

## Research Article

# Natural Background Level and Contamination of Shallow Groundwater Salinity in Various Aquifers in a Coastal Urbanized Area, South China

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Assessing natural background levels (NBLs) of chemical components in groundwater is useful for the evaluation of groundwater contamination in urbanized areas. The present study assessed the NBL of total dissolved solids (TDS) in various groundwater units in the Pearl River Delta (PRD) where urbanization is a large scale and discussed factors controlling groundwater salinity contamination in the PRD. Results showed that the NBL of TDS in groundwater in the coastal-alluvial plain was more than 1.5 times that in other groundwater units because of the seawater intrusion in this groundwater unit. By contrast, interactions of water and soils/rocks were the main factors controlling the NBLs of TDS in other groundwater units. Groundwater salinity contamination in the PRD was positively correlated with the urbanization level. Wastewater from township-village enterprises and industrial wastewater were likely to be the main sources for groundwater salinity contamination in the PRD. Moreover, the wastewater leakage from sewer systems was one of the main pathways for groundwater salinity contamination in urbanized areas, because the proportion of groundwater salinity contamination in urbanized areas formed in 1988–1998 was more than 1.5 times that in urbanized areas formed in 1998–2006 regardless of groundwater units. Besides, sewage irrigation and leakage of landfill leachate were also important sources for groundwater salinity contamination in the PRD.

## 1. Introduction

Groundwater plays a crucial role in our livelihoods by making itself available for drinking. For example, more than half of drinking water is sourced from groundwater globally [1]. However, groundwater is often contaminated due to the large-scale urbanization and industrialization in urbanized areas [2, 3]; thereby, its quality in these areas is often deteriorated [4]. For instance, the Pearl River Delta (PRD) is a rapidly urbanized and industrialized area in China, and groundwater in this area is often contaminated by the salinity because of various anthropogenic sources such as high levels of salinity in wastewaters [3, 5]. In addition, several studies reported that geogenic sources are mainly

responsible for the origin of groundwater salinity in Quaternary aquifers in the PRD by using isotopic, hydrochemical, and microbial evidence [6–8]. However, the origins of groundwater salinity in fissured and karst aquifers as well as the spatial distribution of groundwater salinity contamination in the PRD are unclear. Thus, in order to improve the management and protection of groundwater resources, it is necessary to evaluate the status of groundwater salinity contamination and investigate the factors controlling groundwater salinity in the PRD.

To date, the evaluation of groundwater contamination is often based on the maximum admissible concentrations of chemical components which are harmless to human beings [9–11]. In this case, the impacts of natural factors on the

concentrations of chemical components in groundwater are neglected. However, high levels of chemical components in groundwater may derive from not only anthropogenic contamination but also natural processes [12–15]. Therefore, it is more reasonable to evaluate the groundwater contamination based on natural background levels (NBLs) of chemical components in groundwater. Here, NBLs are defined as concentrations present in groundwater as controlled by natural geogenic, biological, and chemical processes [16]. Preselection is the most common method for the assessment of NBL in groundwater [17]. For example, the method of oxidation capacity (OXC), as one of the preselection methods, was often used to assess the groundwater NBL in coastal areas [16, 18].

Therefore, the objectives of this study are to assess the NBL of salinity (total dissolved solids, TDS) in the PRD by using the OXC method with Grubbs' test, to depict the spatial distribution of groundwater salinity contamination with a Kriging method and to discuss factors controlling groundwater salinity contamination in the PRD. The conclusions would be helpful for improving the management and protection of groundwater resources in the PRD.

## 2. Study Area

The PRD occupies a total area of  $4.17 \times 10^4 \text{ km}^2$  in the southern part of Guangdong province, China. It includes nine cities, such as Guangzhou, Shenzhen, Foshan, Dongguan, Zhongshan, Zhuhai, Huizhou, Jiangmen, and Zhaoqing, in the district. It is bounded by hills in the east, west, and north and by the South China Sea in the south; thereby, its topography inclines from the east, west, and north to the south. Three major rivers, such as East River, West River, and North River, merge in the south and form the Pearl River, which finally discharges into the South China Sea. The PRD can be divided into three areas with different urbanization levels, such as urbanized area (UA), periurban area (PUA), and nonurbanized area (NUA) [13]. Besides, the UA in the PRD formed before 1988, from 1988 to 1998, and from 1998 to 2006 are denoted by UA<sup>1st</sup>, UA<sup>2nd</sup>, and UA<sup>3rd</sup>, respectively. UA<sup>1st</sup> is characterized by a lower proportion of factories in comparison with UA<sup>2nd</sup> and UA<sup>3rd</sup>, while UA<sup>3rd</sup> shows a relatively well-constructed sewer system in comparison with UA<sup>1st</sup> and UA<sup>2nd</sup> [5].

