 2020). High fluoride in groundwater originating from geological sources can be removed by fluorite precipitation in Ca-rich solutions (Chae et al., 2007), or be absorbed from mineral surfaces under neutral to acidic conditions (Tang et al., 2009; Borgnino et al., 2013). Additionally, elevated fluoride concentrations may in some cases be attributed to anthropogenic sources, including over-application of phosphate fertilizers and pesticides (Saxena and Ahmed, 2003; Rao, 2003, 2017; Kim et al., 2011), irrigation return flow from use of deeper HFG (Pettenati et al., 2013; Li et al., 2015), landfill leachate, industrials effluents, coal combustion (Farooqi et al., 2007; Mukherjee and Singh, 2020; Wang et al., 2019a), and brick kilns (Datta et al., 1996; Ali et al., 2016).

In China's coastal aquifers (see Section 4.3 above), HFG is generally associated with fluvial and lacustrine sediments, as opposed to igneous or other hard rock aquifer types, and it is particularly prevalent where sandy aquifers are underlain, overlain and interspersed with clay-rich aquitards (e.g., Tianjin, Cangzhou). The relatively high F concentrations in deep groundwater in the compiled data (Section 4.3) indicate that a) HFG is not (primarily) related to infiltration of polluted surface waters or evaporation and b) that there is a correspondence between severe declines in groundwater levels due to over-exploitation (which occur most markedly in China's confined aquifers) and fluoride enrichment. For example, it is deep, confined aquifers such as the Q3 unit of the North China Plain, where groundwater levels have declined most markedly due to extraction for irrigation water, where high (and increasing) F concentrations occur.

In coastal China, HFG is mainly distributed along with Na-HCO_3 and Na—Cl type water (Li et al., 2020). In many settings worldwide it has been established that HFG is characterized by high sodium and low calcium concentrations, and highly alkaline groundwater is conducive to fluoride enrichment (Chae et al., 2007; Guo et al., 2007; Tirumalesh et al., 2007; Gomez et al., 2009; Wang et al., 2009; Currell et al., 2011; Su et al., 2013; Ali et al., 2016). Fig. S10 shows the pH frequency of groundwater samples collected from Liaoning and Shandong provinces and the North China Plain and indicates that HFG mainly exists in an alkaline environment. This is thought to relate to the propensity for desorption of the F anion from iron and manganese oxy-hydroxide phases at higher pH and/or competitive sorption with bicarbonate ion (Currell et al., 2011; Kim et al., 2012).

Examining the relationship between HFG and key potential governing factors in the Cangzhou area of the North China Plain – one of the most severely affected areas for HFG – provides insight into the multi-stage process by which fluoride enrichment in groundwater takes place across many of China's coastal aquifers (and probably, similar settings worldwide). There were multiple phases of volcanic activity in the Cangzhou region during the Cenozoic period. The deep volcanic lithology is mainly composed of pyroclastics, tuff, and basalt, which are abundant in fluorine-bearing minerals such as tourmaline, amphibole, apatite, biotite, and fluorite (Tian, 1984). The volcanic deposits then became interlayered with, and overlain by, the more recent fluvial and lacustrine sediments of the Quaternary period, derived through the weathering and deposition of pre-existing volcanic rocks and forming the primary aquifer and aquitard units of the Quaternary North China Plain.

Fluoride in deep groundwater in Cangzhou mainly arises from the release of F^- from detrital minerals and exchange/sorption sites - e.g., associated with Fe, Mn, and Al (hydr)oxides, which are particularly abundant in the aquitards deposited by fluvial-lacustrine processes. The most elevated groundwater concentrations are mainly distributed in the deep Q3 aquifer, with concentrations as high as 8 mg/L, and the highest F concentrations are generally at the top and bottom of this unit (i.e., adjacent to overlying and underlying aquitards). With the continuous increase of deep groundwater pumping from the 1970s onwards, the hydraulic gradient between deep confined aquifers and overlying units increased, as did the release of fluoride ion into groundwater (Liu, 1983; Tian, 1984; Su et al., 2018). The HFG in this area thus involves a combination of a geogenic primary source (weathering of volcanic rocks and incorporation of F^- into clay-rich layers in Quaternary sediments), and anthropogenic impact (groundwater pumping) driving the enrichment of fluorine in the deep groundwater in conjunction with aquitard compaction and land subsidence (Li et al., 2020). In addition, the thick clay-rich layers of the Cangzhou area (and similar parts of the NCP) are prone to ion-exchange between Ca^{2+} and Na^+, forming an alkaline water environment, conducive to the fluoride enrichment through exchange reactions (Currell et al., 2011).

