


Comprehensive evaluation of shallow groundwater quality in Central and Southern Jiangsu Province, China

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Abstract Evaluation of the shallow groundwater quality in Central and Southern Jiangsu Province (CSJ) in China is important not only to public health but also to sustainable development and utilization of groundwater resources. In this work, 968 groundwater samples were collected by field surveys during 2006 to 2010 to investigate spatial distribution and extent of pollution. The single factor pollution standard index method, considering the values of both the natural groundwater background and the standard of safe drinking water, was used to assess the groundwater quality in the study area. Results showed that the shallow groundwater was severely polluted with Grade V and VI (i.e., severe and extremely severe pollution) groundwater covered about 1/3 and 1/4 of the study area, respectively. The main groundwater contaminants that cause the severe and extremely severe pollutions were manganese, iron, arsenic, and nitrogen. Findings from this work showed that natural hydrogeochemical processes were the main causes of the iron and manganese pollution, however, human

activities (e.g., industrial, agricultural, and domestic pollutions) mainly contributed to the nitrogen pollution in the study area.

Keywords Groundwater contamination · Background value · Manganese · Iron · Arsenic · Nitrogen · Influences of human activities

Introduction

With rapid economic development, population increase, and urban sprawl, contradiction between water supply and demand becomes a serious problem in many developing countries including China (Jiang 2009; Zhang et al. 2016). Surface water has been increasingly polluted due to industrial and domestic wastewater discharge and agricultural runoff. In May 2007, Taihu Lake, the most important freshwater supply source in Jiangsu Province, China, was choked by blue-green algae, causing panic as over 200,000 people found their tap water was undrinkable. This was China's most serious case of drinking water pollution to date (Shi et al. 2012). The deterioration of surface water quality has made the water supply crisis worsening in the developing countries, particularly China (Shi et al. 2012). Because of its generally better quality than surface water, groundwater is becoming a much more important nature resource to supply potable water for both rural and urban communities. Groundwater quality thus has received increasing attention in recent years.

Groundwater quality can be affected by both natural hydrogeochemical processes (Huang et al. 2013) and human activities such as agricultural irrigation (Chen et al. 2005), land use (Johnson and Belitz 2009), industrial effluents (Lapworth et al. 2012) and so on. For example,

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many studies indicate that excessive use of nitrogen-enriched fertilizers in agricultural areas has directly or indirectly affected groundwater quality, resulting in elevated nitrate concentrations in groundwater (Nolan et al. 1997; Chen et al. 2007). Heavy metals such as metalloids arsenic in groundwater may be resulted from both native geological processes and anthropogenic activities (Bhattacharya et al. 1997; Nickson et al. 1998; Fram and Belitz 2011; Rodríguez-Lado et al. 2013). The economic boom in China, therefore, may also have the deterioration effect caused by pollution on the quality of groundwater due to industrial and domestic pollution and the application of fertilizer and pesticide in agriculture. Particularly, the shallow groundwater is more susceptible to pollutions because of the shallow depth of water level. Comprehensive surveys and evaluations of shallow groundwater quality thus are extremely important in China.

As one of the most developed parts of China, the area of Central and Southern Jiangsu Province (CSJ) belongs to the hinterland of the lower Yangtze Delta that has a total area of 49,000 km² (Fig. 1a). Part of the CSJ area, especially the south (including Suzhou, Wuxi and Changzhou, namely Su–Xi–Chang area), suffered from severe ground settlement caused by extensive groundwater exploitation in the past several decades (Shi et al. 2007; Zhang et al. 2010). Numerous efforts have been devoted since 1995 to control the land subsidence and ground fissures. By the end of 2005, a ban of deep groundwater pumping in Su–Xi–Chang area was fully established and the land subsidence rate was slowed down with the gradual rise of groundwater level. In order to supply adequate water resources, the shallow groundwater in the micro-confined aquifer began to be exploited in the area (Hu et al. 2009). To solve the imbalance of water supply and demand in the CSJ area, previous studies have measured the groundwater quantity (levels, flow directions, fluxes, etc.) through hydrogeological field survey at different scales, such as 1:200,000 or 1:250,000 regional hydrogeological surveys (see the reports of The First Group of Hydrogeologic and Engineering Geology Survey in Jiangsu province, 1978–1980). Comprehensive evaluation of the groundwater quality in the CSJ area, however, has not been conducted in the previous studies.

During 2006 to 2010, a comprehensive study was carried out to survey and evaluate shallow groundwater quality in the CSJ area. A total of 968 shallow groundwater samples were collected from the area (Fig. 1b). Each sample was analyzed for 72 water quality indicators. The single factor pollution index method was used to evaluate the pollution levels of the shallow groundwater in the study area. The main water quality indicators of the area were then further investigated to determine potential pollution sources. The specific objectives of this study were as

follows: (1) evaluate the overall shallow groundwater quality; (2) quantify the pollution levels and determine the main pollution indicators of the shallow groundwater; and (3) determine the potential causes of the shallow groundwater pollution in the study area.

Materials and methods

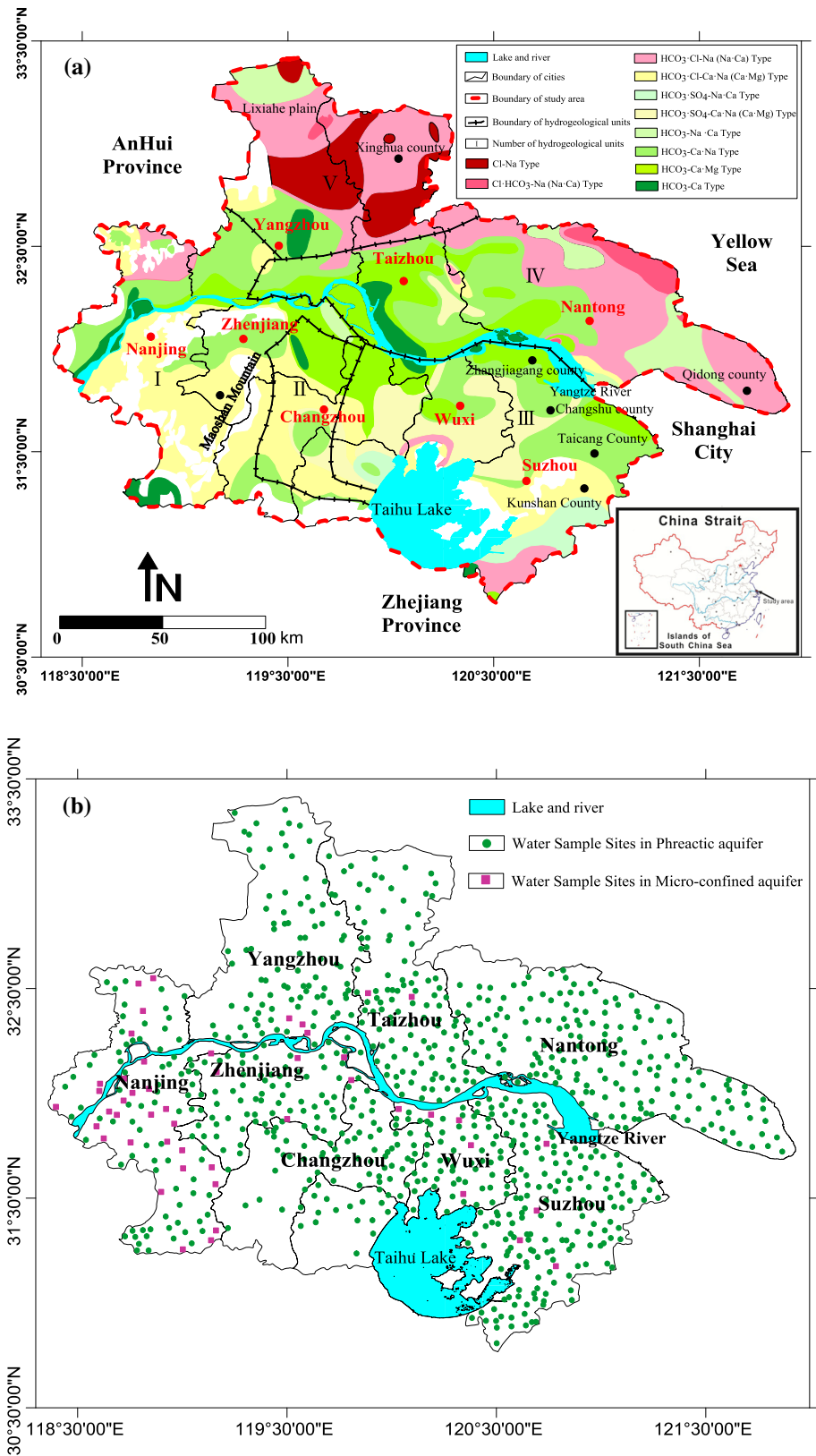
Study area

The study area is in the typically subtropical monsoon climate zone of China with the annual average temperature of 15.6 °C and annual precipitation of 900–1200 mm. There are several rivers, which are belonging to the Yangtze River Catchment and Huai River Catchment. The most important river and lake are the Yangtze River and the Taihu lake (Fig. 1a). The study area, with the slope gradient of ground surface about 1/10 000, mainly comprises wide alluvial and marine plain. Only a few low mountains and hills are located in the south and west, with the average elevations are less than 6 m above the sea level.

