

Petroleum pollution and evolution of water quality in the Malian River Basin of the Longdong Loess Plateau, Northwestern China

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Received: 10 October 2010 / Accepted: 3 October 2011 / Published online: 15 October 2011
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Abstract The possible sources of water pollution and the evolution of water quality in the Malian River Basin of China's Longdong Loess Plateau were studied based on chemical analysis. Concentrations of major chemicals in the river were related to the distance downstream from the source of the river and the surface water in the upstream had good quality, while the river from Southern Huanxian County to Qingcheng County was seriously polluted. Groundwater quality was generally good in the phreatic aquifer beneath the loess plateau, with bicarbonate and calcium dominant, but salinity was high and petroleum pollution was present in many parts of the study area. Both surface water and groundwater quality have declined rapidly during the past 20 years, leading to much of the water unsafe to drink and unsuitable for use in irrigation. The substances released into the water included industrial wastes, domestic sewage, organic and inorganic fertilizers, and pesticides, as well as a range of other substances that are harmful to humans. However, petroleum contamination caused by the oil industry in the Longdong oilfield is the largest source of pollution that infiltrates surface water and groundwater, leading to deterioration of water quality. It is recommended that oilfield management must be improved

and the petroleum products and wastes must be disposed carefully to reduce the risk of further spills and pollution.

Keywords Water quality · Petroleum pollution · Groundwater · Loess plateau

Introduction

During the latter part of the twentieth century, a combination of climate change, population increase, political independence, new technologies, and development of optimism after World War II has accelerated the increase in water withdrawals around the world (IPCC 2007). In addition to climate-induced changes and variations in the global context, river systems respond to human activities such as the construction of dams, levees, and bridges; the diversion of bed materials or flows; sand mining; water withdrawal to meet urban, industrial, and agricultural needs; and changing land use (Walling and Fang 2003; Ye et al. 2003; Choi et al. 2005). These activities can have wide variety of physical, ecological, and environmental effects, such as streambed mobilization, scouring, degradation, and aggradation, as well as changes in the hydrologic regime (Magilligan and Nislow 2005; Magilligan et al. 2008). Many of the world's largest rivers, including the Nile, Colorado, Yellow, Indus, and Ganges, no longer reach the sea for part of the year (Conway and Hulme 1993; Shiklomanov 1997; Archer 2003; Christensen et al. 2004; Liu and Jun 2004; Conway 2005). Water scarcities will become more critical in the future as population growth in arid regions surpasses that in wetter regions. In addition, as urbanization continues, water pollution has become increasingly evident, leading to serious ecological and environmental problems (Chen 2002; Ma et al. 2009).

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Industrial production without adequate regard for environmental impacts has increased both water and air pollution, leading to soil degradation and large-scale global impacts such as acid rain, global warming, and ozone depletion (Seth 2003).

The widespread use of petroleum products around the world has also led to contamination of soil and groundwater at many sites (Culbertson et al. 2008). The subsurface flow and transport of petroleum-derived contaminants, commonly referred to as non-aqueous-phase liquids, is particularly important. Contamination problems arise from disposal dumps, leaking storage tanks, and accidental spills of hydrocarbons used by the petroleum industry (Garg and Rixey 1999; Tam and Byer 2002). Oilfield wastewater is produced by oil rigs as a byproduct of the oil extraction process. This wastewater is enriched in the total *n*-alkane component, but also contains high concentrations of minerals, radioactive substances, benzenes, phenols, and polycyclic aromatic hydrocarbons (Cormack et al. 1998; Toril 2001; Nie et al. 2010). Such wastewater can severely pollute rivers, groundwater, soils, and even the air (Chen et al. 2003; Jerez et al. 2002; Ou et al. 2004; Fakhru'l-Razi et al. 2009). Hydrocarbon contamination of soils, sediments, and groundwater is a global environmental problem. Technological advances for detecting and monitoring hydrocarbon contamination are required to let managers quickly assess and select remediation strategies (Rijal et al. 2010). Especially in northwestern China, serious conflicts over water have arisen; the petroleum industry competes with agriculture to obtain water, agriculture competes for water with the ecological environment, and upstream areas compete for water with downstream areas (Ma et al. 2005). The resulting pressure is most severe in arid and semi-arid zones, where high demand is combined with limited resources.

The Malian River, the subject of the present study, is a major river system in the Longdong Loess Plateau, which surrounds Qingyang City in Gansu Province. The river is the main source of industrial, agricultural, and domestic water supplies. As the region's economy has developed rapidly, causing extensive land use and land cover change, the river's water quality has seriously degraded, particularly as a result of the oil industry's operations in the Longdong oilfield. Such environmental degradation not only impedes sustainable development, but also endangers human health. However, accurate assessment of the kind and extent of water pollution is difficult due to the diverse processes and complex phenomena characterizing these environments. In this context, the main objectives were: (1) to assess the surface water and groundwater quality; (2) to identify the major factors influencing water quality in the region; and (3) to explore the possible pollutant sources, and estimate the potential contributions of petroleum pollution to water quality variables.

Study area and hydrogeology

General settings

The study area is located in eastern Gansu Province, between 106°45' and 108°45'E and between 35°10' and 37°20'N (Fig. 1). It borders Pingliang City of Gansu Province in the south, and shares boundaries with Guyuan City of Ningxia Province in the west and Shaanxi Province in the east and north. The area is in the lower middle reaches of the Yellow River, in the gully region of China's loess plateau (the Longdong Loess Plateau). Elevation ranges from 885 to 2,082 m above sea level. The region experiences severe soil erosion and gully formation, leading to the development of complex landforms.

The average temperature for the region around Xifeng ranges from 7 to 10°C. The annual precipitation averages 505 mm, and decreases moving from ~ 620 mm in the south to ~ 350 mm in the north. Rain falls mostly from July to September, accounting for 58.5% of the annual average. The five largest rivers in the Longdong Loess Plateau are the Malian, Puhe, Honghe, Silanghe, and Hulu rivers. The Malian River originates in the Mahuang Mountains of Ningxia Province, and flows into Qingyang from the north and enters the Jinghe River in Ningxian County. The Malian River is 366 km long and covers an area of 16,900 km². The average annual runoff is 4.47×10^8 m³ with a mean flow of 14.17 m³ s⁻¹.