Apart from water-rock interaction that releases fluoride into groundwater, evaporation (either in modern or geological times) can also play a significant role in HFG enrichment (Kim and Jeong, 2005; Guo et al., 2007; Wang et al., 2009). There is residual ancient seawater in the NCP, which experienced evaporation and complex water-rock interaction and it has been suggested this contributes to enrichment of trace elements including fluoride (Shen et al., 2021). The HFG in Shandong province (for example), is mostly distributed in SWI-affected areas. In this case, the formation of high sodium waters by cation exchange, during seawater intrusion, is likely a controlling factor in mobilizing F (Chen et al., 2020).

The high fluoride water in the west of Shandong is related to the large amount of fluorine-containing minerals in the alluvial sediment brought by the Yellow River, which provides source material for the formation of HFG; Na-HCO_3-type groundwater again plays an important role (Yin, 2015). While most silicate minerals containing F are low in solubility, the alluvial sediments derive from weathering of F-rich source rocks in northwest China and include oxide phases which adsorb F^- and subsequently release it into groundwater. The HFG in north Shandong Province is controlled by semi-arid climatic conditions, slow groundwater flow rates and limited recharge rates, resulting in long-term interaction between groundwater and fluoride-bearing strata (Yang, 2016). Generally, the fluoride concentrations in deep aquifers are higher than in shallow aquifers across all settings, underscoring the importance of the geogenic sources and potential release triggered by high rates of groundwater extraction.

5.3.2. High arsenic groundwater (HAG)

Geogenic occurrence of arsenic in groundwater can cause major adverse health effects for humans and wildlife (Smedley and Kinniburgh, 2002; Ravenscroft et al., 2009). It is estimated approximately 94 to 220 million people around the world (of which 85 to 90% are in South Asia) are potentially exposed to HAG from their domestic water supply (Podgorski and Berg, 2020). In China, over 19 million people are affected by drinking water with As concentrations more than 10 μg/L (Michael, 2013). In mainland of China, HAG is mainly distributed in arid inland basins and river deltas (Figs. 8 and S8), including the Kuitun-Shihezi area in Xinjiang (Luo et al., 2017) and the Datong Basin in Shanxi (Xie et al., 2012, 2015; Li et al., 2020), the Hetao Basin in Inner Mongolia (Smedley et al., 2003; Cao et al., 2018), the Yinchuan Basin in Ningxia (Han et al., 2013; Wang et al., 2017a), the Pearl River Delta(Wang et al., 2012; Zhang et al., 2018a),the Yangtze River Delta (Zeng, 1996; Chen, 1998), and the Jianghan Plain in Hubei (Duan et al., 2015; Yang et al., 2020). Most HAG in China, including coastal aquifers originates from geogenic sources, in groundwater that is (like HFG) in an alkaline state (Welch and Lico, 1998; Smedley and Kinniburgh, 2002; Currell et al., 2011; Guo et al., 2014).

High arsenic concentrations (>50 μg/L), which far exceed the WHO drinking water guideline, have been documented in the uppermost confined aquifer in the Nantong-Shanghai section of the lower Yangtze River Delta since the 1970s (Chen, 1998). Groundwater in this area occurs in a strong reducing environment, where AsO_4^{3-} is readily reduced to AsO_3^{3-}. As well documented in many coastal deltaic sediments, under a reducing environment, iron and manganese oxides or hydroxides are prone to reduction to highly soluble low-valent ions, which facilitates the release of As adsorbed on their surfaces (Zeng, 1996; Nickson et al., 1998; Smedley and Kinniburgh, 2002; Park et al., 2006; Rotiroti et al., 2021; Glodowska et al., 2021). There is a positive relationship between arsenic concentrations and Fe^{2+} in groundwater from the Yangtze River Delta region, which generally has Fe^{2+} concentrations higher than 10 mg/L (Gu and Zhen, 1995; Chen, 1998). Arsenic concentration in the groundwater within 5 km of the river bank along the Nanjing section of the Yangtze River, is generally higher than in the groundwater far from the Yangtze River bank, indicating waterlogged conditions and deposition of organic matter from the river promote As enrichment by this mechanism (Yu, 1999).