The quaternary deposits of fluvial, lake, lagoon, and marine origins, which comprise the aquifers, are widely distributed in the study area. Based on the sedimentary characteristics, hydrogeological conditions, aquifers distributions, and groundwater flow fields, the study area can be classified into five hydrogeological units from Unit I to Unit IV, corresponding to the low mountain and hilly unit, piedmont plain unit, Taihu lake plain unit, Yangtze river delta plain unit, and Lixiahe plain unit, respectively (Fig. 1a; Table 1) (Wu and Fang 1991). In the vertical direction, quaternary sediments are primarily composed of coarse silt, fine sand, medium coarse sand, and pebbled coarse sand with some clay and loam interlayers. The sediments vary from fine to coarse repeatedly and constitute depositional sequence, corresponding to the alluvial, fluvial facies, and sedimentary cycle. Some interlayered lake and river deposits are rich in clay and well consolidated. Most of the lagoonal and marine deposits, however, are soft to plastic, dark clay. Both of these layers have poor hydraulic conductivity and can be treated as aquitard or semi-confining units. As a consequence, the micro-confined aquifer is widely distributed in the Su–Xi–Chang area and Nanjing–Zhengjiang area (Zhang et al. 2013).

There are two aquifer systems in the study area: shallow aquifer system (the groundwater type is phreatic water and micro-confined water) and middle-deep aquifer system (the groundwater type is confined water, including the first, second, third and fourth confined aquifers) (Zhang et al. 2010). This shallow aquifer system is continuous in the study area, with the buried depth ranging from 5 to 50 m.

Fig. 1 Map of the study area in Jiangsu Province: **a** hydrochemical type of groundwater and hydrogeological units division and **b** distribution of the sampling sites



The groundwater level in the shallow aquifer nearly remains steady, only fluctuating within 1–3 m. In this work, only the shallow groundwater is considered. In

general, the regional groundwater flow direction under natural condition is from west to east and the groundwater velocity is relatively very slow with the hydraulic gradient

Table 1 Description of the hydrogeology units of the study area (modified from Wu and Fang 1991)

	Hydrogeological unit	Area (km ²)	Topography characteristics
II	Low mountain and hilly unit in Nanjing–Zhenjiang area	10,979	Low mountains and hills of Nanjing–Zhenjiang, Yixing–Liyang area, Intermontane basin, the floodplains of the Yangtze river
III	Piedmont plain unit	2679	Piedmont plain of Maoshan area
IIII	Taihu lake plain unit	13,114	Water network plain of Taihu lake
IIV	Yangtze River delta plain unit	14,521	Alluvial plain of delta
I V	Lixiahe plain unit	7707	Dishing low-lying plain

about 2/10,000. For the shallow aquifer system, the main recharge is the rainfall and irrigation water infiltration due to the close connection with the surface environment, and the main discharge is via evaporation, leakage to the middle-deep aquifer system and exploitation (Wu and Fang 1991).

The groundwater hydrochemical-type distribution in the study area shows an obvious trend and zoning from west to east (Fig. 1a). The groundwater quality in hilly area and piedmont undulating plain of the Midwest is better due to the faster rate of water cycling. In the Taihu lake plain, the concentration of Na⁺ and Cl⁻ increases gradually in shallow groundwater, causing freshwater transform into brackish water. The hydrochemistry type in Yangtze coastal region indicates the surface water influence to the shallow aquifers. Brackish water and saline water is mainly distributed in the Lixiahe plain of the northern area due to the lagoon sediments. The rest of brackish water and saline water distributed in the east of the study area is due to the seawater intrusion.

Field sampling

The survey of groundwater quality was carried out during 2006–2010 in the study area. Firstly, a groundwater sampling plan is designed meticulously after understanding of the hydrogeology and flow dynamics of the shallow groundwater system combined with survey of probably contamination sources to collect representative samples. Monitor-pumped wells and some water supply wells installed by the local government were selected as the sampling sites. The sampling density was controlled into about 2 samples per 100 km². Furthermore, electric submersible pumps are chosen as the sampling devices. The buried depth of the shallow groundwater system is usually about 4–5 m, however, the range of wells depths and the positions of the screening of the wells can be variable over distances due to the variation of hydrogeology. As a result, the sampling intake should be placed at the desired sampling point across or just above the screened interval. In accordance with the standard requirements of DD 2008-01, totally 968 water samples from the phreatic and micro-

confined aquifers were collected. The distribution of the sampling sites is shown in Fig. 1b.

Sample preparation and analysis

Due to the flow velocity of the shallow groundwater system under natural condition is relative small, low-flow purging (sometimes referred to as micro-purging) was recommended before sampling to minimize the disturbance of the stagnant water column above the screened interval. On-site water quality measurements (including temperature, pH, specific electrical conductance, dissolved oxygen and oxidation–reduction potential) were taken predominantly to monitor effective purging of water at the sampling point before sample collection and to measure unstable parameters that cannot be subsequently reliably determined in the laboratory. The samples were stored in plastic bottles and brown glass. Before sampling, containers were rinsed with groundwater to be taken. After sampling, one plastic bottle was acidified with HNO₃, and all containers were sealed immediately. The sample in brown glass for determining the organic compounds was filled without air to avoid evaporation, oxidation, and reduction (Li et al. 2013). Before loading, the sampled containers were labeled and packed into incubators.

A bulk chemical analysis was carried out for total of 72 testing indicators, including 33 inorganic indicators and 39 organic indicators. They were further divided into 5 categories (see Table 2) including: (1) general chemical indicators; (2) inorganic toxicology indicators; (3) toxic heavy metal indicators; (4) volatile organic indicators; and (5) semi-volatile organic indicators. Volatile organic compounds in water were analyzed by purge and trap gas chromatography/mass spectrometry. Particularly, organic chlorine pesticides and polychlorinated biphenyls were analyzed by electron capture detector gas chromatography/mass spectrometry. Organophosphorus pesticides were analyzed by flame ionization detection gas chromatography/mass spectrometry. High-performance liquid chromatography coupled to an ultraviolet, diode array or fluorescence detector (HPLC/UV-FLD) has been used to detect polycyclic aromatic hydrocarbons (PAHs). More

Table 2 Classification of shallow groundwater pollution assessment indicators

General chemical indicators (15)	pH, TDS, Total hardness, Sulfate, Chloride, Fe, Mn, Cu, Zn, Al, COD, Sulfide, Na + K, Se, Ammonium
Inorganic toxic indicators (6)	Nitrate, Nitrite, Iodide, Fluoride, Be, Bo
Toxic heavy metal (ions) indicators (12)	Hg, As, Cd, Cr, Pb, Ba, Ni, Mo, Ag, Co, Antimony, Thallium
Volatile organic indicators (24)	Trichloromethane, Carbon tetrachloride, 1,1,1-Trichloroethane, Trichloroethylene, Tetrachloroethylene, Dichloromethane, 1, 2-Dichloroethane, 1,1,2-Trichloroethane, 1,2-Dichloropropane, Bromoform, Chloroethylene, 1,1-Dichloroethylene, 1,2-Dichloroethylene, Bromo dichloromethane, Dibromochloromethane, Chlorobenzene, O-dichlorobenzene, Para-dichlorobenzene, Trichlorobenzene, Benzene, Methylbenzene, Ethylbenzene, Xylene, Styrene
Semi-volatile organic indicators (15)	Hexachlorobenzene, Total BHC, γ -BHC, Total DDTs, Heptachlor, DDVP, Parathion-methyl, Malathion, Dimethoate, Benzo(a)pyrene, Naphthalene, Anthracene, Fluoranthene, Total Polychlorodiphenyls, PCB

details about the requirement for Preservation/Handling of Samples and analytical protocols are referred the standards of China Geological Survey (DD 2008-01, 2008). For example, NH_4^+ was detected by using the salicylic acid spectrophotometric method; NO_3^- was determined by ion chromatography. Total Mn and Fe were analyzed by using flame atomic absorption spectrophotometry.