Geology and hydrogeology

The Longdong Loess Plateau is on the western edge of the Ordos Platform, which is part of the stable Northern China Platform and was a huge inland freshwater lacustrine basin that formed in the Late Triassic and filled with Mesozoic fluvial lacustrine clastics (Yang 2002). This intracontinental sedimentary basin covers 37×10^4 km², and contains abundant Mesozoic crude oil reserves estimated at 10×10^8 m³. From the middle Triassic to the early Cretaceous, large-scale depression basins developed, forming the Tongchuan–Qingyang Depression. During the late Triassic, the depression rose sharply due to uplift, and the large Triassic basins fragmented into large Jurassic depression basins that form the current structural profile of the Malian Basin. After the early Cretaceous, the basins rose until the Pliocene, subsided over large areas, and then received deposits of Tertiary red clay. The Quaternary deposits formed on the surface of the Pliocene series. The fluvio-lacustrine Sanmen Formation developed within the ancient hydrologic network during the early Pleistocene and the Wucheng Loess developed in the northern Qingyang Depression, ranging from 10 to 60 m in

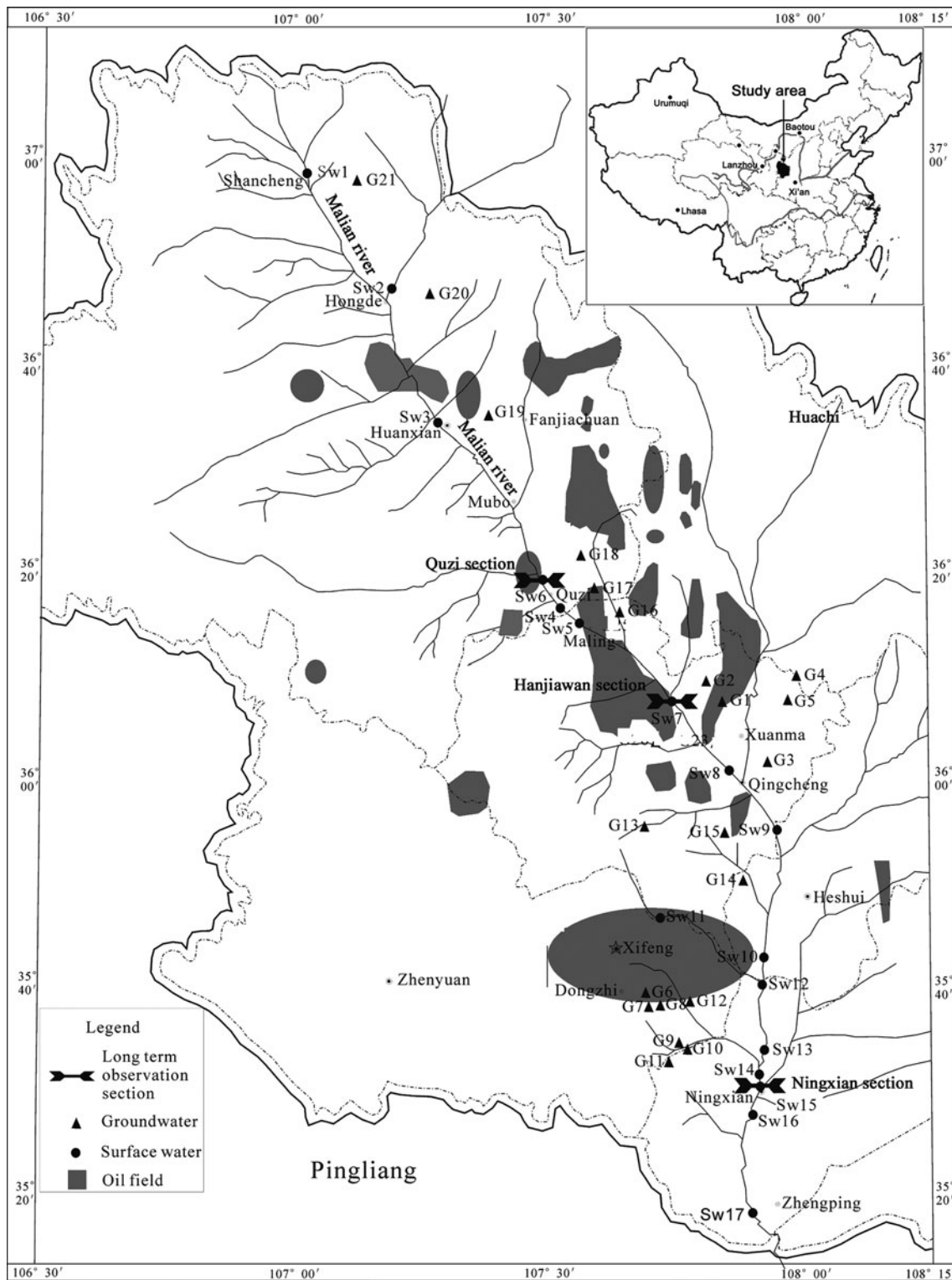


Fig. 1 Hydrogeological map of the Malian River Basin in China's Loess Plateau, and locations of the sampling sites used for measurements of surface water and groundwater characteristics

thickness. The Lishi Loess of the middle Pleistocene and the Malan Loess of the early Pleistocene were influenced by the imbalance uplifts and formed the thickest Loess plateau in the south of Qingyang, reaching a thickness of

100–180 m. These deposits are responsible for the current landforms and hydrological systems.

The highest residual terrace of the Lishi Loess formed the western bank of the Malian River and the Malan Loess

Terrace is distributed along the current watershed, with a thickness of 10–20 m. Holocene alluvial and proluvial sandy loams are distributed along the rivers. The sandy loams on terraces I and II have a thickness of 3–8 m, and an underlying layer of sandy gravel 3–5 m thick in the river bed which forms a good aquifer. On ridge and hilly areas of the loess plateau, the phreatic aquifer occurs primarily in the loess of the Pleistocene. As a result of ravine development and uplift, most of the aquifer structure was damaged and the aquifer is thin and discontinuous. At the center of the loess tableland (*yuan*, a kind of loess landform with flat and complete stratigraphies), which covers more than 100 km², the aquifer is 40–80 m thick, with a 20–40 m depth of water level.

The late Cretaceous system in northern Huanxian County is conglomerate and sandstone of 800 m depth, with an aquifer of 400–700 m thickness and a general specific water flow of 2,000 m³ day. The Huanhe Formation of the late Cretaceous system is distributed throughout the Longdong Loess Plateau, which is a part of an artesian basin far from the source and discharge areas. The aquifer is a sandy shale, mudstone and siltstone with sandstone interbedded, which is 400–600 m thick, and the specific water flow is generally 200–500 m³ day, with a maximum of 700 m³ day. The aquifer of the Luohandong Formation is coarse to fine sandstone, and is mainly distributed west of the Malian River, with a thickness of 100–250 m.

Methods

Field work took place during October 2009 and the sampling sites used to provide the data reported in this paper appear in Fig. 1. A total of 17 surface water samples and 21 groundwater samples were obtained for chemical and pollutant analysis. Each sample was separated into three aliquots. Two of these were filtered through pre-cleaned 0.45- μ m acetate filters using a pre-cleaned Nalgene filter apparatus, and the filtrate of one of these two aliquots was acidified with suprapure HNO₃ (1% v/v) and stored in pre-cleaned polyethylene bottles (60 mL) for analysis of major cations. The other filtered portion was stored un-acidified in polyethylene bottles for the anion determinations. These samples were stored at 4°C until they could be analyzed. The third aliquot (3,000 mL) was stored in two sterile plastic bacteriological bottles. These samples were immediately placed in the dark, cooled to 4°C in an insulated container, and transported to the laboratory within 10 h; they were then processed within 2 h following standard methods (SEPA 2002; CREH 1999).