As concentrations in HAG from the Pearl River Delta (PRD) range from 2.8 to 161.0 μg/L (Huang et al., 2010; Wang et al., 2012). These occur in a neutral or weakly alkaline and reducing environment, mainly characterized by Na—Cl type water, with high NH_4^+ and organic matter concentrations (up to 390 mg/L and 36 mg/L, respectively), and low concentrations of SO_4^{2-} and NO_3^- (Jiao et al., 2010; Wang et al., 2012; Zhi, 2015). There is no obvious effect of salinity content on As enrichment. Enrichment of arsenic in this setting is again mainly from the release of primary arsenic in the aquifer media by reductive dissolution, and the infiltration and recharge to groundwater of surface irrigation with sewage (Huang et al., 2010). Irrigation by wastewater with As content up to 16.8 μg/L has resulted in As enrichment in soils and groundwater locally in the PRD (Huang et al., 2011). However, unlike the HAG in more arid inland basins of China – which generally results due to desorption of oxy-anions, under oxic, alkaline conditions (Currell et al., 2011), the affected groundwater in the PRD has significantly higher Fe and Mn concentrations, consistent with reductive dissolution. There is abundant iron oxide documented in the PRD sediments (Jiao et al., 2010).

There is also a negative correlation between dissolved organic carbon (DOC) concentrations (average 8.7 mg/L) and redox potential for the HAG, indicating that DOC concentration is a key factor promoting the formation of reducing environment in the groundwater system (Michael, 2013; Guo et al., 2013). Silt with abundant organic matter is widely present in the deltaic aquifers associated with these large river systems, intensifying the decomposition of iron oxides in anoxic groundwater (Zhang, 2011). In addition, there is a high content of iron and organic matter (e.g., COD) in the wastewater associated with the extensive industrial pollution discharges that have characterized the southern area of China for some decades, which may exacerbate generation of high HAG through reductive dissolution (Zhang et al., 2018b).

It has been reported that the occurrence of HAG in Shandong and Jiangsu provinces is widely distributed (Guo et al., 2013); however, apart from north plain of Shandong Prov. (Liu et al., 2013), there are few detailed hydrogeochemical studies on the mechanism(s) of enrichment in these areas. The spatial patterns of geogenic (fluoride and arsenic) contaminants in coastal groundwater is evidently complicated according to the source and geochemical behaviors. Local enrichment of F may in some cases largely represent the discharge of regionally deeply-circulated groundwater, which has undergone substantial geochemical evolution (largely toward alkaline Na-HCO_3 type water), which is associated with secondary F release by the mechanisms described above.

5.4. Urban growth / land-use change

Coastal zones of China are densely populated with large urban centers (Fig. 1), and urbanization and associated coastal development has continuously accelerated, along with economic growth (Li et al., 2017b). Urbanization results in land cover changes and alterations to hydrological systems, biogeochemistry, climate, and biodiversity (Grimm et al., 2008) which are broadly associated with deterioration of the eco-environment (Hua et al., 2020).

Adverse effects of urbanization on coastal groundwater quality are a fundamental concern in China's heavily populated coastal zones. This is related to various factors linked to urban growth/land-use change, including increases in groundwater extraction, land reclamation, wastewater infiltration, sewage exfiltration, and seawater intrusion (Onodera et al., 2008; Kløve et al., 2014; Shi et al., 2018). Urban growth drives the development of industry, agriculture, tourism, and fast-growing populations, leading to increased water demand (Flörke et al., 2018; Sanchez et al., 2020). Due to limited surface water resources (caused by drought or pollution), groundwater is often an essential water resource in coastal zones (Bricker et al., 2017; Wen et al., 2019). As a result, groundwater level decline, seawater/saltwater intrusion (Han et al., 2011, 2015a) and groundwater salinization (Han and Currell, 2018; Yolcubal et al., 2019) are associated with coastal urbanization. In addition, urban land-use change leads to an increased share of artificial, impervious surfaces impacting on recharge rates, mechanisms, and quality (Nuissl and Siedentop, 2021).