QA/QC

All target analytes in procedural blanks were below their corresponding limits of quantification (LOQs). A procedural blank and a procedural recovery sample were also analyzed for each batch of samples to check laboratory contamination and accuracy. For the inorganic compounds, the mean error of anion–cation balance was less than 3%. For the organic compounds, the mean procedural recovery ($n = 3$) ranged from 70 to 130% for VOCs, and 65 to 130% for SVOCs, respectively.

Pollution assessment methods

Groundwater pollution assessment is a pollution-level assessment based on background value or pollution initial values, which is usually to reveal the cause or mechanism of changes in water quality. The single factor pollution index method was selected to evaluate the water quality of the samples because of its clear physical significance, simple calculation process, and wide adaption in China (Li et al. 2009, 2013). There are graded Class I to VI based on the threshold values defined in the *Standard for groundwater quality (DZ/T 0290-2015)*. Class III is corresponded to the drinkable quality water, which means Class I to III are classified into drinkable water and Class IV and V are classified into undrinkable water. In order to consider both the natural groundwater background and the Class III standard, the single factor pollution index method has recently been modified and renamed as the single factor

pollution standard index method (Li et al. 2013). The index is calculated as following:

$$P_{ki} = \frac{C_{ki} - C_{0i}}{C_{IIIi}} \quad (\text{Except for pH value}) \quad (1)$$

$$P_{ki} = \frac{C_{imin} - C_{ki}}{C_{ki} - C_{imax}} \begin{matrix} (C_{ki} \leq \overline{CO}_i) \\ (C_{ki} > \overline{CO}_i) \end{matrix} \quad (\text{for pH value}) \quad (2)$$

where P_{ki} —pollution index of the i th indicator for the k th sample. C_{ki} —analyzed result of the i th indicator for the k th sample. C_{0i} —the background value of the i th indicator for the k th sample for the inorganic compounds; the detection limit of indicators for organic compounds. C_{IIIi} —limit value of the i th indicator in the *Standard for groundwater quality*. \overline{CO}_i —Standard mean value of pH. C_{imax} —upper limit of the standard value of pH. C_{imin} —lower limit of the standard value of pH.

Equations (1) and (2) are utilized to calculate the pollution index P_{ki} for each indicator (72 total, see Table 2) for each water sample, which is further classified as Grade I to VI based on the threshold values in Table 3. The single factor pollution standard index method thus can be easily used for comprehensive pollution assessment (i.e., $P_k = -\max\{P_{ki}\}$) to evaluate the pollution index for each sample.

To assess pollution status and calculation of contamination indices, background analyses are essential for distinguishing between chemicals present in the environment due to naturally occurring geochemical processes and sources, or to human activities. High concentration in the water samples do not automatically imply that contamination has occurred, but may reflect the natural mineralogical composition of the parent geological material (Mendizabal and Stuyfzand 2009). Background chemicals are derived from natural and anthropogenic sources not associated with chemical releases. It is noted that the background values are varied with hydrological conditions and have different values for different hydrogeological

Table 3 Grading standards for the single factor pollution standard index (Li et al. 2013)

Pollution category	No pollution	Light pollution	Moderate pollution	Less severe pollution	Severe pollution	Extremely severe pollution
Grading level	I	II	III	IV	V	VI
Index range	$P \leq 0$	$0 < P \leq 0.2$	$0.2 < P \leq 0.6$	$0.6 < P \leq 1.0$	$1.0 < P \leq 1.5$	$P > 1.5$

units (Zeng 1997). The study area was subdivided into totally 20 hydrochemical zones based on the sedimentary characteristics, hydrogeological units and hydrogeochemical characteristics (Fig. 2). Here, the background values for the inorganic indicators (i.e., K^+ , Na^+ , Ca^{2+} , Mg^{2+} , Cl^- , HCO_3^- , SO_4^{2-} , NO_3^- , NO_2^- , NH_4^+ , COD, F, As, pH, and TDS (expressed in mg/l) were determined based on the reported values of 348 groundwater samples, which were taken and analyzed during the 1970s to 1980s (see the reports of The First Group of Hydrogeologic and Engineering Geology Survey in Jiangsu province 1978–1980). The background values for the heavy metals indicators (more details see Table 4), including Cu, Zn, Pb, Cd, Ba, Co, Cr, Hg, Mo, Ni, Se, Sulfide, and Cyanide, were determined by using statistical analysis as described in Li et al. (2013). Meanwhile, the background values of organic

pollution indicators (Table 2) were set to zero. All statistical analysis was conducted by Excel 2003 (Microsoft Office).

Results and discussion

Statistical analysis of individual indicators

Statistical analyses of individual indicators were preliminarily carried out to describe the general physiochemical characteristics and to further understand which indicators have high pollution induces. In order to find out whether it is suitable for drinking, the maximum values were compared with the acceptable limits which are the Class III standards for drinking in the *Standard for groundwater quality (DZ/T 0290-2015)* (Ministry of Land and Resources

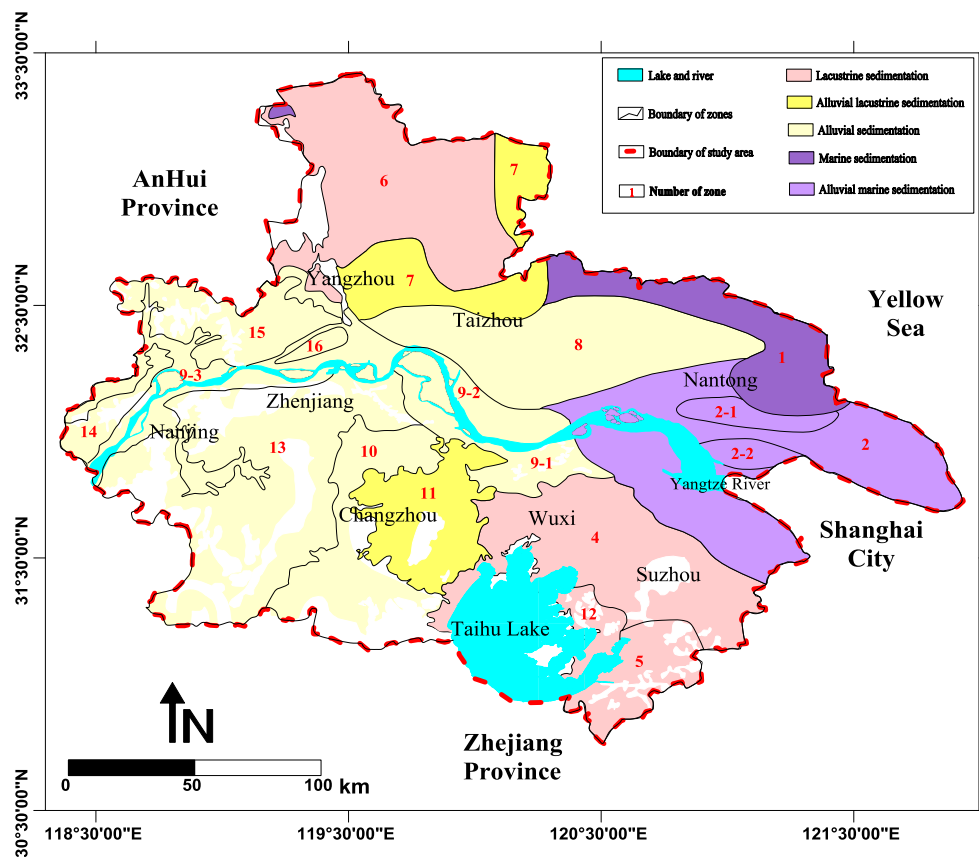
**Fig. 2** Hydrogeological zone with different background values for shallow groundwater system in the study area

Table 4 Background values of different hydrogeological zones (see Fig. 2) for the shallow groundwater system in the study area