Total alkalinity (as HCO₃⁻) and the electrical conductivity (EC), total dissolved solids (TDS), pH, temperature, and dissolved oxygen content (DO) were measured in the

field. Alkalinity was measured using a Hath field titration kit (titration with 0.1 M HCl). The EC and TDS ($\pm 5\%$, standardized to 25°C), pH (± 0.05), temperature, and DO were measured using portable field kits with different electrodes (Mettler-Toledo, Columbus, OH) that had been previously calibrated using standard buffers. For field measurements of groundwater, an in-line flow cell was utilized to prevent atmospheric contamination and improve measurement stability. Major anions (Cl⁻, SO₄²⁻, NO₃⁻ and F⁻) and major cations (Na⁺, K⁺, Ca²⁺, Mg²⁺) for surface water and groundwater samples were undertaken by ion chromatography (Dionex ICS-2500) at the Key Laboratory of Western China's Environmental Systems (MOE), Lanzhou University. An AS4A analytical column with carbonate/bicarbonate was used for the anions (uncertainty 10% with a coverage factor $k = 2$). Calibrations for cation analyses were performed using appropriately diluted laboratory standards (uncertainty 10–15%, $k = 2$). The precision and accuracy of analyses were within 5% (evaluated through repeated analyses of standards and samples) (Ma et al. 2010). The pollutants analyses including ammonium (NH₄⁺-N), chemical oxygen demand (COD), biological oxygen demand (BOD), permanganate index (PI), Cr⁶⁺, phenol, petroleum, total nitrogens (TN) and total phosphorus (TP) were undertaken at the Key Laboratory of Qingyang Environmental Observation Station, according to the standard analytical method for water quality provided by the China State Environmental Protection Administration (GB3838-2002) (SEPA 2002), which is similar to the international standard method of analysis of water and wastewater (APHA 1992).

To investigate water pollution, all sources of industrial pollution, urban domestic sewage, and agricultural pollution were surveyed from September to October 2009. More than 30 enterprises and 20 oil wells were surveyed to collect information about the water pollution sources, the main types of pollutants and composition of the wastes, waste discharge locations and volumes, waste control methods, and existing problems. The survey of urban domestic pollution included the total population, the floating population of seasonal migrant workers, water consumption per capita, and the location and scale of sewage treatment plants and sewer outlets. Evaluation of agricultural sources of pollution focused on pesticide and fertilizer use. Data on domestic and agricultural pollution from 1991 to 2008 were provided by the Qingyang Environmental Protection Office (QEPO 2009). The long-term water quality monitoring data provided by QEPO (2009) were used for the previously mentioned parameters in surface water and groundwater.

In recent years, water quality assessment has been performed using the water quality standards for surface water. A synthetic pollution index was developed to integrate the

Table 1 The analytical data and statistical results for the chemicals and pollutants measured in surface water samples from the Malian River Basin by Qingyang Environmental Observation Station (2008)

Sample no.	pH	DO	Cl ⁻ (mg L ⁻¹)	NO ₃ ⁻ (mg L ⁻¹)	SO ₄ ²⁻ (mg L ⁻¹)	COD (mg L ⁻¹)	BOD ₅ (mg L ⁻¹)	PI (mg L ⁻¹)	NH ₄ -N (mg L ⁻¹)	Cr ⁶⁺ (mg L ⁻¹)	Phenol (mg L ⁻¹)	Petroleum (mg L ⁻¹)	TP (mg L ⁻¹)	Coliform (cfu L ⁻¹)	P _i
SW1	7.98	7.97	1,532.47	11.45	1,211.32	76.40	2.00	5.52	0.83	0.108	0.006	0.035	0.030	20	13.30
SW2	7.82	7.47	1,059.42	11.06	1,003.77	52.30	2.00	4.88	2.25	0.300	0.002	0.053	0.028	70	14.29
SW3	7.91	7.27	1,037.25	11.43	1,024.53	84.20	2.00	4.34	0.35	0.162	0.002	0.050	0.106	50	11.84
SW4	7.56	8.23	739.13	10.53	354.72	59.60	2.00	3.63	1.69	0.132	0.002	0.035	0.027	90	6.74
SW5	7.82	8.17	699.71	10.26	575.09	34.20	2.00	4.24	1.75	0.136	0.002	0.045	0.036	60	6.31
SW6	8.48	2.40	759.60	60.16	769.55	51.80	2.85	2.20	0.87	0.126	0.002	0.032	0.251	110	13.02
SW7	8.64	2.80	832.62	78.03	816.81	53.80	4.42	6.25	1.36	0.082	0.002	0.047	0.298	95	15.17
SW8	7.38	8.37	202.03	1.57	332.45	8.44	2.00	2.96	1.86	0.044	0.002	0.040	0.025	120	1.19
SW9	7.58	7.63	512.47	5.06	89.43	8.04	2.09	4.27	1.66	0.044	0.002	0.098	0.025	140	2.67
SW10	7.79	8.39	149.30	5.65	294.34	26.90	2.41	6.99	2.04	0.048	0.002	0.042	0.120	20	1.72
SW11	7.88	8.13	263.13	2.24	98.11	115.00	2.00	41.20	1.03	0.151	0.006	0.061	0.295	120	13.62
SW12	7.80	8.80	85.74	1.10	43.40	50.70	4.34	8.40	3.35	0.050	0.002	0.058	0.111	40	4.53
SW13	7.65	8.75	77.86	10.62	62.26	34.20	5.46	8.98	3.66	0.067	0.002	0.010	0.370	40	5.48
SW14	7.82	8.96	448.41	9.14	426.42	35.00	2.35	6.16	2.08	0.108	0.002	0.030	0.108	70	4.52
SW15	8.41	1.60	397.98	27.54	472.77	51.10	3.97	4.78	2.27	0.065	0.002	0.010	0.026	70	7.04
SW16	7.65	7.96	453.33	2.12	445.28	120.60	2.41	2.74	0.91	0.111	0.002	0.035	0.025	80	7.84
SW17	7.39	8.21	404.06	1.12	261.32	76.40	2.34	14.80	0.75	0.029	0.002	0.028	0.025	50	4.95
Mean	7.86	7.12	567.91	15.24	487.15	55.22	2.74	7.78	1.69	0.10	0.00	0.04	0.11	73	7.90
Minimum	7.38	1.60	77.86	1.10	43.40	8.04	2.00	2.20	0.35	0.03	0.00	0.01	0.03	20	1.19
Maximum	8.64	8.96	1,532.47	78.03	1,211.32	120.60	5.46	41.20	3.66	0.30	0.01	0.10	0.37	140	15.17
National standard values															
Drinking water	-	5.00	250.00	10.00	250.00	20.00	4.00	6.00	1.00	0.05	0.01	0.05	0.20	10,000	-
Irrigation water	-	2.00	-	-	-	40.00	10.00	15.00	2.00	0.10	0.10	1.00	0.40	40,000	-

Parameter definitions: DO dissolved oxygen content, COD chemical oxygen demand, BOD biological oxygen demand, PI permanganate index, phenol total volatile phenols, petroleum all petroleum compounds, TP total phosphorus, P_i the synthetic pollutant index defined in "Methods" section of the paper

individual impact of the various pollutants on water quality. This approach is widely used because it provides a simpler overview parameter (Ma et al. 2009). The index was calculated using the following equation:

$$P_i = \sum_{j=1}^n (C_j - S_j) / S_j$$

where P_i represents the synthetic pollution index for sample i , n represents the number of parameters, and $C_j - S_j = 0$ if $C_j < S_j$. C_j is the analytical concentration of the j th parameter, and S_j is the standard concentration for the j th parameter based on the national standards for irrigation water (Table 1). The water quality was classified into five categories based on the P_i value: clean, versus lightly, moderately, significantly, and seriously polluted for P_i values of <0.5 , $0.5\text{--}3.0$, $3.0\text{--}5.0$, $5.0\text{--}10.0$, and >10.0 , respectively.