The PRD was formed as a result of the Tibetan Plateau uplift during the Tertiary and Quaternary Periods. It can be divided into four groundwater units (Figure 1). Unit A is related to a coastal-alluvial plain, which is located in the central and southern parts of the PRD and consists of two marine layers and two terrestrial layers [2]. The younger marine layer (M1) has an elevation of above  $-20 \text{ m}$ , and the older marine layer (M2) is located between  $-15$  and  $-40 \text{ m}$  (Figure 1). The younger terrestrial layer (T1) can be sandy fluvial deposits or clayey silt. The older terrestrial layer (T2) is dominated by sand and gravel [19]. Groundwater in this unit is recharged primarily by atmospheric precipitation, agricultural irrigation, and seawater intrusion [20, 21]. Unit B (alluvial-proluvial aquifer) is outside of unit A, which is related to the valley and interhill plains where marine layers

are absent but terrestrial layers are common. Unit C (fractured aquifer) is related to hilly areas where bedrocks are fractured. Unit D is related to karst aquifers and accounts for less than 10% of the total area [22].

## 3. Materials and Methods

**3.1. Sampling and Analytical Techniques.** Approximately 400 groundwater samples were collected from the PRD. Specifically, 124 samples, 134 samples, 132 samples, and 9 samples were collected from units A to D, respectively. All the samples were stored at  $4^\circ\text{C}$  until the laboratory procedures could be performed. Redox potential (Eh), pH, and dissolved oxygen (DO) were measured on site using a multiparameter instrument (WTW Multi 340i/SET, Germany). Metals (K, Na, Ca, and Mg) were measured by inductively coupled plasma mass spectrometry (Agilent 7500ce ICP-MS, Tokyo, Japan).  $\text{HCO}_3^-$  and TDS were measured using acid-base titration and gravimetric methods, respectively.  $\text{NH}_4^+$  and other anions ( $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$ , and  $\text{Cl}^-$ ) were analyzed by ion chromatography (Shimadzu LC-10ADvp, Japan). To ensure data quality, each sample was analyzed in triplicate, and sample batches were regularly interspersed with standards and blanks. The relative errors of inorganic parameters were  $\leq \pm 5\%$ .

**3.2. NBL Assessment.** Griffioen et al. reported that oxidation capacity is calculated as  $7[\text{SO}_4] + 5[\text{NO}_3]$  with the concentration of the species in  $[\text{mmol/L}]$  [16]. The oxidation capacity will be useless to identify groundwater with anthropogenic influence under the condition of a strongly reductive environment because groundwater  $\text{SO}_4$  and  $\text{NO}_3$  are at low levels. In this case, high levels of  $\text{NH}_4^+$  will be a useful indicator for the identification of contaminated groundwater, because high levels of  $\text{NH}_4^+$  in shallow groundwater in the PRD originated from human activities [23]. Therefore, a preselection method, consisting of two indicators such as the oxidation capacity and  $\text{NH}_4^+$  concentration, was used for the assessment of NBL in groundwater in this study. Specifically, groundwater samples with oxidation capacity  $> 3 \text{ meq/L}$  or  $\text{NH}_4\text{-N}$  concentrations  $> 0.5 \text{ mg/L}$  in original datasets were deleted, and the remaining datasets were denoted as PS datasets. Note that a higher value of  $3 \text{ meq/L}$  was selected as the threshold value for oxidation capacity in comparison with previous studies because geogenic sources are sometimes responsible for high levels of  $\text{SO}_4$  in groundwater in the PRD [24]. Then, TDS concentrations in PS datasets in various groundwater units were tested by Grubbs' test ( $\alpha = 0.01$ ) until normal distributions were obtained, and the outliers in various groundwater units were deleted (Supplementary material, Section 3.2) [25]. The remaining datasets were denoted as residual datasets. The maximum concentrations of TDS in residual datasets in various groundwater units were extracted as NBLs.

**3.3. Evaluation of Contamination Levels.** In this study, the contamination level of salinity in groundwater was quantified by the ratios of groundwater TDS concentrations to the