Zone number	1	2	2-1	2-2	3	4	5	6	7	8	9-1	9-2	9-3	10	11	12	13	14	15	16
Na ⁺ + K ⁺	6371.40	292.37	95.22	114.99	89.18	89.50	91.37	185.28	230.43	237.60	76.14	50.44	90.12	105.95	73.56	71.43	57.98	60.31	87.23	91.86
Fe ²⁺ + Fe ³⁺	0.48	1.21	0.78	0.45	0.05	0.05	0.03	0.10	0.00	0.16	0.13	1.29	0.37	0.00	0.21	0.01	0.14	0.14	0.08	2.53
Chloride	11985.50	360.33	93.63	101.60	129.47	109.64	90.86	173.40	414.45	264.00	148.52	61.12	73.62	164.05	104.27	122.05	106.10	70.92	93.74	88.60
Sulfate	829.63	20.97	94.50	97.90	87.81	74.76	44.74	125.46	36.98	20.20	57.27	19.20	65.71	97.27	64.36	77.59	134.50	93.05	74.14	70.48
Nitrate	4.13	1.24	38.16	31.40	14.72	19.34	33.32	0.21	0.00	0.46	38.16	0.00	20.17	55.01	45.02	80.49	31.59	37.84	28.02	7.37
Nitrite	0.00	1.33	0.01	0.00	0.01	5.77	0.14	0.01	0.01	0.04	0.16	0.00	0.13	0.30	1.57	0.29	0.02	0.00	0.01	0.04
Ammonium	17198.84	4.27	0.06	0.00	0.04	0.02	0.10	0.23	0.12	5.00	0.06	12.40	0.43	0.20	0.06	0.05	0.11	0.03	0.01	6.50
COD	251.91	3.47	1.07	1.06	1.93	1.33	2.94	0.73	0.60	2.90	1.20	3.92	2.06	1.00	1.94	1.59	0.97	1.42	1.31	3.40
Fluoride	0.13	0.00	0.28	0.23	0.14	0.18	0.16	0.79	0.46	0.24	0.19	0.10	0.07	0.04	0.05	0.11	0.16	0.39	0.35	0.14
TDS	0.00	1185.00	800.34	726.05	873.69	678.92	562.79	960.85	1190.00	823.67	663.92	658.20	596.28	846.00	777.29	614.00	658.19	647.00	692.09	727.00
TH	0.19	23.94	419.42	354.34	30.66	21.64	16.49	25.45	33.95	22.77	22.41	27.64	19.63	27.36	21.13	19.76	23.86	313.40	309.05	329.11
Mn	1.37	0.40	0.35	0.53	0.22	0.09	0.49	0.44	0.53	0.25	0.19	0.48	0.24	0.12	0.11	0.06	0.02	0.22	0.10	0.65
Al	0.00	0.00	0.00	0.00	0.03	0.04	0.10	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.12	0.00	0.00	0.00	0.00
Sulfide	0.19	0.15	0.00	0.00	0.00	0.61	0.00	0.00	0.21	0.12	0.00	0.00	0.00	0.10	0.10	0.15	0.31	0.00	0.00	0.00
As	1.37	1.94	4.51	4.46	4.00	0.92	1.71	2.63	0.66	0.83	0.00	74.00	3.16	1.33	1.42	1.75	2.25	2.32	2.12	5.32
Hg	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Cr	11.66	7.21	12.74	13.03	11.20	8.65	12.25	6.37	12.56	9.72	7.98	6.09	11.16	6.01	4.84	9.12	8.53	6.94	6.34	6.85
Cd	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Cu	1.58	1.38	2.44	1.35	0.87	0.58	0.36	1.23	1.64	0.63	60.00	0.98	2.33	20.00	60.00	0.09	1.87	2.51	2.21	1.21
Pb	0.00	0.21	0.30	0.26	0.00	0.00	0.00	0.06	0.00	0.00	1.40	0.15	0.23	0.11	0.00	0.00	0.15	0.18	0.16	0.20
Zn	34.14	16.26	19.40	22.00	8.18	8.82	2.27	6.40	12.93	21.02	93.30	11.79	20.65	17.77	49.60	4.60	14.80	31.90	26.85	18.00
Se	3.79	3.78	3.28	1.97	1.84	1.35	0.94	1.97	1.05	1.75	1.48	2.26	0.51	0.81	1.41	3.32	1.38	1.00	0.39	0.62
Be	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Ba	15.97	57.42	28.56	26.40	82.48	59.44	63.71	79.20	78.59	27.72	57.58	172.57	88.95	74.06	77.89	71.05	96.36	67.20	76.38	182.17
Ni	1.33	3.22	4.80	0.67	4.59	2.68	1.40	3.23	4.19	3.20	3.65	3.27	5.98	3.69	5.45	1.15	5.10	4.23	3.93	4.23
Mo	1.56	1.56	0.44	0.57	0.51	0.54	1.71	0.90	0.76	0.78	0.44	1.80	2.73	0.54	0.39	5.98	0.87	2.02	1.41	1.06

(1) mg/L unit for Na⁺ + K⁺ + Fe²⁺ + Fe³⁺, Chloride, Sulfate, Nitrate, Nitrite, Ammonium, COD, Fluoride, TDS, Total hardness (TH), Mn, Al, Sulfide; (2) µg/L unit for the ions of As, Hg, Cr, Cd, Cu, Pb, Zn, Se, Be, Ba, Ni, and Mo

of China 2016) of China. The statistical results are presented in Fig. 3 and Table 5, Fig. 4, and Table 6 for inorganic and organic indicators, respectively.

As shown in Table 5 and Fig. 3, there are total 11 inorganic indicators with more than 10% recovery rate of less than Class III (corresponding to unsuitable for drinking) samples, including 7 general chemical indicators (i.e., total Fe, Mn, pH, Total hardness, TDS, COD, and ammonium); 3 Inorganic toxicology indicators (i.e., nitrate, nitrite, iodide); and 1 toxic heavy metal indicators (arsenic).

Similarly, Table 6 and Fig. 4 both show that there are total 10 organic indicators in the less than Class III (corresponding to unsuitable for drinking) samples, including Carbon tetrachloride, Trichloroethylene, 1,2-Dichloroethane, 1,1,2-Trichloroethane, 1,2-Dichloropropane, Chloroethylene, 1,2-Dichloroethylene, Benzene, Naphthalene, PCB.

Evaluation of the shallow groundwater quality

The single factor pollution indices P_{ki} for each indicator (72 total, see Table 2) were statically analyzed to evaluate the shallow groundwater quality in the study area. Due to the page limitation, detailed distributions of the pollution indices P_{ki} for each indicator in the area are not listed. The comprehensive shallow groundwater quality assessment results, which combined index results of all the 72 indicators, were thus used to evaluate the shallow groundwater quality in the study area.

Figure 5 shows that the frequencies (referred to as pollution contribution rate) of individual compounds for Grade V and VI samples. As shown in Fig. 5, the main contributors to the shallow groundwater pollutions in the study area were Mn, total Fe, As, nitrogen (including nitrate, nitrite, and ammonium), and COD, which are mainly the inorganic contaminants. The contribution of organic contaminants to the severe (Grade V) and extre-

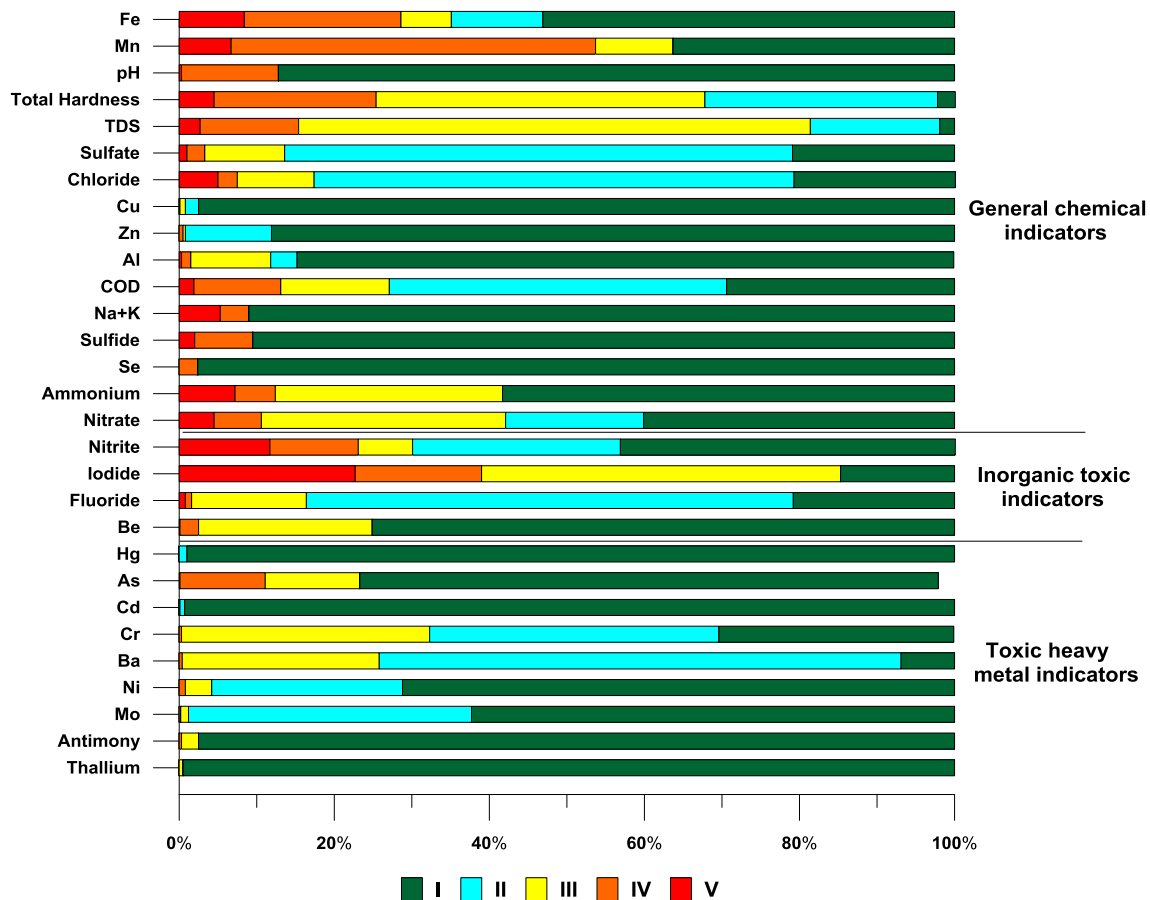


Fig. 3 Statistical analyses of individual inorganic indicators based on the classification standard defined in the *Standard for groundwater quality (DZ/T 0290-2015)* of China. There are five classes (Class I to VI). Among them, Class III is corresponded to acceptable limits of the

drinkable water quality, which means Class I to III are classified into drinkable water and Class IV and V are classified into undrinkable water