Results and discussion

Evolution of surface water quality

Table 1 shows the measured results for 17 surface water samples collected in 2008 by Qingyang Environmental Observation Station. Figure 2 presents the main surface water chemical and quality parameters in the Malian River Basin as a function of the distance from the northern border of Huanxian County. The samples were generally moderately alkaline, with pH ranging from 7.4 to 8.6. The DO concentrations were higher than 7 mg L^{-1} for most samples (Table 1), with exceptions of three samples lower than 3 mg L^{-1} (SW6, SW7, and SW15). The Cl^- and SO_4^{2-} concentrations were high in the upstream samples, but decreased with increasing distance downstream (Fig. 2). The main pollutants in the surface water were COD, $\text{NH}_4^+\text{-N}$, and Cr^{6+} , and phenol pollution was serious in the Hanjiawan section. The COD and BOD values decreased with increasing distance downstream (to about 200 km), then became particularly high farther downstream, reaching 120 mg L^{-1} (SW16) and 115 mg L^{-1} (SW11) COD, which greatly exceeded China's national standard of 40 mg L^{-1} . PI averaged 7.8 mg L^{-1} , with the highest concentration at Xifeng (SW11, 41.2 mg L^{-1}). Cr^{6+} pollution was serious, with a mean value of 0.1 mg L^{-1} and a maximum of 0.3 mg L^{-1} (SW2), versus a national standard value of $0.05\text{--}1.0 \text{ mg L}^{-1}$. $\text{NH}_4^+\text{-N}$ and TP also increased with distance downstream and exceeded the safety criteria for irrigation or drinking water (Table 1). The P_i value belonged to the severely polluted class (>10.0) in six samples and was significantly polluted (>5) in five other samples; only three samples were classed as lightly polluted.

The overall water quality varied greatly over time in the Malian River from the long-term observations by the Qingyang Environmental Monitoring Station (Quzi, Hanjiawan, and Ningxian sections (Figs. 1, 3). Most of the surface water in the Malian River from the Quzi section to the Ningxian section is severely polluted. At the same time, the polluted river in Longdong Loess Plateau has taken 48% of the total river length (QEPO 2000). The pollution began to become serious at the end of the 1970s, when development of the oil fields began. By 1984, pollution in the Malian River caused by the oil industry was particularly severe, with an average petroleum content of 44.85 mg L^{-1} and a maximum value of 450.0 mg L^{-1} , which is 1,499 times the value in the national standard for drinking water (QEPO 2000). However, the petroleum pollution decreased after 1994; from 1995 to 1998, its value ranged between 0.5 and 3.0 mg L^{-1} , and has been lower than 1.0 mg L^{-1} since 2000. However, COD and PI increased since 2000 and reached a maximum around 2004.

Groundwater hydrochemistry and pollution

Groundwater geochemical properties

According to the Piper diagram for all samples obtained in the study area, the groundwater was generally alkaline, and the concentrations of the individual ions varied greatly (Fig. 4). The cations in shallow groundwater were mainly Na^+ and K^+ , but the anions showed no clear domination. The cations in deep groundwater were mainly Ca^{2+} and Mg^{2+} , and the anions were mainly HCO_3^- . Because the aquifer is mainly loess, which has a limited cation exchange capacity, the Ca^{2+} and Mg^{2+} contents in the groundwater were higher than the Na^+ (Table 2). However, owing to the concentration of clay minerals in the aquifer gradually increasing in the discharge area, the absorbed Na^+ is easily replaced by Ca^{2+} and Mg^{2+} in groundwater, leading to increasing Na^+ in the groundwater and decreasing Ca^{2+} and Mg^{2+} . The mCa/Mg ratio increases along the water's flow direction, indicating that dissolved Ca^{2+} replaces Mg^{2+} in MgCO_3 and produces CaCO_3 precipitate. The TDS of shallow groundwater is higher due to evaporation, ranging from 246 to $3,410 \text{ mg L}^{-1}$. In contrast, the deep groundwater has lower TDS of $230\text{--}552 \text{ mg L}^{-1}$.

Along the groundwater flow direction, the concentrations of Mg^{2+} and Na^+ increase to different degrees, whereas that of Ca^{2+} decreases, and the concentrations of all three cations are positively correlated with Cl^- and SO_4^{2-} (Fig. 5). The concentration of Na^+ in groundwater is higher than that of Cl^- , indicating that intense water-rock interactions have occurred. The imbalance between Na^+ and Cl^- indicates that in addition to the dissolution of