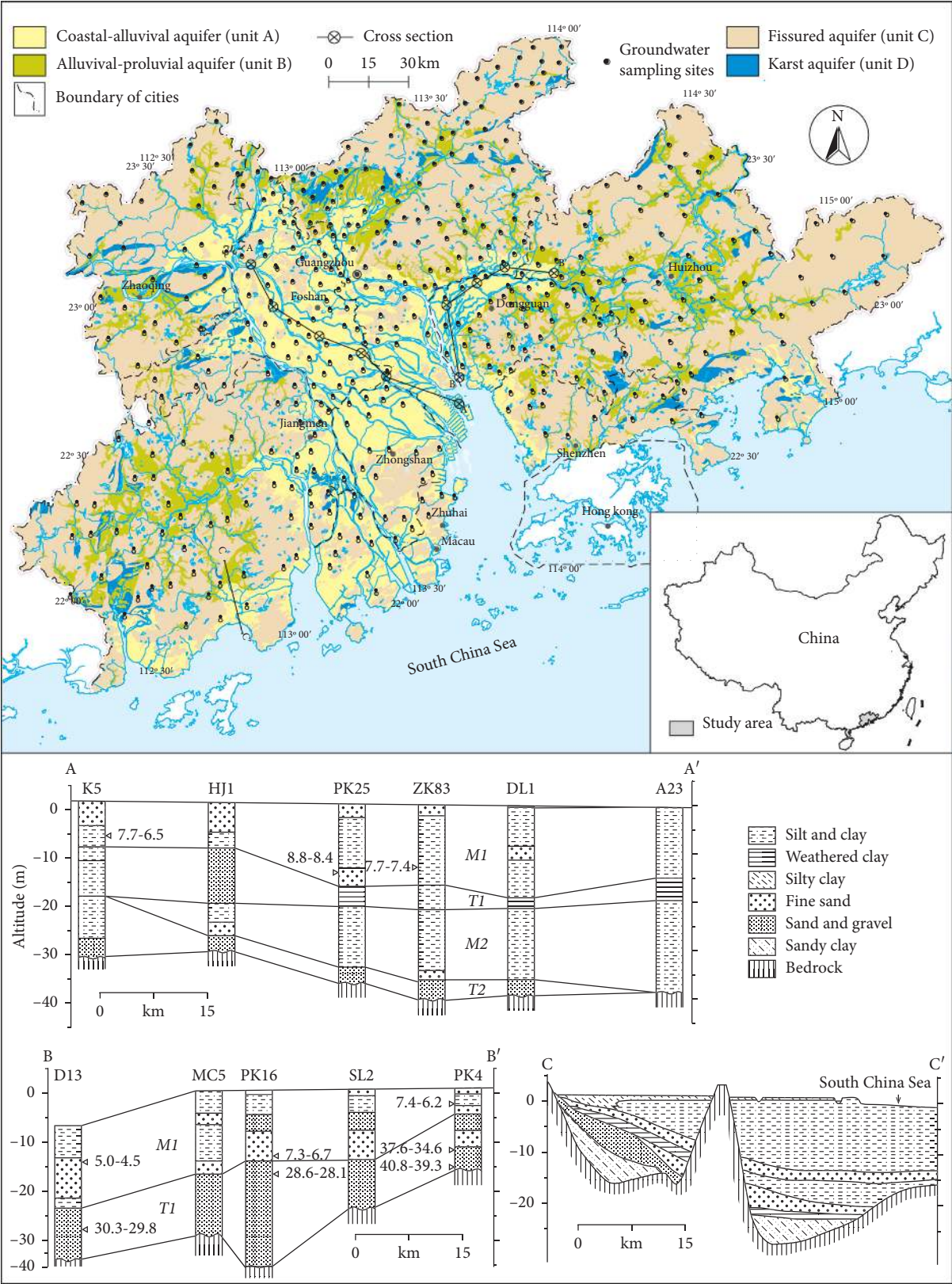


FIGURE 1: Groundwater units and sampling sites in the Pearl River Delta.

NBLs of TDS in various groundwater units. Thus, groundwater samples with different contamination levels are divided into four categories as follows: uncontaminated

(contamination level  $\leq 1$ ), low ( $1 < \text{contamination level} \leq 2$ ), moderate ( $2 < \text{contamination level} \leq 4$ ), and high (contamination level  $> 4$ ). The spatial distribution of groundwater



salinity contamination in the PRD is depicted by a geo-statistical analysis (MapGIS 10.2, China University of Geosciences, China). The universal kriging method and the estimation of empirical semivariogram models are used in MapGIS 10.2 software [26, 27]. The details are in the supplementary material (Section 3.2).

**3.4. Socioeconomic Data.** Socioeconomic data related to nine major cities in the PRD were compiled and used in this study. Relevant data were taken from the Statistical Yearbook of Guangdong Province and are compiled in Table S1 [28]; they include population density (PD, number of people/km<sup>2</sup>), gross domestic product (GDP, millions of Chinese yuan/km<sup>2</sup>), domestic sewage discharge (DSD, ton/km<sup>2</sup>), industrial wastewater discharge (IWD, ton/km<sup>2</sup>), ratio of urbanized areas (RUA, %), industrial enterprises above a specified size (IE, number/km<sup>2</sup>), township-village enterprises (TVEs, number/km<sup>2</sup>), agricultural output (AO, millions of Chinese yuan/km<sup>2</sup>), livestock output (LO, millions of Chinese yuan/km<sup>2</sup>), and livestock density (LD, number/km<sup>2</sup>).

**3.5. Principal Components Analysis (PCA).** The PCA is a useful technique for reducing a large number of variables to a small number of principal components (i.e., PCs) by linearly combining measurements made on the original variables [13]. In this study, the PCA was used to identify the relationships between the contamination level of groundwater salinity and the relevant socioeconomic data. In the PCA, only PCs with eigenvalues greater than one were retained for analyses, and the Varimax method was adopted. The terms “strong,” “moderate,” and “weak,” as applied to PC loadings, referred to absolute loading values of >0.75, 0.75–0.5, and 0.5–0.3, respectively [2].