Table 5 Detection frequency of the inorganic toxic indicators and toxic heavy metal indicators in the samples

Indicator	Mean concentration ^a (mg/L)	Detection frequency (%)
Nitrite-N	1.41	57.2
Nitrate-N	8.30	94.2
Iodide	0.08	85.5
Fluoride	0.38	94.5
Be	0.00013	0.4
Bo	0.18	45.6
Hg	0.0001	1.0
As	0.01	88.7
Cd	0.0005	0.9
Cr	0.01	94.6
Pb	0.001	28.1
Ba	0.09	99.0
Ni	0.0046	87.9
Mo	0.0019	90.6

^a Mean concentration represents the mean value of the corresponding concentration in the detected samples

mely severe (Grade VI) pollution indices was relatively small. The major organic contaminants classified into Halogenated hydrocarbons and polycyclic aromatic hydrocarbons. These findings are consistent with the previous results as shown in Figs. 3 and 4.

Figure 6 represents the spatial distribution of comprehensive evaluation for the shallow groundwater pollution, using different colors to represent the zones of different pollution levels (as categorized in “Grades”). As shown in the map (Fig. 6), the shallow groundwater in the study area was all polluted without any Grade I (none polluted) samples. Light pollution samples of Grade II only accounted for 1.03% of the total samples. Moderate pollution samples of Grade III accounted for 16.04%. Less severe pollution samples of Grade IV accounted for 22.41%. Severe pollution samples of Grade V accounted for 27.19% and extremely severe polluted samples of Grade VI accounted for 33.33% of the total samples. From the point of the area distribution, Grade V and VI samples covered 13385.05 and 10920.69 km², respectively,

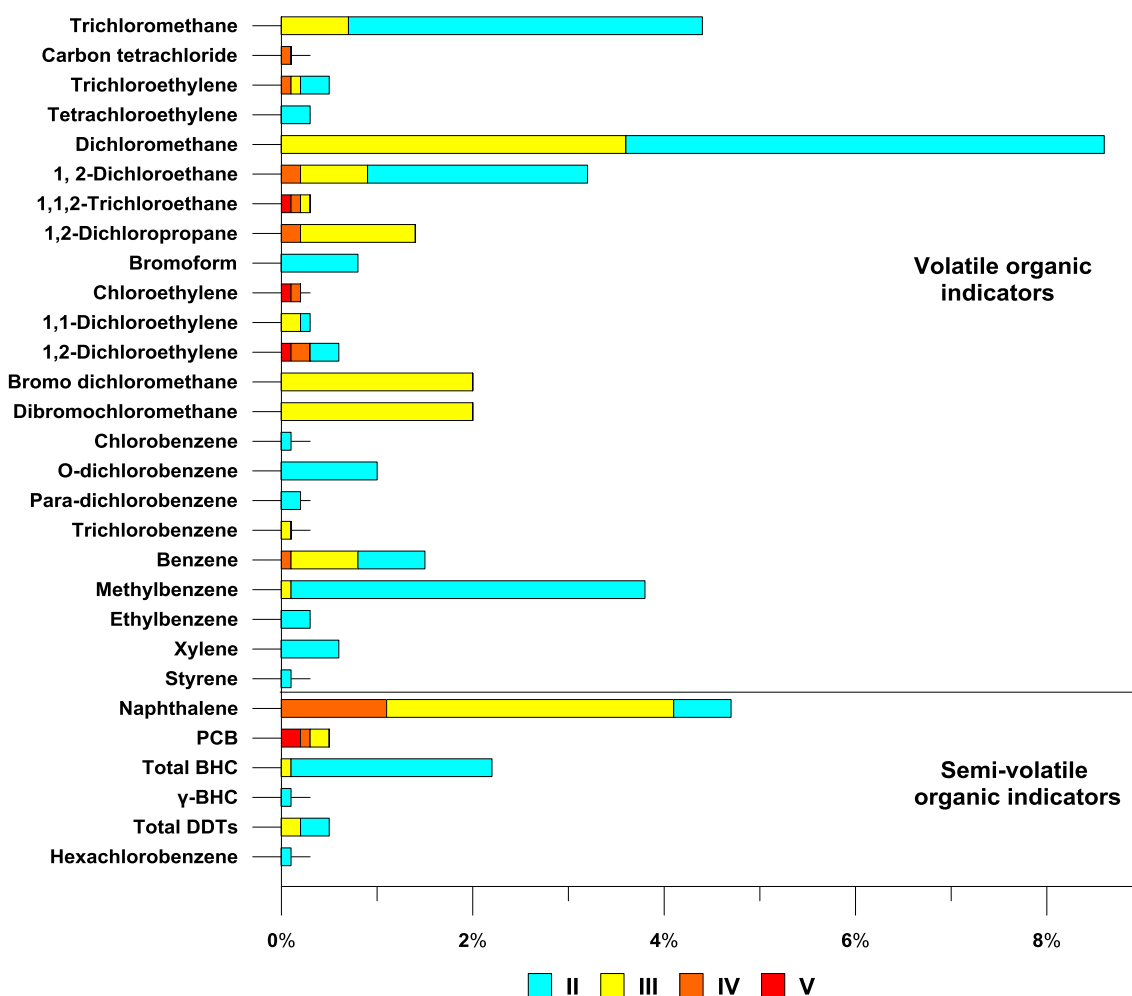


Fig. 4 Statistical analyses of individual organic indicators based on the classification standard defined in the *Standard for groundwater quality (DZ/T 0290-2015)* of China. It is noted that, Class I is not shown here to make it more readable

Table 6 Detection frequency of organic indicators in the samples

Indicator	Concentration ($\mu\text{g/L}$)		Detection frequency (%)
	Minimum	Maximum	
Trichloromethane	0.53	30.62	4.44
Tetrachloromethane	6.07	6.07	0.1
1,1,1-Trichloroethane	nd	nd	nd
Trichloroethylene	0.7	124	0.52
Tetrachloroethylene	0.58	2.76	0.31
Dichloromethane	0.53	14.6	17.5
1,2-Dichloroethane	0.5	34.7	3.31
1,1,2-Trichloroethane	0.75	17.1	0.31
1,2-Dichloropropane	0.5	17.7	1.55
Bromoform	1	7.72	0.83
Chloroethylene	28.6	382	0.21
1,1-Dichloroethylene	2.37	4.2	0.31
1,2-Dichloroethylene	0.52	785.1	0.62
Chlorobenzene	10.8	10.8	0.1
1,2-Dichlorobenzene	0.55	1.9	1.03
1,4-Dichlorobenzene	0.97	1.97	0.21
Total trichlorobenzene	5.15	5.15	0.1
Benzene	0.55	49.05	1.55
Methylbenzene	0.51	446.5	3.82
Ethylbenzene	0.51	5.47	0.31
Xylene	0.87	11.08	0.62
Styrene	0.92	0.92	0.1
Dimethoate	nd	nd	nd
Total benzex	0.02	1.070	2.17
γ -BHC	0.056	0.056	0.1
Total DDTs	0.034	0.180	0.52
Hexachlorobenzene	0.028	0.028	0.1
Heptachlor	nd	nd	bd
DDVP	nd	nd	nd
Prarathion-methyl	nd	nd	nd
Malathion	nd	nd	nd
Benzo(a)pyrene	nd	nd	nd
Naphthalene	nd	120	5.26
Anthracene	nd	nd	nd
Fluoranthene	nd	nd	nd
Total polychlorodiphenyls	nd	0.64	0.57
Bezo(b)flouranthene	nd	nd	nd

accounting for about 1/3 and 1/4 of the total study area. Overall, the pollution of shallow groundwater in the north of the Yangtze river was more severe than that in the south, and the relatively light pollution areas were mainly distributed in the piedmont plain (Fig. 6).