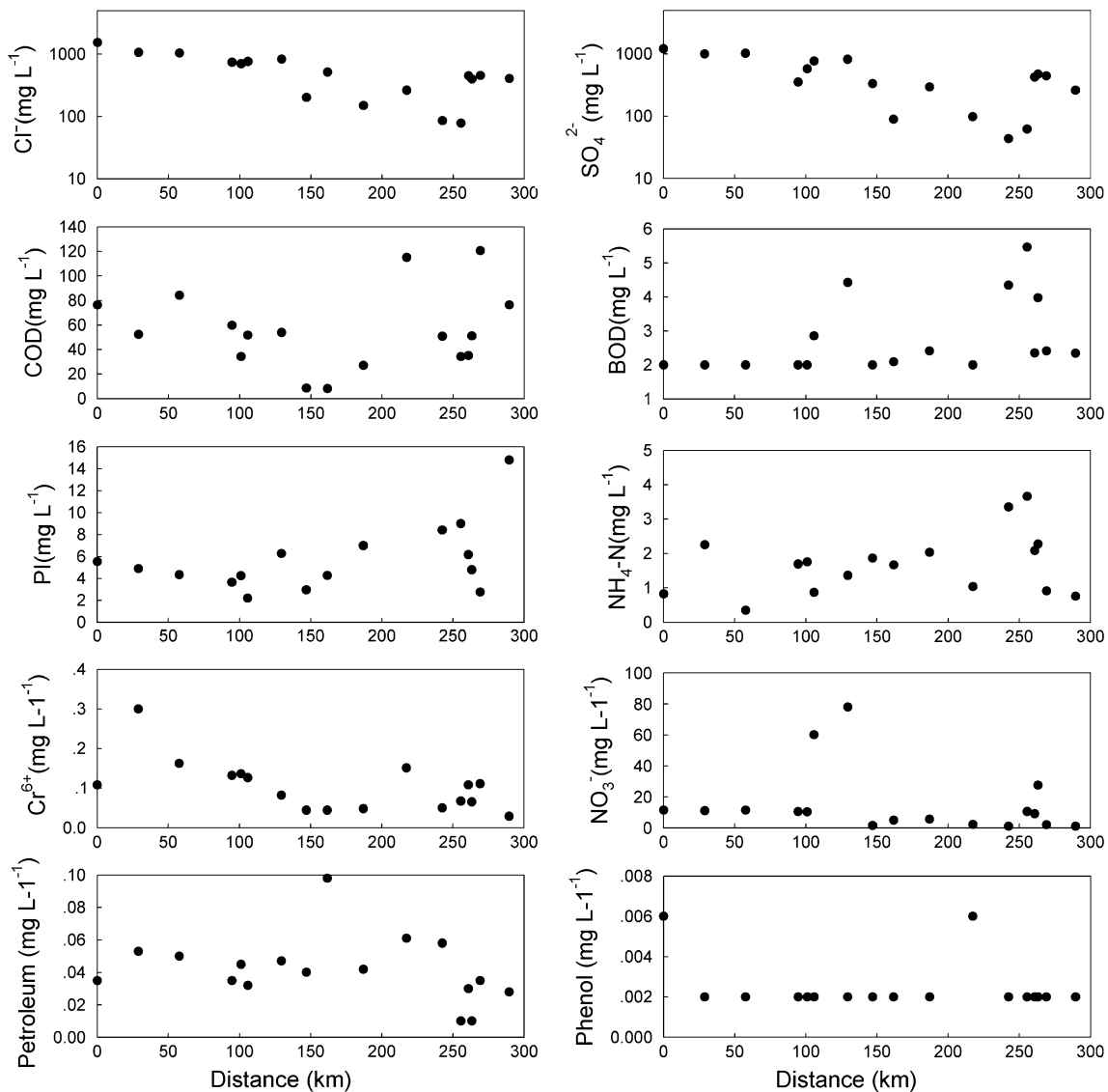


Fig. 2 Plots of the main surface water parameters as a function of distance downstream from the border of Huanxian County in the direction of Ningxian. Cl^- chloride, COD chemical oxygen demand, P total phosphorus, Cr^{6+} chromium, petroleum all petroleum

compounds, SO_4^{2-} sulfate, BOD biological oxygen demand, NH_4-N ammonium nitrogen, NO_3^- nitrate nitrogen, phenol total volatile phenols

NaCl, Na-containing minerals such as sodalite are dissolving, which is similar to the majority area of northwestern China (Ma et al. 2008). Along the water flow direction, the decrease of Ca^{2+} and increase of SO_4^{2-} and Mg^{2+} indicates that dissolved Ca^{2+} replaces Mg^{2+} in dolomite to produce $CaCO_3$ precipitate.

Groundwater quality and pollution

The groundwater in 6 of 21 samples was seriously polluted by permanganate, with the maximum value reaching 465.7 mg L⁻¹ (Table 2). These polluted areas were mainly distributed between Huanxian County and Qingcheng County. Under normal circumstances, the natural

background value of groundwater PI is less than 2, but shallow wells with a groundwater depth less than 15 m (G4, G17, G18) are vulnerable to surface pollution. Nitrite is another potential problem because it is a toxic (carcinogenic) substance, and commonly results from agricultural use of nitrogen fertilizer, which is converted into nitrite through biotic and abiotic processes. The nitrite level was less than 0.1 mg L⁻¹ in most groundwater samples, but values were greater than 200 mg L⁻¹ at four deep wells (G3, G13, G14, G15). The water sample analysis by local Environmental Office revealed that the concentration of NO_3^- -N in the groundwater is remarkably high and reached 10–12 mg L⁻¹ near some agricultural areas in Huanxian and Qingcheng Counties (QEPO 2009). These

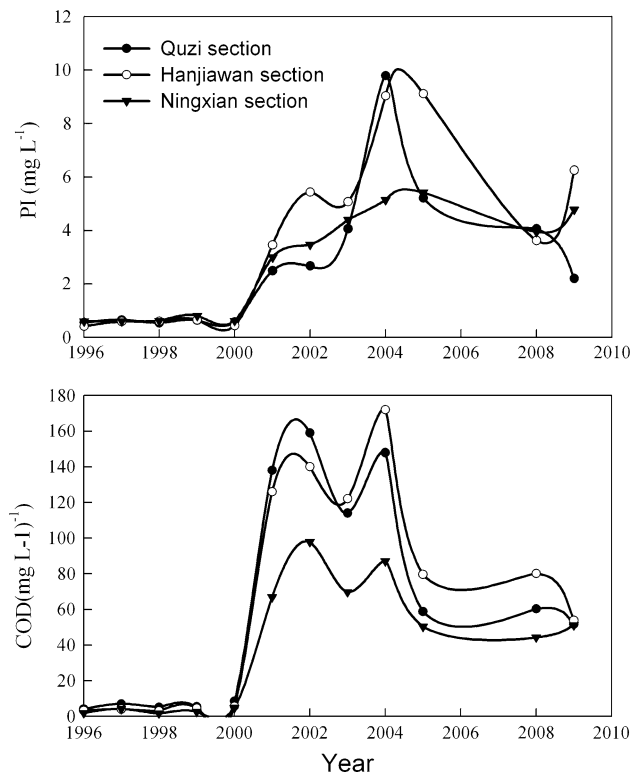


Fig. 3 Changes in permanganate index (PI) and chemical oxygen demand (COD) in three key sections of the Malian River since 1996

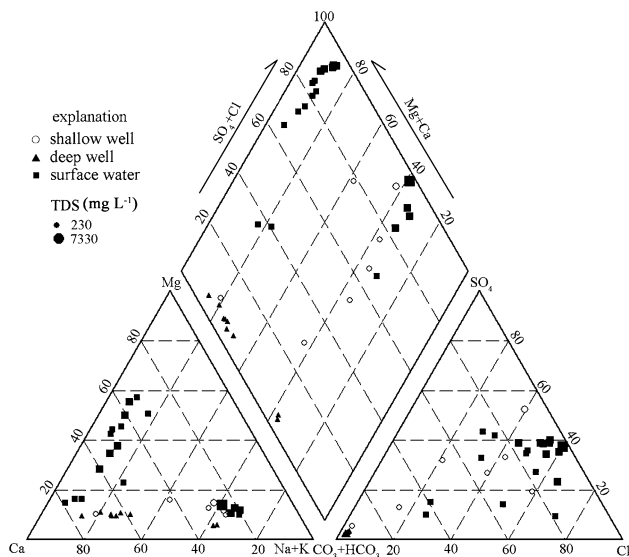


Fig. 4 Piper diagrams for the geochemistry of the major ions for all water samples in the study area

concentrations are more than two times the drinking water standard (5.0 mg L^{-1}). Trace elements such as Fe, Mn, and Cr^{6+} were also detected at levels higher than the drinking water standards in several wells. The highest Fe level was 0.44 mg L^{-1} (G1), whereas the highest level of Mn was 1.20 mg L^{-1} . $\text{NH}_4\text{-N}$ ranged from 0.05 to 1.09 mg L^{-1} ,

with a mean value of 0.26 mg L^{-1} . The average coliform bacteria count was 124, with the highest value (372) in well G8, which was 180 m deep. This pollution is related to the rapid industrial development of the Xifeng region. There is no sewage treatment plant in Xifeng, as a result, most industrial wastewater is not treated and directly pollutes the groundwater of the loess tableland.

Pollution sources

The results revealed high variation in groundwater quality, with severe pollution levels in some regions, which relates to factors such as the main local industry, agricultural fertilizer and pesticide, levels of domestic sewage discharge.