## 4. Results

**4.1. NBL Assessment for Groundwater Salinity.** Using oxidation capacity >3 meq/L and NH<sub>4</sub>-N concentrations >0.5 mg/L to identify groundwater samples with anthropogenic influence in this study, nearly 60% of groundwater samples with anthropogenic influence in original datasets were removed out (Figure 2), and the remaining datasets were denoted as PS datasets. Then, TDS concentrations in PS datasets in various groundwater units were tested by Grubbs' test ( $\alpha = 0.01$ ), and outliers in PS datasets were eliminated (Figure 3). The remaining datasets were denoted as residual datasets. The maximum concentrations of TDS in residual datasets in various groundwater units are extracted as NBLs; thereby, NBLs of TDS in units A to D were 688 mg/L, 432 mg/L, 321 mg/L, and 99 mg/L, respectively.

### 4.2. Distribution of Groundwater Salinity Contamination in the PRD

**4.2.1. Salinity Contamination in Various Groundwater Units.** As shown in Table 1, 21.1% of groundwater samples in the PRD were contaminated by salinity. Among them, low to high contamination levels of groundwater samples

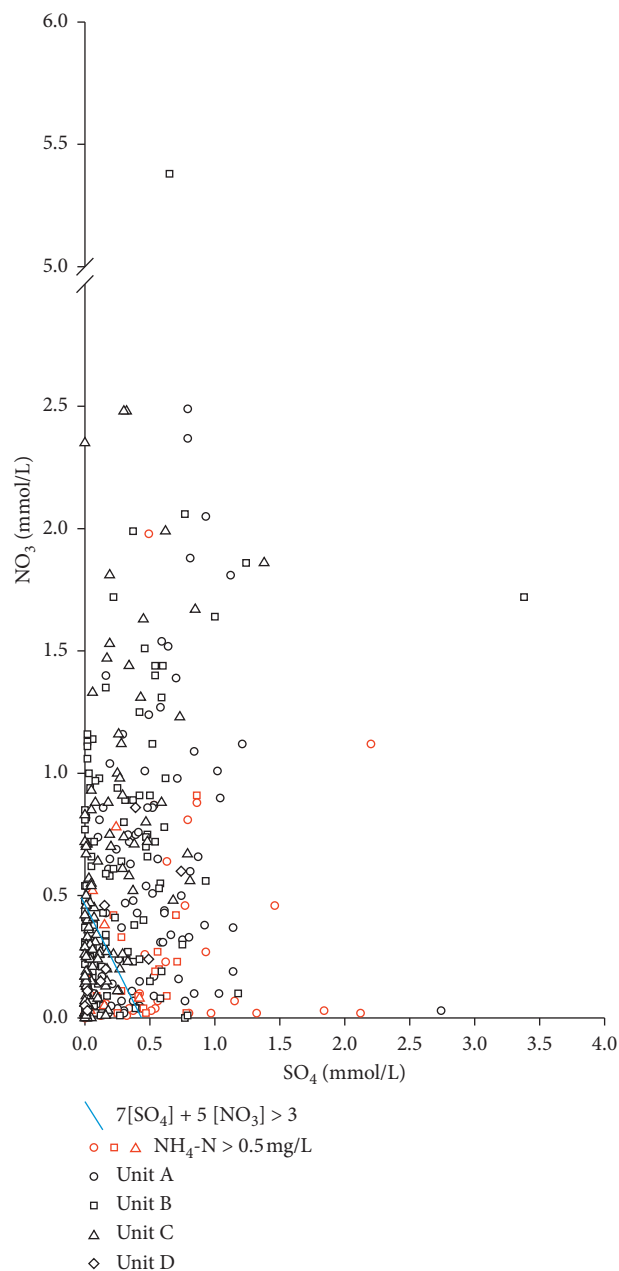


FIGURE 2: High levels of NH<sub>4</sub> as well as oxidation capacity in various groundwater units in the Pearl River Delta.

accounted for 16.5%, 3.8%, and 0.8%, respectively. Salinity contamination in various groundwater units in the PRD was significantly different. High levels of salinity contamination occurred in groundwater units A and B, but not in units C and D. By contrast, moderate levels of salinity contamination in groundwater unit D accounted for 44.4%, which was 10 times that in unit C and 20 times that in units A and B. Meanwhile, a low level of salinity contamination in groundwater unit A accounted for 26.6% and was more than 2 times that in other groundwater units. As far as the proportion of salinity contamination is concerned, groundwater unit D was the highest, which was 1.8 times that in unit A and 3.6 times that in units B and C.