Discussion of the contaminants source

The study area is among the most populated regions in China (with more 50 million of total population in 2012) and

underwent booms in industry and agriculture in the past several decades (Du et al. 2014; Shi et al. 2007, 2012). In addition to the heavy industrial activities, the study area also showed intensive agricultural activities. The human activities, not exclusively, might pollute the shallow groundwater through processes such as sewage discharge and solid waste leaching, industrial waste disposal and wastewater discharge, and agricultural runoff and waste disposal (Katz et al. 2009).

The comprehensive evaluation of the shallow groundwater quality showed that the main contaminants that cause

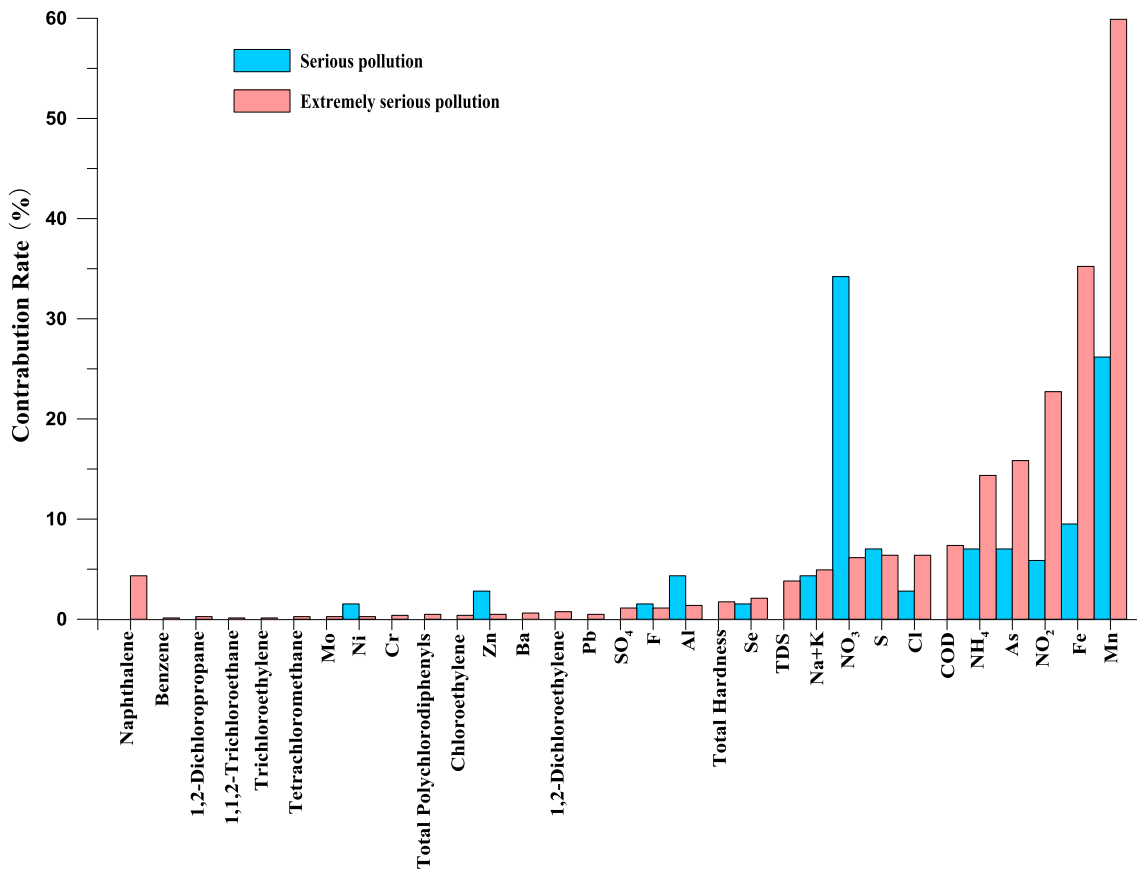


Fig. 5 Contribution rate of individual pollution indicator for the corresponding pollution levels in the study area

the severe and extremely severe pollutions in the study area were manganese, iron, arsenic, and nitrogen. The distributions and sources of these main contaminants thus are further discussed in following sections.

Iron and manganese

The distributions of total iron and manganese pollutions are shown in Fig. 7a, b, respectively. The results showed heavy pollutions of shallow groundwater by iron and manganese in the study area. The severely iron-polluted areas include the part along the Yangtze River, east part of the Maoshan Mountain, areas from Changshu County to Kunshan County, coastal areas of the eastern Qidong County and the south and east parts of Xinghua County. Among these, the areas of severe (V) and extremely severe (VI) pollutions were 2261.55 and 1858.87 km², respectively. Compared to the iron pollution, manganese pollution of shallow groundwater was worse with the total areas of severe and extremely severe pollutions of 4130.0 and 3701.34 km², respectively.

The main sources of the iron and manganese pollutions in the shallow groundwater could be attributed to natural hydrological and geochemical process. Because iron and

manganese are the main component in the crust (Fe 4.65%, Mn 0.1%), weathering, decomposition, and lixiviation of the large area of bedrock could release both iron and manganese into the shallow groundwater. In general, the manganic contamination level was higher than total ferrous contamination level because manganese ions were more stable than ferrous ions (Fig. 7a, b). Meanwhile, the ferrous and manganic contamination levels in the western mountain and hill areas where bedrocks are exposed to air (oxygen), were generally lower than that in the eastern region due to the fact that the ferrous and manganic ions can be easily transported in soluble compound forms, particularly in the reducing environment (Homoncik et al. 2010). The high content of iron and manganese in the Lixiahe plain (Fig. 7a, b) was due to the low-lying terrain, shallow buried depth of groundwater, poor hydrodynamic condition, and the high evaporation of shallow groundwater (Fig. 1a; Table 1). In the east of Suzhou City and the east of the Taihu Lake, the deposition of the shallow strata was filled with large amount of organic matters of coastal tidal flat facies and limnetic facies, leading to the higher content of iron and manganese in the shallow groundwater in the region (Fig. 7a, b).

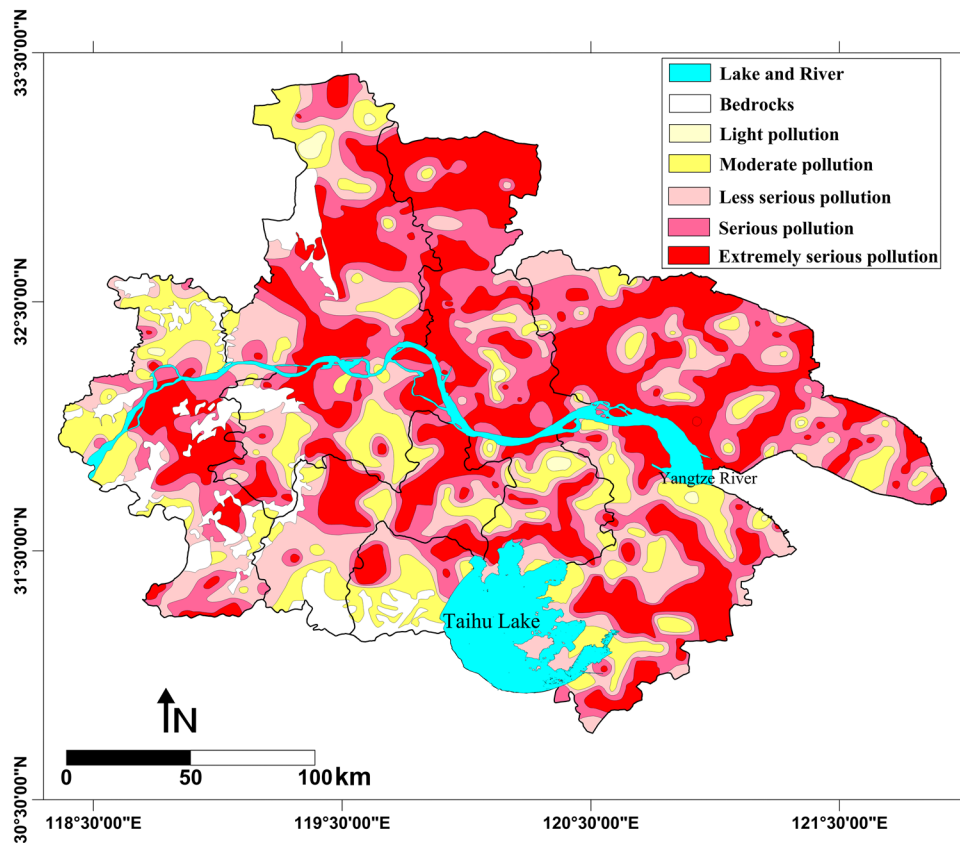


Fig. 6 Comprehensive evaluation of the shallow groundwater pollution using single factor pollution index method

Arsenic

The distribution of arsenic pollution is illustrated in Fig. 7c. The unpolluted areas were mainly in Nanjing, Zhenjiang, Changzhou and part of Nantong City, with total area of 6200.25 km². The medium and moderate polluted areas were distributed all over the study area with about 27,600 km². The severe (V) and extremely severe (VI) polluted areas were 1599.62 and 979.55 km², respectively.