Sources of agricultural pollution

Agricultural pollution in the study area results mainly from the use of pesticides and fertilizers. In the 1990s, the total annual pesticide used in the study area averaged $302.04 \times 10^3 \text{ kg}$ per year, which amounts to 46.32 kg ha^{-1} , and about 79% of the cultivated area was used pesticides. The fertilizer use in the study area totaled $23.36 \times 10^9 \text{ kg}$ per year, which amounts to an average use of 265.7 kg ha^{-1} , and fertilizer was used in 96% of the study area.

Along rivers and in areas with shallow groundwater, agricultural fertilizer easily enters the water through leaching, percolation, and soil erosion. On mountain ridges, groundwater is deep and rainfall is rare, although it is hard to directly pollute the groundwater resource, residual pesticides and fertilizers in steep, cultivated land enter ravines through water loss, soil erosion, and overland flow, finally entering and polluting rivers. When excessive nitrogen fertilizer is used, the nitrogen not absorbed by plants is transformed into NH_3 (which is lost into the air through volatilization or transformed into NH_4^+ in water) or is transformed into NO_3^- (mainly in the surface soil). Because NO_3^- is stable in oxidation condition and is difficult to be absorbed by soil particles, most of the NO_3^- enters the groundwater and causes long-term and extensive pollution. Pesticides enter the water in a similar manner because of seldom biotic or abiotic processes. It is clear that agricultural pollution caused by pesticides and fertilizers is serious in these areas. However, the proposed development of green agriculture will gradually reduce the use of pesticides and fertilizers.

Sources of domestic pollution

The Malian River receives domestic sewage from Huanxian, Qingcheng, Huachi, Heshui, and Ningxian and Xifeng.

Table 2 The main chemical and pollutant data and statistical results for the groundwater samples collected in the Longdong Loess Plateau along the Malian River Basin

Sample no.	Depth (m)	pH	TDS (mg L ⁻¹)	Cl ⁻ (mg L ⁻¹)	SO ₄ ²⁻ (mg L ⁻¹)	HCO ₃ ⁻ (mg L ⁻¹)	NO ₃ ⁻ (mg L ⁻¹)	Na ⁺ (mg L ⁻¹)	K ⁺ (mg L ⁻¹)	Mg ²⁺ (mg L ⁻¹)	Ca ²⁺ (mg L ⁻¹)	PI (mg L ⁻¹)	Fe (mg L ⁻¹)	Mn (mg L ⁻¹)	Petroleum (mg L ⁻¹)	NH ₄ ⁺ -N (mg L ⁻¹)	Cr ⁶⁺ (mg L ⁻¹)	Coliform (cfu L ⁻¹)	
G1	470	7.70	1,701	245.2	857.9	172	3.75	696.87	1.97	7.32	73.01	1.09	0.442	0.01	0.026	0.096	0.005	76	
G2	26	7.71	457	41.56	45.62	324	37.00	103.38	1.57	12.06	47.73	-	-	-	-	-	-	-	
G3	14	7.76	851	76.82	155.4	364	37.22	302.71	4.35	10.00	62.01	0.0	0.161	0.022	0.010	0.057	0.004	88	
G4	13	7.60	1,813	370.7	625.3	306	12.83	665.21	6.99	17.15	113.0	237.51	0.261	0.045	0.010	0.385	0.003	91	
G5	15	7.67	576	72.13	144.9	271	12.18	148.25	1.79	11.90	51.02	-	-	-	-	-	-	-	
G6	7.83	255	5.80	7.52	281	13.15	21.82	0.30	6.26	65.44	0.50	0.03	0.01	0.010	0.276	0.058	36		
G7	7.80	252	4.92	7.01	274	13.64	29.15	0.53	5.56	62.64	0.64	0.03	0.01	0.010	0.289	0.004	136		
G8	8.11	236	4.41	5.14	271	9.47	72.55	0.39	3.27	32.51	0.50	0.03	0.01	0.010	0.173	0.072	372		
G9	8.14	256	5.69	4.63	292	13.80	34.29	0.51	5.83	57.49	-	-	-	-	-	-	-	-	
G10	8.26	252	3.87	5.43	308	10.39	29.93	0.47	6.02	59.54	0.57	0.03	0.01	0.020	0.285	0.041	158		
G11	8.15	238	4.47	6.36	271	10.80	24.78	0.73	5.08	58.45	-	-	-	-	-	-	-	-	
G12	8.30	230	4.31	6.15	265	7.63	23.73	0.45	5.06	55.96	-	-	-	-	-	-	-	-	
G13	7.30	239	3.49	5.75	263	9.70	13.14	2.21	4.79	65.63	0	0.075	0.01	0.010	0.106	0.032	62		
G14	7.81	246	5.22	12.16	261	10.73	19.80	0.32	5.59	64.51	0	0.03	0.01	0.010	0.052	0.027	130		
G15	8.14	249	3.43	4.19	288	5.63	75.53	0.74	3.71	32.24	0.0	0.03	0.01	0.010	0.099	0.034	280		
G16	7.95	998	315.6	315.6	138.5	200	46.02	133.70	7.70	27.58	120.45	-	-	-	-	-	-	-	
G17	7.90	970	234.3	234.3	215.9	346	13.27	234.80	1.69	20.68	75.80	261.16	0.03	0.01	0.010	0.146	0.055	143	
G18	7.83	1,189	295.1	295.1	306.6	287	34.27	274.78	1.61	26.24	97.80	465.65	0.407	0.028	0.024	0.36	0.017	120	
G19	7.67	3,410	818.7	818.7	1,462.7	283	221.53	810.95	9.16	110.41	335.66	1.28	0.092	0.01	0.019	0.198	0.003	196	
G20	8.24	2,730	760.4	760.4	903.4	112	26.28	1,231.25	31.31	16.83	139.81	1.45	0.289	0.01	0.014	0.132	0.003	112	
G21	7.63	3,544	199.5	199.5	861.9	219	0.00	728.45	9.86	7.79	147.23	0.64	0.03	0.01	0.010	0.097	0.002	12	
Sample number	-	21	21	21	21	21	21	21	21	21	21	91	91	91	91	91	91	91	91
Mean	-	7.88	985	165.5	275.4	269	26.16	270.24	4.03	15.20	86.57	75.62	0.48	0.06	0.04	0.30	0.03	118	
Minimum	-	7.30	230	3.43	4.19	112	0.00	13.14	0.30	3.27	32.24	0.00	0.03	0.01	0.00	0.03	0.00	4.00	
Maximum	-	8.30	3,544	818.7	1,462.7	364	221.53	1,231.25	31.31	110.41	335.66	346.43	7.47	1.20	2.10	2.77	0.11	499	
SD	-	0.26	1,064	241.4	414.8	56.52	46.43	345.52	6.96	22.99	65.32	122.54	1.27	0.19	0.22	0.43	0.03	109.9	

Parameter definitions: TDS total dissolved solids, PI permanganate index, petroleum all petroleum compounds

Fig. 5 Graphs of TDS, major ions (Na^+ , SO_4^{2-}) and two ratios, $m(\text{Na}/\text{Cl})$ and $m(\text{Mg}/\text{Ca})$, in groundwater as a function of the Cl^- concentration and Ca^{2+} as a function of the SO_4^{2-} concentrations