With the long-term coastal reclamation activities for urban growth in China, the length of the coastlines in the Bohai Bay-Yellow River Estuary Zone, the Yangtze River Estuary-Hangzhou Bay Zone and the Pearl River Estuary Zone increased from 717.6 km, 716.3 km, and 519.6 km in 1980 to 1252.8 km, 822.6 km and 780.9 km in 2018, respectively (Wang et al., 2021). The three zones are located in the most developed economic regions in China. Land reclamation and deep foundations may reduce groundwater discharge, changing coastal groundwater quality, and (in some areas) increase the groundwater levels in coastal areas (Hall, 1989; Stuyfzand, 1995; Jiao et al., 2001, 2006; Jiao, 2002; Guo and Jiao, 2007; Chen and Jiao, 2008). Water-rock interactions and water-concrete interaction are also important factors affecting coastal groundwater chemistry in highly urbanized areas (Chen and Jiao, 2008; Shi et al., 2018; Zhang et al., 2018c). Concrete material dissolution – particularly severe in the coastal zone due to high salt flux from sea-spray, adds Ca^{2+} and SO_4^{2-} to groundwater in urbanized mega-cities such as Shenzhen City (Shi et al., 2018). Industrialization was found to be the main driving force for the frequent occurrences of SO_4-type groundwater in this rapidly urbanizing area (Huang et al., 2018). Gradual release of heavy metals such as V, Cr, Mn, Ni, Cu and Cd from marine sediment to coastal groundwater has also been documented after reclamation in Shenzhen and Zhoushan Island (Chen and Jiao, 2008; Zhang et al., 2018c).

The infiltration from urban runoff carrying chemical pollutants from sanitary sewers, industrial activities, landfill leachate, and fertilizers also contribute to groundwater quality degradation (Arunprakash et al., 2014; Chitsazan et al., 2019; Nuissl and Siedentop, 2021). Chloride (Cl^-) and Nitrate-nitrogen (NO_3-N) are primary indicator contaminants of more complex mixtures of locally elevated pollutants (Bertrand et al., 2016). While fertilizer from agricultural activities (up to 652.7 mg/L) is the primary source of nitrate in shallow coastal groundwater in North China (Qin et al., 2013; Zhang et al., 2017; He et al., 2020), septic tanks have also been found to pollute groundwater rapidly, increasing NO_3-N from below 20 mg/L to about 120 mg/L following increased leakage in one reported instance (Lu et al., 2008).

5.5. Other potential drivers of coastal groundwater quality in China

Accelerated development of aquaculture in the last three decades has created multiple negative environmental impacts along China's coastline changing the ecological environment and biodiversity (Duarte et al., 2003; Li et al., 2017a; Zhao et al., 2018; Liang et al., 2019), degrading surface water and sediment quality (Yang et al., 2017; Zhang et al., 2019; Zhang et al., 2020a) and causing the shrinkage or disappearance of native coastal wetlands (Alonso-Pérez et al., 2003; Yan et al., 2017; Duan et al., 2020; Ding et al., 2020). In the process of aquaculture, excessive feeding can cause overnutrition and high turbidity in the water, and aquaculture farm effluents discharge high levels of organic matter into coastal waters, causing frequent algal blooms (Eng et al., 1989; Putheti et al., 2008; Ferrera et al., 2016).

From 1984 to 2016, the total area of China's coastal aquaculture ponds increased by 10,463 km^2, an increase of 327 km^2/yr (Ren et al., 2019). The area of aquaculture increased significantly in Guangdong, Shandong, Jiangsu, Liaoning, and Hebei provinces, accounting for 83% of the total expansion. Coastal waters of the Bohai Sea are the main aquaculture areas in northern China, and the area within 10 km of the nearshore buffer zone has expanded rapidly (Ren et al., 2019). The water quality problems brought by nearshore aquaculture mainly center on the input and complex transport process of nutrients, antibiotics, POPs and other pollutants, but there are few studies of the influence on coastal groundwater quality caused by nearshore aquaculture activities.