The arsenic pollution of the shallow groundwater in this study area could be attributed to natural hydrogeochemical processes (Mariner et al. 1996; Langner et al. 2001; Guo et al. 2008; Reza and Singh 2010) and human activities (Klump et al. 2006). Leaching from solid waste leaching and wastewater discharge from the pharmaceutical, dyeing, and various chemical industry enterprises were the main contributor of arsenic pollution. In 2012, the total number of industrial enterprises with revenue over 20 million yuan per year was between 2446 and 10,444 for the eight cities, especially in Changzhou, Wuxi, Nantong, and Suzhou cities (Fig. 8a), which was corresponded to the distributions of moderate to extremely severe arsenic pollution of shallow groundwater in this study area (Fig. 7c).

Meanwhile, hydrogeochemical processes might cause the release of arsenic in the shallow groundwater.

Geothermal water with abundant arsenic species is mainly distributed in Nanjing, Zhenjiang, Wuxi, Suzhou and part of Nantong, which is consistent with the spatial distribution of the severe contamination in the western and southern parts of the study area. The pH values of the groundwater in the study area were weakly alkaline, which can promote the release of the arsenic (Smedley et al. 2002; Boyle et al. 1998) in the form of arsenate and arsenite (Stollenwerk et al. 2007). Higher pH value thus can reduce their attachment to sediments and thus increase arsenic concentration in groundwater (Park et al. 2006). Organic matter in groundwater can also facilitate the release of arsenic mainly through either reducing arsenate into arsenite (more mobile than arsenate in groundwater) or competing for adsorption sites (Bhattacharya et al. 1997). The stratum in the Lixiahe plain is the lagoon sediment, which is rich in organic matter. This is consistent with the result that severe arsenic contaminations in the shallow groundwater were observed in Yangzhong, Xinghua, Zhangjiagang, Nantong, Wuxi and Suzhou City (Fig. 7c).

Nitrogen

The distribution of total inorganic nitrogen pollution (including ammonium, nitrate, and nitrite) in the study area is

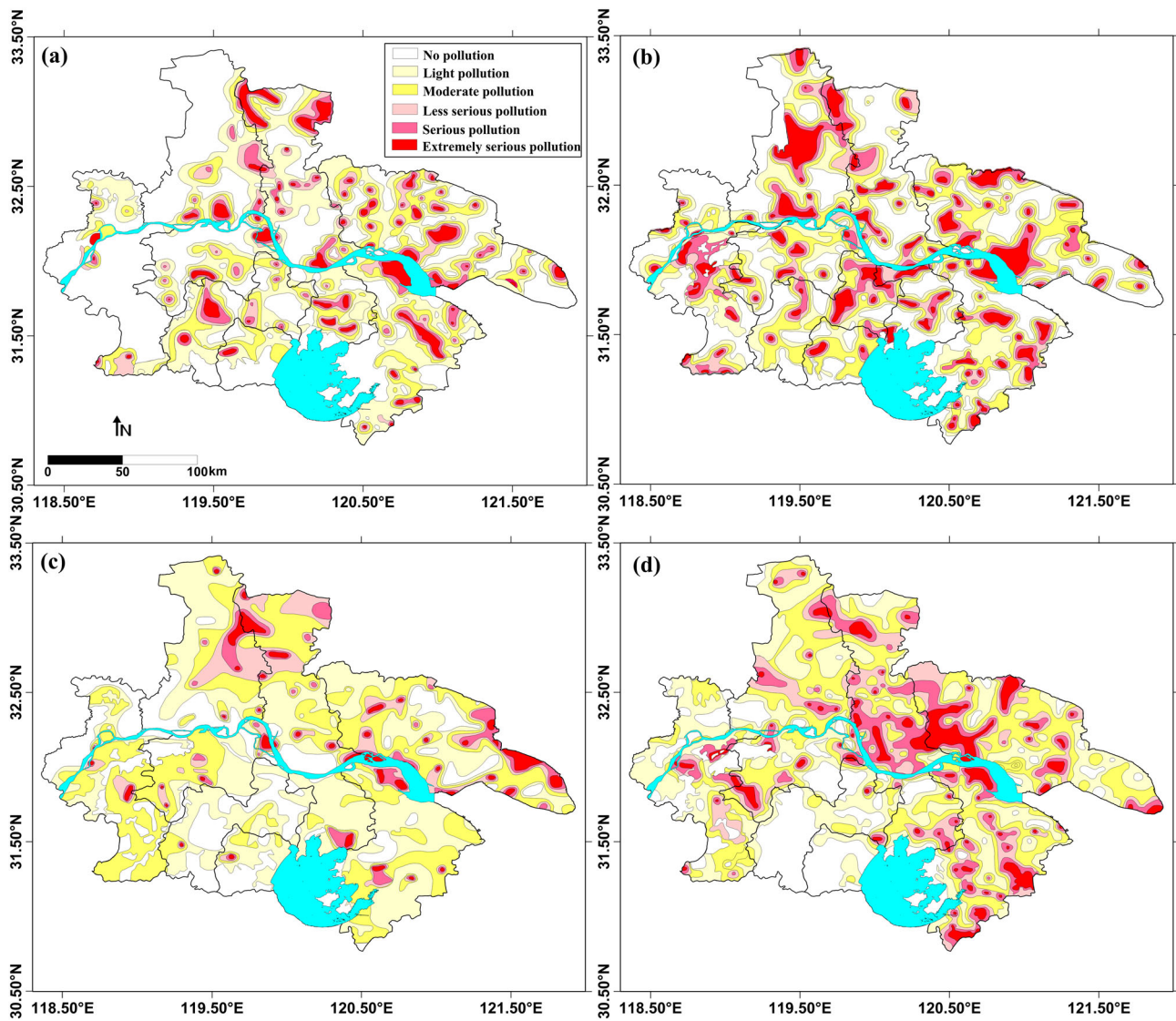


Fig. 7 Distributions of total ferrous iron (a), manganese (b), arsenic (c), and nitrogen (d) pollutions in the study area

illustrated in Fig. 7d. In general, the nitrogen pollution of the shallow groundwater was common with unpolluted (I) area of only 5773.11 km². The total area of the lightly (II) and medium (III) polluted parts was about 20,900 km². The severely contaminated parts were connected together (IV, V, and VI) with the area of about 13,500 km², accounting to the 33.50% of the study area. Based on the nitrogen pollution index evaluation (Fig. 7d), the Yangtze River delta plain unit was the most severely contaminated area, where the samples with the moderate to extremely severe pollution levels accounted for 62.63% of the total. The Taihu lake plain unit was moderately contaminated and the piedmont undulating plain unit was the lightest contaminated area.

The main sources of nitrogen pollution in the groundwater could be attributed to human activities, including usage of agricultural fertilizer, livestock industry, and

domestic and industrial sewage discharge (DeSimone and Howes 1998; Terblanche 1991). Figure 9 shows the land-use map of the study area in 2010, in which 6 first-level land-use types of the datasets were used, namely including cropland (including paddy field and dry land), woodland, grassland, water body, built-up land and unused land. The spatial distribution of the nitrogen pollution (Fig. 7d) was consistent with the degrees of urbanization, agricultural activities, and industry development (Fig. 9).

Industrial wastewater discharged from the light industries (i.e., dyeing mills, textile mills, and chemical plants) in the study area might also contribute to the nitrogen pollution. The distribution of the nitrogen contamination in the study area (Fig. 7d) was correlated with that of the light industries, especially in the Suzhou and Nantong city (Fig. 8a).