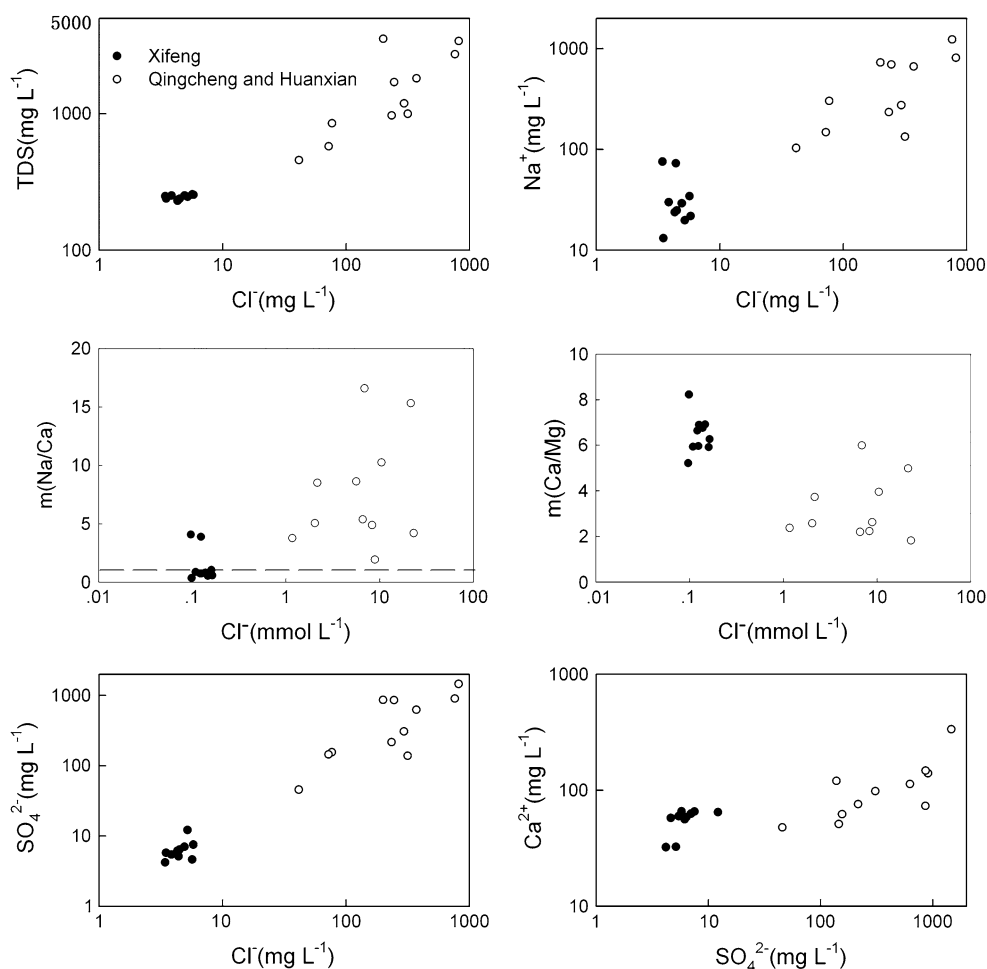


Table 3 Total domestic water consumption and wastewater discharge from different towns in the Longdong Loess Plateau of the Malian River Basin

Location	Water consumption ($\times 10^4 \text{ m}^3$ per year)	Domestic sewage ($\times 10^4 \text{ m}^3$ per year)	COD ($\times 10^3 \text{ kg}$ per year)	$\text{NH}_4^+\text{-N}$ ($\times 10^3 \text{ kg}$ per year)
Xifeng	357.33	268	2,012.6	234.80
Qingcheng	161.33	122	1,462.92	170.67
Huanxian	49.43	40	477.4	55.70
Huachi	48.97	37	446.8	52.10
Total	617.06	467	4,399.72	513.27

In 2006, the annual volume of domestic sewage was $467 \times 10^4 \text{ m}^3$, amounting to 76.1% of the total wastewater received by the Malian River. This pollution includes $4,400 \times 10^3 \text{ kg}$ of COD and $513 \times 10^3 \text{ kg}$ of NH_4 (Table 3). Because no sewage treatment plant exists, domestic sewage is discharged directly into the surface water, becoming one of the main factors on the Malian River's water quality.

Sources of industrial pollution

The industrial wastewater and domestic sewage from Huanxian County and Qingcheng County is discharged into the Malian River, and that from Huachi County is discharged into the Rouyuan River, the tributary of Malian River. There are 28 major industrial wastewater sources in the study area, with annual emissions of $185.58 \times 10^4 \text{ m}^3$, accounting for 23.9% of the total emission volume entering the Malian River. These emissions include $566.74 \times 10^3 \text{ kg}$ of COD, $19.81 \times 10^3 \text{ kg}$ of $\text{NH}_4\text{-N}$, $1.58 \times 10^3 \text{ kg}$ of volatile phenols, and $15.80 \times 10^3 \text{ kg}$ of petroleum. The Qingyang Petrochemical Company is the largest producer of industrial wastewater, with an annual volume of $86.2 \times 10^4 \text{ m}^3$, which includes $319.5 \times 10^3 \text{ kg}$ of COD, $2.6 \times 10^3 \text{ kg}$ of $\text{NH}_4\text{-N}$, $1.5 \times 10^3 \text{ kg}$ of volatile phenols, and $15.5 \times 10^3 \text{ kg}$ of petroleum (QEPO 2009). The major industrial pollution is concentrated from the Quzi section to the Hanjiawan section. Because there is no sewage treatment plant, these industrial wastes are discharged directly into the Malian River together with domestic wastes. The polluted river water is the main source of domestic water

supply for many residents in this area, so this pollution represents a serious problem.

Discussion

Human activity has become an important geological agent that affects surface water and groundwater both directly and indirectly, leading to imbalances in the chemical composition of this water. The substances released by humans included industrial wastes, domestic sewage, organic and inorganic fertilizers, and pesticides. However, petroleum contamination in the Longdong oilfield is the largest source of pollution.

The Longdong oilfield is one of the three main oilfields operated by the Changqing Branch of PetroChina Co. Ltd, and supplies Oil Plant 2 of the Changqing Oilfield Company, the largest crude oil producer in Gansu Province (Huang 2006). There are currently 11 large oilfields, including the Maling, Fanjiachuan, Huachi, Chenghao, Nanliang, and Xifeng fields, which cover an area of nearly 26,000 km² (Fig. 1). The estimated total crude oil reserves in these fields is about 250 Mt. The Longdong oil field included more than 2,800 working oil wells by the end of 2008 (Table 4), and the number was increasing rapidly. Oilfield development comprises exploration, drilling of wells, borehole operation, oil extraction, oil and gas collection and transportation, and infrastructure construction. In each of these phases of the rapidly expanding operation, inefficient techniques and equipment defects, operating errors, and accidents cause pollution (Tellez et al. 2002).

Well drilling is one of the main oil development processes, and uses large amounts of fresh water. During drilling, solid wastes, wastewater discharge, and pollutants discharged by drilling accidents all affect the water quality.