The huge South-North Water Diversion Project (SNWDP) has alleviated severe water shortages and the consequences of groundwater exploitation in northern China. Planned for completion in 2050, it will eventually divert 44.8 billion m^3 of water annually to the population centers of the drier north. Generally, surface water transfer is very effective in relieving pressure on groundwater resources - groundwater level recovery has already been observed in the NCP during 2015-2019 (Long et al., 2020; Zhang et al., 2020c). This may mitigate environmental problems such as reversible land subsidence (noting that some component of subsidence is irreversible), and seawater intrusion. As discussed above, both these processes are major contributors to groundwater quality problems in China's coastal aquifers – e.g. SWI drives significant salinization of fresh groundwater, while aquitard compaction releases fluoride into groundwater. Thus, there is potential to alleviate these issues in the coming years through water transfer. However, careful monitoring of the quality indicators surveyed in this review in areas of water level recovery will be needed to verify this.

Due to a lack of historical precedent for water transfer on this scale, the impacts of water diversion on groundwater quality remain largely unknown. Water transfer projects can modify the hydraulic connection between different water bodies in the receiving areas, such as interaction between surface water and groundwater, and affect the associated water and solute fluxes in aquifers (Zhu et al., 2019; Yuan et al., 2020). On the one hand, water diversion with fresh water sources may rapidly improve local groundwater quality through dilution and/or offsetting extraction that is driving degradation. On the other hand, water diversion may result in indirect effects on groundwater quality. The historical exploitation of groundwater has led to the thickening of the unsaturated zone (Cao et al., 2016) and the accumulation of significant quantities of 'latent' pollutants in the unsaturated zone, under the influence of intensive agriculture (and other pollutant sources). These pollutants may be intercepted by rising groundwater and pollute these aquifers as the groundwater table rises, thereby indirectly polluting groundwater (e.g. Wang et al., 2019b). As groundwater levels recover/rise, it is difficult to determine whether salinization associated with seawater intrusion will be slowed or reversed; however, preliminary studies show promise (e.g., Han et al., 2015a; Han and Currell, 2018).

6. Implications for coastal aquifer management and sustainability considerations in China

Our review has shown that groundwater quality problems in coastal areas of China include significant distributions of high fluoride, arsenic, and nitrate groundwater along with significant areas of salinization. The quality of coastal groundwater is controlled by primary geological environment, hydrological processes and hydrogeochemical (and biological) reactions. As shown here, it is often the combination of geogenic source, plus the influence of secondary hydrological changes associated with anthropogenic activity, which releases pollutants into groundwater. In response to the HFG and HAG caused by geogenic sources, the most important strategy for immediately reducing health risks and burdens is improving the reach and affordability of water treatment technologies to remove pollutants. Seeking alternative water sources may be another strategy, although this is likely to be difficult in many parts of China experiencing water stress. As described, land subsidence caused by groundwater over-exploitation from deep aquifers can be responsible for release of geogenic elements - artificial recharge is one strategy for controlling land subsidence (Lebbe et al., 1995; Hellauer et al., 2018).

Nitrate pollution, derived mainly from NH_4-fertilier, followed by manure and septic waste, requires further strategies to minimize nutrient loss (e.g. excess fertilizer application) as well as improved sanitation, especially in rural sewerage. Agriculture is by far the largest contributor to nutrient export to coastal ecosystems on a global scale (Howarth et al., 1996); and there is still (after many years of debate) urgent need to improve China's agricultural practices to maximize crop yield while reducing the rate of groundwater contamination from nitrogen fertilizer (e.g., Liu and Yang, 2012).