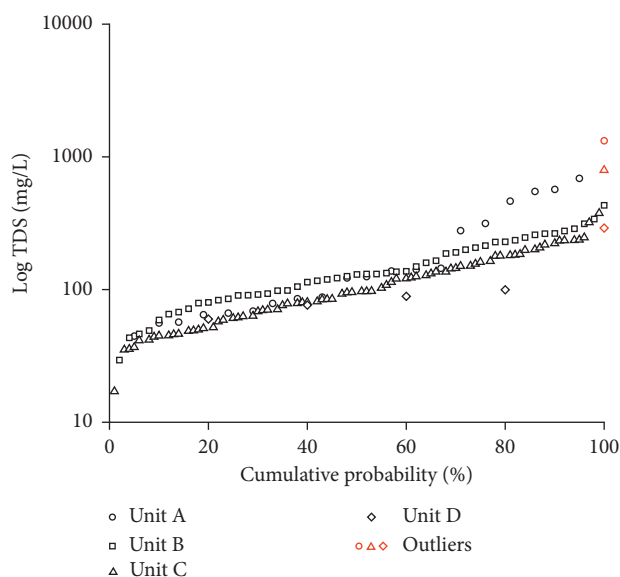


FIGURE 3: Outliers in PS datasets in various groundwater units tested by Grubbs' test.

TABLE 1: Groundwater salinity contamination in various groundwater units in the Pearl River Delta.

Contamination levels	PRD	Unit A	Unit B	Unit C	Unit D
Uncontaminated (%)	78.9	69.4	84.7	84.8	44.5
Low (%)	16.5	26.6	12.4	11.4	11.1
Moderate (%)	3.8	2.4	2.2	3.8	44.4
High (%)	0.8	1.6	0.7	0	0

**4.2.2. Groundwater Salinity Contamination in Areas with Different Urbanization Levels.** As seen in Table 2, the proportion of groundwater salinity contamination in UA was 29.7%, which was 1.2 times and 3.3 times that in PUA and NUA, respectively. This indicates that groundwater salinity contamination in the PRD was positively correlated with the urbanization level. Similarly, in groundwater unit A, the proportion of salinity contamination in UA was 1.8 times and 2.3 times that in PUA and NUA, respectively. Likewise, in groundwater unit B, the proportion of salinity contamination in UA was 1.2 times and 9.1 times that in PUA and NUA, respectively. By contrast, in groundwater unit C, the proportion of salinity contamination in PUA was the highest, which was 1.6 times and 3.9 times that in UA and NUA, respectively. This indicates that the correlation between the salinity contamination and the urbanization level in groundwater unit C was insignificant. Note that the salinity contamination in areas with different urbanization levels in groundwater unit D was not investigated because of too few samples. In addition, groundwater salinity contamination in UA formed in different periods in the PRD was also investigated. In groundwater unit A, proportions of salinity contamination in UA<sup>1st</sup> and UA<sup>2nd</sup> were close to each other and were approximately 1.5 times that in UA<sup>3rd</sup> (Table S2). By contrast, the proportion of salinity contamination in groundwater unit B in UA<sup>2nd</sup> was the highest and

was more than 5 times and 2 times that in UA<sup>1st</sup> and UA<sup>3rd</sup>, respectively (Table S2). Similarly, the proportion of salinity contamination in groundwater unit C in UA<sup>2nd</sup> was also much higher than that in UA<sup>1st</sup> and UA<sup>3rd</sup> (Table S2).

**4.2.3. Spatial Distribution of Groundwater Salinity Contamination in Various Cities.** As shown in Table 3, the high level of groundwater salinity contamination occurred in Dongguan and Guangzhou cities only, which accounted for less than 1% of the total area (Figure 4). The moderate level of groundwater salinity contamination occurred in six cities except for Shenzhen, Zhuhai, and Zhaoqing, which accounted for approximately 5% of the total area (Figure 4). Among them, the occurrence of a moderate level of groundwater salinity contamination was mainly distributed in Dongguan and Zhongshan cities (Table 3). By contrast, the low level of groundwater salinity contamination occurred in all of the nine cities, which accounted for approximately 16% of the total area (Figure 4). As far as the proportion of groundwater salinity contamination is concerned, Zhongshan was the highest, which was followed by the order of Dongguan, Foshan, Guangzhou, Jiangmen, Huizhou, Shenzhen, Zhuhai, and Zhaoqing, respectively (Table 3).

**4.3. Relationship between Contamination Levels of Groundwater Salinity and Socioeconomic Parameters.** In this study, the relationship between the proportions of groundwater salinity contamination and socioeconomic parameters in various cities in the PRD was investigated by the PCA. Note that differences in the socioeconomic parameters of the cities resulting from differences in size were eliminated by using the values of the socioeconomic parameters per square kilometer [3, 5]. PC2 shows strong positive loadings with the proportion of groundwater salinity contamination, TVE, and IWD (Table 4).

## 5. Discussions

**5.1. Natural Factors Controlling Salinity in Various Groundwater Units.** Generally, after the exclusion of the influence of anthropogenic activities, TDS in groundwater originates from the seawater intrusion, the soil-water interaction, and the rock-water interaction [7, 29]. In this study, NBL-TDS in groundwater unit A was much higher than that in other groundwater units (Figure 3). This probably ascribes to seawater intrusion, because the intrusion of seawater with extremely high levels of TDS often occurs in groundwater unit A [21]. NBL-TDS in groundwater unit B was lower than that in groundwater unit A but much higher than that in the other two groundwater units (Figure 3). This may be attributed to the more strong interactions of water and soils/rocks in groundwater unit B in comparison with that in groundwater units C and D because the groundwater flow rate in unit B is slower in comparison with that in units C and D [22]. This is also likely to be the main reason for the much higher NBL-TDS in groundwater unit C than that in groundwater unit D.