The wastewater produced during drilling includes equipment drilling and cleaning water, both of which contain pollutants such as petroleum, suspended substances, and compounds that increase the COD. Under normal conditions, this wastewater is stored in pools protected against seepage and is not discharged, but if the storage area is not properly constructed, these pollutants may enter the groundwater through seepage. Solid wastes produced by drilling are mainly discarded slurry and rock debris, and the main components in the discarded slurry are clays, colloids, and alkali materials. The discarded slurry produced by a single well in the Longdong oilfield covers about 200 m² and the rock debris covers about 30 m³ (QEPO 2009). If the mud storage area is not sealed against seepage, rainfall can cause the pollutants contained in these solid wastes to enter the ground, polluting the groundwater.

In the Longdong oilfield, the water concentration in the crude oil averages 75.8% v/v (Table 4), and after separation of the oily water from the oil at the collection and transportation station, the resulting water is stored at a depth of 1,000 m below ground. During the extraction and transportation of crude oil, some oil is spilled and pipes may leak, polluting the soil. The expanding oil production at the Longdong oilfield is steadily increasing the volume of water consumed, and the ever more extracted water is being used for injection after treatment (Wang et al. 2008). Because the extracted water comes from different layers, its water composition is complex, with large quantities of bacteria, suspended substances, oil, SO₂, H₂S, CO₂, and other contaminants that make the water strongly corrosive. As a result, the water easily corrodes the oilfield equipment. If daily maintenance of injection wells is not performed or is performed poorly, corrosion of the oil jacket may let the extracted water seep into non-target layers, causing groundwater pollution (Wang et al. 2003). Oxygen

Table 4 The statistics for oil wells and injection wells in the Longdong oilfield

Oilfield	Total no. of wells	No. of oil wells	No. of injection wells	Oil production (× 10 ⁴ m ³ per year)	Water rate of oil (%)	Injection water (m ³ per day)
Maling	491	355	136	25.69	81.3	4,468
Huachi	520	344	176	34.34	64.8	5,663
Yuancheng	175	164	11	20.67	66.1	453
Nanliang	377	254	123	21.61	36.7	4,846
Chenghao	125	90	35	8.28	78.1	1,223
Xifeng	1,657	1,235	422	106.95	25.4	12,299
Fanjiachuan	109	84	25	5.48	75.3	632
Zhengbei	300	227	73	25.59	21.3	1,331
Wujiao	53	45	8	4.71	77.4	268
Yanwu	17	13	4	0.69	77.7	104
Total	3,824	2,811	1,013	254.01	–	31,287
Average	–	–	–	–	75.8	–

is the most harmful corrosion factor during the treatment of oil-containing wastewater and wastewater injection. The salinity and hardness of the wastewater is so high that precipitates can clog equipment and pipelines, leading to additional seepage of injected wastewater and the spread of pollutants into non-target layers (Li et al. 1996; Wang et al. 2003).

Abnormal circumstances such as oil blowouts, pipe corrosion, and poor well cementation may cause additional pollution of the groundwater. Oil blowouts are serious accidents that occur when the drill encounters a high-pressure oil–water layer, leading to an explosion of oily water from the well’s mouth. This spreads crude oil, oily sewage, and other materials over a large area, eventually leading to groundwater contamination because it is difficult or impossible to recover these materials.

During or after drilling, poor well cementation quality or poor casing pipe quality can let oily water seep into other layers, including gaps between strata. There are 81 water source wells used for oil extraction and water injection in the Longdong oilfield, including 23 water source wells created from old oil wells and 20 water source wells created from injection wells. It is important to carefully control the conversion of oil wells and injection wells into water sources so as to avoid seepage of oily water. The potential for an oil spill is significant, and the effects of spilled oil pose serious threats to the environment. Leaking underground and aboveground storage tanks, improper disposal of petroleum wastes, and accidental spills are major routes of soil and groundwater contamination by petroleum products. Pipeline breaks are relatively rare accidents during oil production, but the resulting damage is great. When a pipeline breaks, the pressurized crude oil escapes at a high velocity, and the oil and oily wastes can pollute groundwater over a large area. When an oil pipeline is installed in an area that cannot support its weight, natural collapse of the underlying soil and rocks can lead to breakage of the pipeline under its own weight. Petroleum products are mostly lighter than water, and when they reach aquifers, a pool of floating non-aqueous-phase liquids forms over the water table. The major problem associated with subsurface petroleum contamination is the dissolution of highly toxic light aromatic hydrocarbons in the groundwater.

Pollution by the oil industry is a particularly serious problem. Nadim et al. (2000) summarize the techniques used for treating hydrocarbon-contaminated soils and groundwater and limitations on their application. For example, the “pump and treat” technique can be used to extract contaminated groundwater from below the surface using extraction wells. The contaminated groundwater can then be treated by means of air stripping, passage through activated carbon, or bioremediation. However, such

treatments are expensive and time-consuming. It would be more effective to improve the management of oil extraction operations and handling of the extracted petroleum products and of wastes to reduce the possibility of spills and pollution.

Conclusions

The ion and pollutant concentrations in surface water and groundwater have changed with unprecedented speed, making much of the water unsafe to drink and unsuitable for use in irrigation in the Malian River Basin and Longdong Loess Plateau. The substances released by humans included industrial wastes, domestic sewage, organic and inorganic fertilizers, and pesticides, as well as a range of other substances that are harmful to humans. However, petroleum contamination produced by the oil industry in the Longdong oilfield is the largest source of pollution that infiltrates surface water and groundwater in various ways, leading to severe deterioration of water quality.

Protecting the water resources in the study area will be a formidable challenge. The discharge of wastes into the surface water and groundwater must be drastically reduced so the ecosystem’s natural ability to degrade these wastes will not be exceeded. Achieving this goal will require modernization of all the industrial, agricultural, and domestic processes that use water, thereby improving the recovery and recycling of wastewater, especially in the oilfields. Water treatment facilities must be established for all urban and industrial areas, which currently discharge their wastes directly into the river. Sufficient water treatment facilities must be made available, and old facilities must be upgraded to increase their capacity. Agricultural techniques must be modified to greatly reduce the quantities of pesticides and fertilizer that are used; for example, integrated pest-management techniques should be adopted to reduce the need for pesticides, slow-release fertilizers should replace older forms to reduce leaching of nutrient, and drainage water from agricultural fields should be captured and recirculated in those fields so that the dissolved nutrient ions can be taken up by crops. The subsurface medium must be protected against any kind of hazardous contamination because soil acts as a long-term residence for pollutants, and dynamic movements of the hydrological cycle transport them into groundwater aquifers.

Acknowledgments The research was supported by the Keygrant Project of Chinese Ministry of Education (No. 310005), NSFC Innovation Team Project (No. 40721061) and Foundation of Changqing Oilfield Limited Company (09AK-KF-003). We thank Mr. Liu of Qingyang Environmental Observation Station for pollutants analyses and valuable advices and Mrs Wang of Key Laboratory of

Western China's Environmental Systems (Ministry of Education), Lanzhou University for chemical analysis. We also appreciate the contributions of anonymous reviewers and Geoff Hart, all of whom contributed to improve the manuscript.

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