Many pollutants – including those covered in this review, but also others, for which the groundwater quality data are relatively scarce (such as POPs and heavy metals) are also associated with industrial discharges (Han and Currell, 2017) which are highly likely to reach groundwater in coastal areas due to strong ground-surface water connectivity. The Bohai Sea is estimated to have received 588 million tons of direct sewage discharge, including 7858 tons of chemical oxygen demand, 2531 tons of total nitrogen, 70 tons of total phosphorus, and 48.4 tons of petroleum pollutants in the most current official annual data (China Marine Ecology and Environment Bulletin, 2020). As a result, water quality has deteriorated in Liaodong Bay, Bohai Bay, and the Yellow River Estuary, with serious inorganic nitrogen and active phosphate pollution and eutrophication. If the problem of seawater intrusion (SWI) due to groundwater over-extraction in this area cannot be controlled, the long-term intrusion of polluted seawater into freshwater aquifers may inevitably result not only in groundwater salinization, but also groundwater contamination with additional pollutants that are difficult to remediate from these contaminated marine environments.

It is thus critical to formulate and enforce strict laws and regulations to control pollutant discharge at the source (Han et al., 2016a). The influence of policy changes such as new pollutant discharge and land use planning laws on groundwater quality, may however take decades or even centuries to take full effect, due to lag times in groundwater systems (Boulton, 2005; Currell et al., 2016). Thus, long-term planning and groundwater monitoring are fundamental. Moreover, management strategies should be forward-looking and incorporate analysis of relevant time scales and lag-times (Currell et al., 2016).

Our review found that the investigation of groundwater quality problems and the availability of data still lags far behind surface water – particularly for organic pollutants. It is thus necessary to strengthen monitoring to enable more complete characterization of groundwater quality and put forward corresponding countermeasures based on the analysis of its controlling factors. This is also critical to effective protections for surface water in coastal zones; as our review of SGD rates indicates, much solute flux in these surface waters ultimately derives from groundwater in coastal areas. Effective improvement of coastal groundwater quality should be therefore be based on integrated land-sea-river monitoring, accounting for the surface and subsurface fluxes of water and pollutants.

Finally, potential effects of climate change on groundwater quality remain poorly understood (Treidel et al., 2012). While climate change may not have major direct impacts at the local scale on local groundwater quality in coastal areas (notwithstanding some influence from rising sea-levels and changes to groundwater recharge rates), the indirect effects through intensification of water demand in climate-vulnerable regions such as northern China may be severe. Being subject to droughts, hurricane, storm surges and other meteorological phenomena that are intensifying under climate change, coastal aquifers may also be affected by major one-off disasters – e.g. storm surge floods resulting in large, temporary ingress of saline water into fresh aquifers, which may be difficult to reverse.

The coastal hydrological environment has been constantly influenced by human activities, such as intensive groundwater extraction, the rapid development of nearshore aquaculture, urbanization, land reclamation, and cross-basin water transfer projects implemented in recent years. However, there have to date been few if any studies reviewing the extent or impact of these processes on a national scale for coastal areas of China. It is imperative to establish a strict framework for maximizing environmental protection while achieving social and economic development; this review has provided a state-of-play with respect to coastal groundwater quality and a basis for management actions contributing to this goal.

Management of groundwater quality requires protection of aquifers from contaminants, and remediation and treatment of contaminated water. However, the remediation procedure to deal with groundwater pollution is generally very complicated and expensive, and it is difficult to achieve the purpose of remediation within a short period (e.g. Siegel, 2014). Groundwater pollution is often irreversible or difficult to repair. The protection of aquifers thus depends to a high degree on the formulation of strict laws and regulations to restrict actions which drive the degradation of groundwater quality in the first place. Coastal areas, as low-lying zones, are the regions where regional discharge from aquifers and rivers converge. As such management actions must involve coordination with inland regions to restrict unregulated wastewater discharge, land contamination and associated seepage along with local-level policies to reduce groundwater depletion responsible for seawater intrusion and further release and mobilization of geogenic elements in coastal areas. Detailed coastal groundwater protection plans should be formulated through the coordination of water and soil management departments in the coastal provinces, with mechanisms for wider regional cooperation with inland provinces as well.

CRediT authorship contribution statement

Dongmei Han: Conceptualization; Funding acquisition; Investigation; Data collection; Formal analysis; Writing-Original Draft; Writing-Review & Editing.

Matthew J. Currell: Conceptualization; Writing-Review & Editing.

Declaration of competing interest

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

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Appendix A. Supplementary data

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