TABLE 2: Groundwater salinity contamination in areas with different urbanization levels in the Pearl River Delta.

Contamination levels	PRD			Unit A			Unit B			Unit C		
	UA	PUA	NUA	UA	PUA	NUA	UA	PUA	NUA	UA	PUA	NUA
Uncontaminated (%)	70.3	75	90.9	57.4	76.8	81.5	77.3	80.5	97.5	80	68.4	91.8
Low (%)	24.3	18.5	7.0	40.7	18.6	11.1	17	17.1	2.5	12.5	21.1	8.2
Moderate (%)	4.7	5.6	1.4	1.9	2.3	3.7	3.8	2.4	0	7.5	10.5	0
High (%)	0.7	0.9	0.7	0	2.3	3.7	1.9	0	0	0	0	0

TABLE 3: Groundwater salinity contamination in various cities in the Pearl River Delta.

Contamination levels	Guangzhou	Shenzhen	Foshan	Dongguan	Huizhou	Zhaoqing	Jiangmen	Zhuhai	Zhongshan
Uncontaminated (%)	79.3	86.7	75.8	65	84.4	96.2	80.9	92.3	52.7
Low (%)	16.5	13.3	21	20	12.5	3.8	15.9	7.7	36.8
Moderate (%)	2.1	0	3.2	12.5	3.1	0	3.2	0	10.5
High (%)	2.1	0	0	2.5	0	0	0	0	0