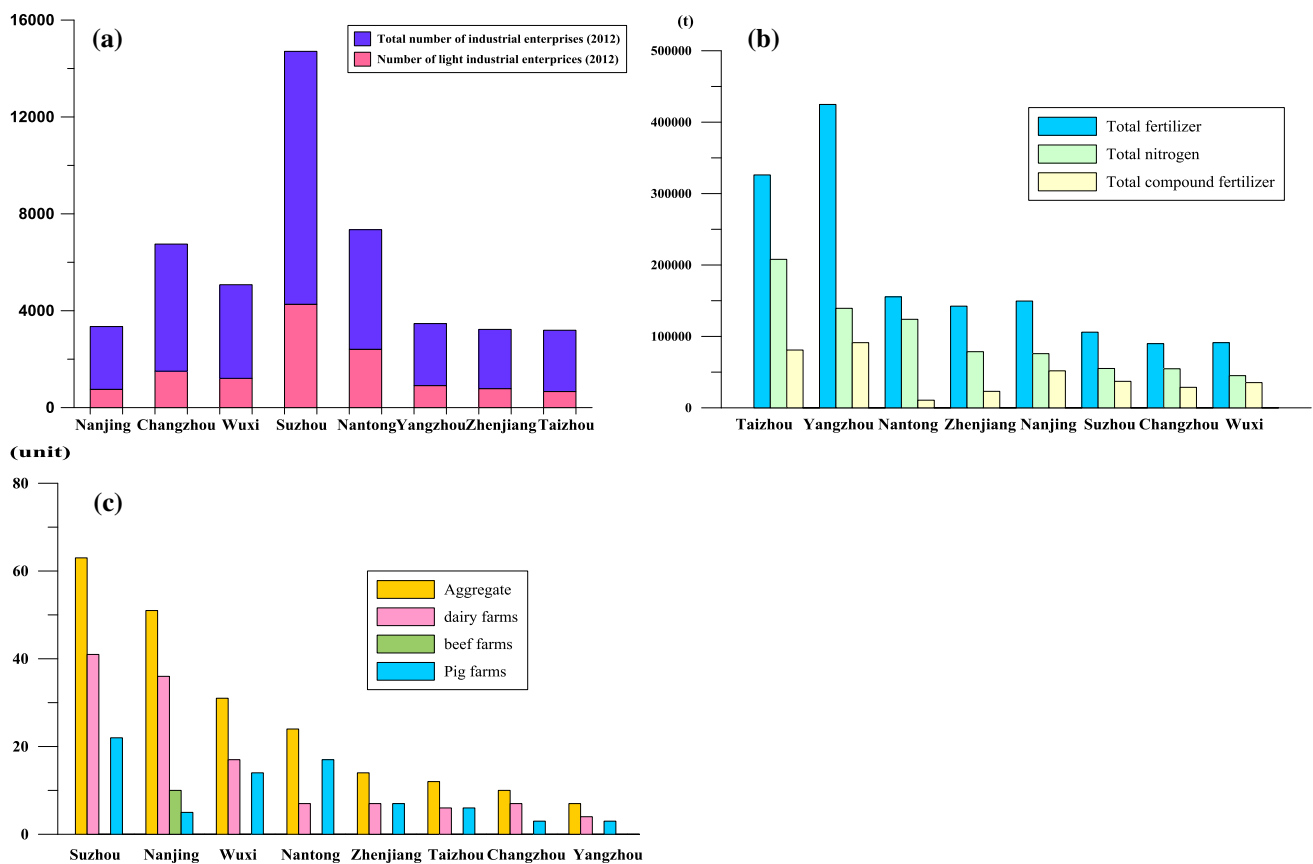


Fig. 8 Industrial and agricultural activities in the study area in 2012: **a** total number of major industrial enterprises (revenue >20 million yuan per year); **b** total amount of fertilizer usage (unit: tons); and **c** total number of large-scale farms

However, the usage of agricultural fertilizer has the strongest impact on nitrogen in groundwater (McLay et al. 2001; Ledoux et al. 2007). There are total 2051.2×10^3 ha of cropland area in the study area with three major crops (wheat, rice, and oilseed rape). In each of the cities in the study area, the total amount of total fertilizer, total nitrogen fertilizer, and total compound fertilizer usage in 2012 was between 89,864–424,813, 45,086–207,893, and 10,756–91,223 tons, respectively (Fig. 8b). The extremely severe nitrogen pollution was distributed in Taizhou, Nantong and Yangzhou, corresponding to the area that had the highest usages of nitrogen fertilizer (Fig. 8b). The average fertilizer usage is 733.79 kg/ha of arable land in 2006, far higher than the standard limited value of 225 kg/ha, which is defined in the developed countries to prevent fertilizer pollution to the groundwater. Because the utilization rate of nitrogen is generally lower than 50% (Dinnes et al. 2002), excessive fertilization may lead to the excessive reserve of the fertilizer. During precipitation and irrigation events, the dissolved fertilizer may infiltrate into the shallow groundwater.

Meanwhile, the study area also had many large-scale livestock farms (Fig. 8c), particularly in Suzhou and

Nanjing (>50 units), which might be another source of the nitrogen pollution in groundwater because fecal contamination from the livestock industry contains lots of nitrogen. The farms were mainly in Suzhou, Nanjing, Wuxi and Nantong area (Fig. 8c), corresponding to the sporadic severe nitrogen contamination of shallow groundwater in these cities (Fig. 7d).

Conclusions

This study firstly presents the qualitative and comprehensive evaluation of shallow groundwater quality in the CSJ area. A total of 968 groundwater samples were taken during 2006–2010 and analyzed for 72 groundwater quality indicators (see Table 2). The single factor pollution index method was used to determine the groundwater pollution levels of the study area. The results indicated that the shallow groundwater in CSJ was heavily polluted with both inorganic and organic contaminants, but the inorganic pollutions were more severe. The main inorganic contaminants that cause the severe and extremely severe pollution were manganese, iron, arsenic, and nitrogen. While natural

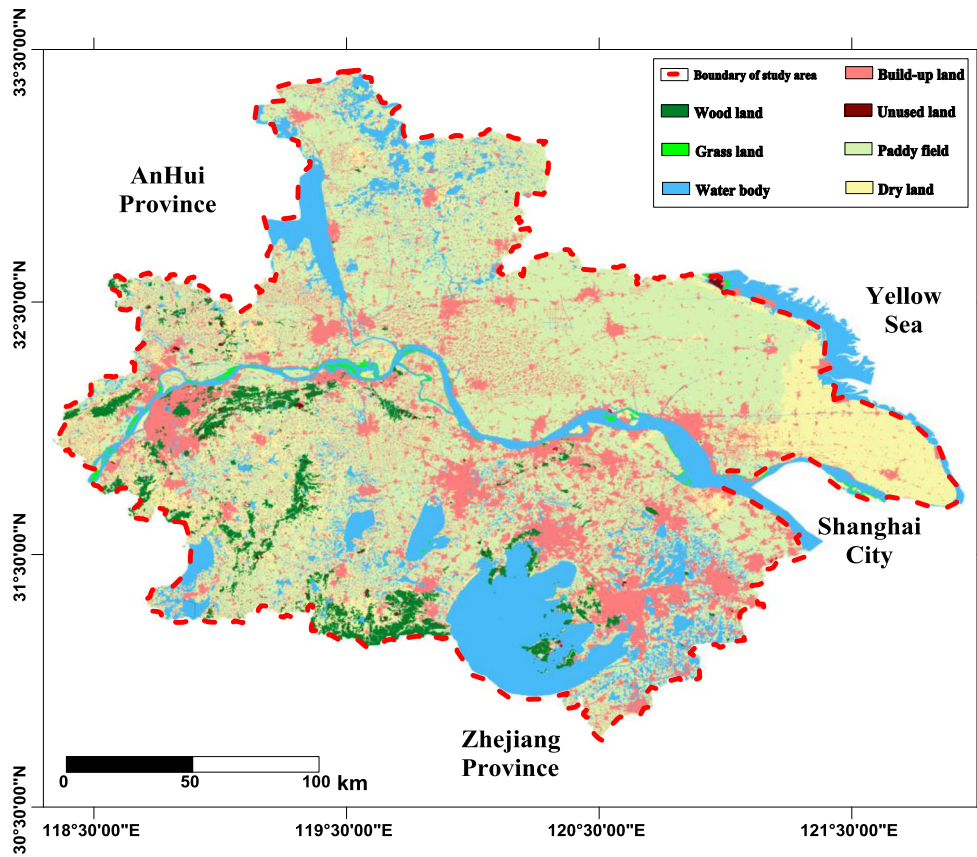


Fig. 9 Land-use map of the study area in 2010. The dataset was obtained from the National Science & Technology Infrastructure of China, National Earth System Science Data Sharing Infrastructure (<http://www.geodata.cn>)

processes contributed to the iron and manganese pollution in the study area; human activities such as industrial, agricultural, and domestic pollutions could be the main pollution source, especially for nitrogen. It is urgent to decrease the contamination sources to control the groundwater pollution. Pollution control strategies thus are needed in the study area to improve the shallow groundwater quality to satisfy the increasing water demands.

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