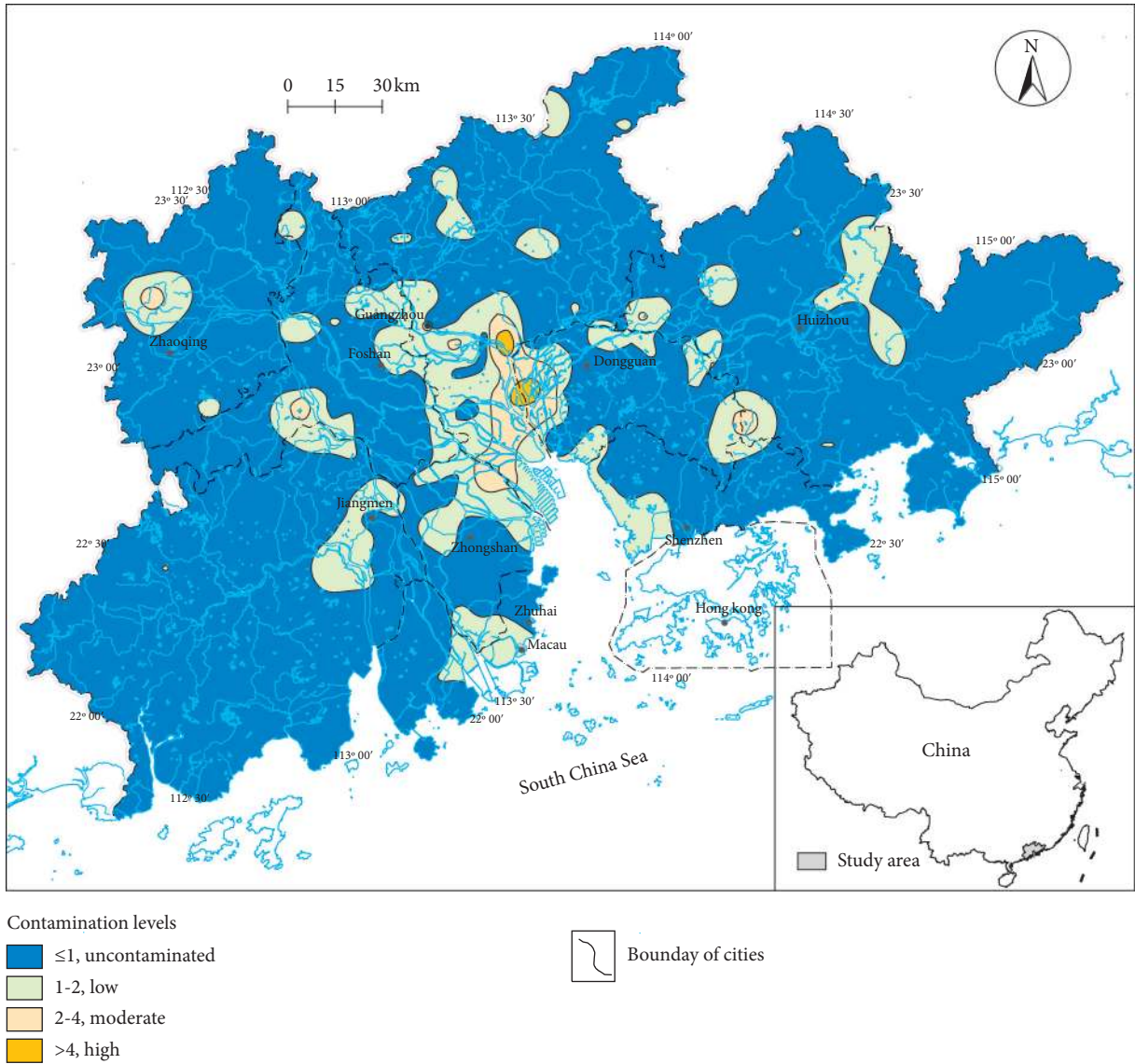




TABLE 4: Principal component (PC) loadings for proportions of groundwater salinity contamination and socioeconomic parameters in various cities in the Pearl River Delta

Items	PCs			
	PC1	PC2	PC3	PC4
GDP	<b>0.990</b>	−0.06	−0.051	−0.057
DSD	<b>0.975</b>	0.07	0.055	−0.188
PD	<b>0.966</b>	0.16	−0.136	−0.109
RUA	<b>0.940</b>	0.062	−0.243	0.019
IE	<b>0.833</b>	0.482	−0.159	0.13
PGSC <sup>a</sup>	0.095	<b>0.956</b>	0.104	0.136
TVE	−0.075	<b>0.943</b>	0.111	−0.276
IWD	0.386	<b>0.819</b>	−0.247	0.081
LO	−0.051	−0.113	<b>0.976</b>	−0.098
AO	−0.207	0.153	<b>0.949</b>	−0.008
LD	−0.142	−0.017	−0.083	<b>0.985</b>
Eigenvalue	4.7	2.8	2.1	1.1
Explained variance (%)	42.4	25.3	18.7	10.5
Cumulative % of variance	42.4	67.7	86.4	96.8

<sup>a</sup> PGSC: proportion of groundwater salinity contamination. Bold numbers represent maximum absolute PC loading of one parameter.

The proportions of salinity contamination in various groundwater units were significantly different (Table 1), indicating that groundwater salinity contamination in the PRD is affected by natural factors. A much higher proportion of salinity contamination in groundwater unit D than that in other groundwater units indicates that groundwater unit D has a higher risk of groundwater salinity contamination in comparison with other groundwater units in the PRD (Table 1) because the vadose zone in groundwater unit D is commonly characterized by more coarse-grained media in comparison with other groundwater units [22]. In addition, the proportion of salinity contamination in groundwater unit A was approximately 2 times that in groundwater units B and C (Table 1). This may not ascribe to natural factors but anthropogenic factors. In theory, the proportion of salinity contamination in groundwater unit A should be lower than that in groundwater units B and C if groundwater salinity contamination in various units is controlled by the property of media of vadose zone, because vadose zone in groundwater unit A is characterized by more fine-grained media in comparison with groundwater units B and C [22], which will result in the lower groundwater vulnerability of groundwater unit A in comparison with groundwater units B and C in the PRD. On the other hand, the ratio of UA in groundwater unit A was much higher than that in groundwater units B and C (Figures 1 and S1), and UA was commonly accompanied by more factories and industrial wastewater in comparison with other areas [5]. Thus, it can be concluded that a higher ratio of UA in groundwater unit A than that in groundwater units B and C is likely to be responsible for the higher proportion of salinity contamination in groundwater unit A in comparison with that in groundwater units B and C.

**5.2. Anthropogenic Factors Controlling Groundwater Salinity Contamination in the PRD.** A much higher proportion of groundwater salinity contamination in UA than that in PUA

and NUA indicates that human activities during urbanization should be the main factors (Table 2). Groundwater contamination such as nitrate and phosphorus contaminations in urbanized areas resulting from human activities was often identified by various socioeconomic parameters [3, 5]. Therefore, the relationship between the proportions of groundwater salinity contamination and socioeconomic parameters in various cities in the PRD was investigated by the PCA, to identify which human activities mainly result in groundwater salinity contamination in the PRD. As shown in Table 4, the proportion of groundwater salinity contamination is accompanied by TVE and IWD in the same PC. This indicates that wastewater from TVE and industrial wastewater is probably the main sources of groundwater salinity contamination in the PRD on a regional scale because industrial wastewater and wastewater from TVE with high levels of TDS were sometimes illegally discharged into rivers or ground surface without treatment [20]. For instance, the amounts of TVE in Zhongshan and Dongguan were more than 3 times that in other cities (Table S1), and the industrial wastewater discharges in Zhongshan and Dongguan were also significantly higher than those in other cities (except for Foshan) (Table S1). Correspondingly, the proportions of groundwater salinity contamination in both Zhongshan and Dongguan were more than 1.4 times that in other cities (Table 3). By contrast, the amount of industrial wastewater discharge in Zhaoqing was markedly lower than that in other cities (Table S1). Correspondingly, the proportion of groundwater salinity contamination in Zhaoqing was significantly lower than that in other cities (Table 3). This is also supported by the evidence that the proportions of groundwater salinity contamination in UA<sup>1st</sup> were much lower than those in UA<sup>2nd</sup> and UA<sup>3rd</sup> in some groundwater units such as B and C (Table S2) because UA<sup>1st</sup> has a lower proportion of factories as well as a lower discharge of industrial wastewater in comparison with UA<sup>2nd</sup> and UA<sup>3rd</sup> [5]. In addition, the proportions of groundwater salinity contamination in various units in UA<sup>2nd</sup> were more than 1.5 times those in UA<sup>3rd</sup> (Table S2), indicating that the wastewater leakage from sewer systems may be one of the main pathways for groundwater salinity contamination in UA in the PRD because UA<sup>3rd</sup> shows a relatively well-constructed sewer system in comparison with UA<sup>2nd</sup> [5].

The proportions of groundwater salinity contamination in PUA were significantly higher than those in NUA in various groundwater units (Table 2). This is probably attributed to the infiltration of industrial wastewater and domestic sewage. On one hand, illegal discharge of industrial wastewater with high levels of TDS from factories into the nearby ground surface sometimes occurred in PUA but none in NUA because some factories such as TVE were distributed in PUA but none in NUA [12, 13]. On the other hand, domestic sewage with high levels of TDS was also often discharged into the nearby ground surface in PUA in the PRD because a large number of people lived in the PUA where the sewer system was sometimes missing [2, 5]. Therefore, the infiltration of industrial wastewater and domestic sewage is likely to be the main source of groundwater salinity contamination in PUA in the PRD.

Though the proportion of groundwater salinity contamination in NUA was much lower than that in UA and PUA, it was still considerable, especially in groundwater unit A (Table 2). As shown in Figures 4 and S1, NUA contaminated by groundwater salinity was mainly located at a river network area adjacent to the Pearl River Estuary. This indicates that sewage irrigation may be mainly responsible for groundwater salinity contamination in NUA in the PRD. On one hand, most rivers in this river network area were contaminated by industrial and domestic sewage due to urbanization and industrialization, thereby resulting in high levels of TDS occurring in these polluted river waters [30]. On the other hand, agricultural lands within NUA near rivers were often irrigated by river waters [4, 20]. Besides, a few groundwater samples characterized by relatively high levels of TDS within NUA were near landfills such as Datianshan landfill and Hongmei town landfill [5, 31], and these groundwater samples in the west boundary of Dongguan city and southeast of Guangzhou city showed low to high levels of salinity contamination (Figure 4). This indicates that the leakage of landfill leachate should also be an important source for groundwater salinity contamination in NUA in the PRD because these landfills often lacked antiseepage measure and landfill leachate showed extremely high levels of TDS [13, 31].

## 6. Conclusions

Results showed that NBLs of TDS in groundwater units A to D were 688 mg/L, 432 mg/L, 321 mg/L, and 99 mg/L, respectively. The seawater intrusion was likely to be responsible for the much higher NBL-TDS in groundwater unit A than that in other groundwater units. By contrast, higher NBL-TDS in groundwater unit B than that in groundwater units C and D was probably attributed to the more strong interactions of water and soils/rocks in groundwater unit B in comparison with that in groundwater units C and D. This was also likely to be the main reason for the much higher NBL-TDS in groundwater unit C than that in groundwater unit D.

The proportion of salinity contamination in groundwater unit D was much higher than that in other groundwater units. This may be attributed to the more coarse-grained media of vadose zone in groundwater unit D in comparison with other groundwater units. By contrast, a higher ratio of UA in groundwater unit A than that in groundwater units B and C was probably responsible for the higher proportion of salinity contamination in groundwater unit A in comparison with that in groundwater units B and C.

Groundwater salinity contamination in the PRD was positively correlated with the urbanization level. Furthermore, proportions of salinity contamination in UA<sup>2nd</sup> were much higher than that in UA<sup>3rd</sup> regardless of groundwater units. By contrast, the proportion of salinity contamination in UA<sup>3rd</sup> was also much higher than that in UA<sup>1st</sup> in both groundwater units B and C but contrary in groundwater unit A. Wastewater from TVE and industrial wastewater were likely to be the main sources for groundwater salinity contamination in the PRD, and the wastewater leakage from

sewer systems was one of the main pathways for groundwater salinity contamination in UA. Besides, sewage irrigation and leakage of landfill leachate were also important sources for groundwater salinity contamination in the PRD.

## Data Availability

The data used to support the findings of this study are included within the article.

## Conflicts of Interest

The authors declare that there are no conflicts of interest regarding the publication of this paper.

## Acknowledgments

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## Supplementary Materials

Table S1: socioeconomic data for the nine major cities of the PRD in 2006. Table S2: groundwater salinity contamination in UA formed in different periods in the Pearl River Delta. Figure S1: the expansion of urbanization in the PRD. (*Supplementary Materials*)

## References

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