Review

Deep challenges for China's war on water pollution☆

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ABSTRACT

China's Central government has released an ambitious plan to tackle the nation's water pollution crisis. However, this is inhibited by a lack of data, particularly for groundwater. We compiled and analyzed water quality classification data from publicly available government sources, further revealing the scale and extent of the crisis. We also compiled nitrate data in shallow and deep groundwater from a range of literature sources, covering 52 of China's groundwater systems; the most comprehensive national-scale assessment yet. Nitrate pollution at levels exceeding the US EPA's maximum contaminant level (10 mg/L NO_3-N) occurs at the 90th percentile in 25 of 36 shallow aquifers and 10 out of 37 deep or karst aquifers. Isotopic compositions of groundwater nitrate ($\delta^{15}N$ and $\delta^{18}O_{NO3}$ values ranging from $-14.9‰$ to $35.5‰$ and $-8.1‰$ to $51.0‰$, respectively) indicate many nitrate sources including soil nitrogen, agricultural fertilizers, untreated wastewater and/or manure, and locally show evidence of de-nitrification. From these data, it is clear that contaminated groundwater is ubiquitous in deep aquifers as well as shallow groundwater (and surface water). Deep aquifers contain water recharged tens of thousands of years before present, long before widespread anthropogenic nitrate contamination. This groundwater has therefore likely been contaminated due to rapid bypass flow along wells or other conduits. Addressing the issue of well condition is urgently needed to stop further pollution of China's deep aquifers, which are some of China's most important drinking water sources. China's new 10-point Water Pollution Plan addresses previous shortcomings, however, control and remediation of deep groundwater pollution will take decades of sustained effort.

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1. Introduction: China's 'war on pollution' and the new 10-point plan

In 2014, Chinese Premier Li Keqiang publicly declared 'war on pollution' and the Chinese Central Government has since announced major policies in the area of pollution control and remediation (Branigan, 2014; Zheng, 2015). The Water Pollution Prevention and Control Action Plan ("10-Point Water Plan") was released in April 2015 (Central People's Government of the People's Republic of China, 2015). This is arguably the most comprehensive policy yet aimed at tackling pollution of groundwater and surface water, which are recognized as being among the most severely degraded natural resources in China, and among the most heavily polluted water sources in the world (Ma, 2004; Gleick, 2009; Shapiro, 2012). Despite recent improvements in the provision of clean drinking water (Liu, 2015), it is estimated that more than 200 million people in China are still using unsafe water sources (Tao and Xin, 2014; Liu, 2015). Since 1995, it is estimated that 11,000 water quality-related emergencies have occurred — a recent example being the contamination and temporary shutdown of the city water supply (sourced from groundwater) in Lanzhou, because of contamination by benzene from a nearby petrochemical facility (Yan, 2015). It is also estimated that each year thousands of incidents of civil unrest related to water pollution take place in China, predominantly in rural areas (Gilbert, 2012).

The new water pollution action plan breaks from previous approaches to China's water crisis that focused on large-scale engineering solutions for the provision of clean water (e.g. Liu and Yang, 2012). The plan includes ambitious targets for improvement of

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groundwater and surface water quality over coming decades. The promulgation of this and the analogous 10-point plan for air pollution (Central People's Government of the People's Republic of China, 2013) signifies that China's new generation of leaders recognize that pollution poses a risk to China's ongoing development and social stability, and are serious about improving the prospects for all Chinese people to have access to clean and secure water.

Control and remediation of water pollution requires accurate data to diagnose the nature and extent of the problem. While data collection and transparency has improved in China recently, there remain major gaps which hamper accurate assessment at the range of scales required, e.g., major river catchment or groundwater basin to sub-catchment or sub-basin (Liu, 2015). This is particularly true for groundwater, where data is sparse apart from aggregated regional or national statistics (Ministry of Land and Resources, 2014). In this review we address these gaps to the extent currently possible by: 1) compiling and visualizing the most up-to-date public data on water quality classes in China's rivers and aquifers from government statistics at national and regional scales; 2) compiling water quality analyses from published and unpublished research sources, focusing on nitrate – a ubiquitous indicator of anthropogenic groundwater contamination; 3) compiling and analyzing data on the isotopic composition of nitrate, providing insight into major sources of pollution.

In doing so, we help to better illustrate the scale of China's water pollution challenges and uncover key processes driving groundwater quality degradation. These data indicate that contamination of deep groundwater, which is generally thought to be a relatively safe source of drinking water, is occurring on a large scale, a phenomenon which is poorly documented and may not yet be well understood by the relevant authorities. We argue that protection of this high quality groundwater should be a key focus in China's ongoing war against water pollution.

2. Materials & Methods

Data for the water pollution maps presented below (Fig. 1) were compiled from three major sources: China's Ministry of Land and Resources, Ministry of Environmental Protection and State Oceanic Administration bulletins (Ministry of Land and Resources, 2010; Ministry of Land and Resources, 2014; Ministry of Environmental Protection, 2014; State Oceanic Administration, 2015). The data in these bulletins do not include Hong Kong, Macao and Taiwan. Surface and seawater quality class data were aggregated onto a single map (Fig. 1a), color-coded for the water quality classes set out in China's national standards. Groundwater data was converted into charts showing the percentage of water quality occupying the 5 different classes for shallow and deep groundwater, respectively, within six sub-regions of China – the highest resolution data currently available (Fig. 1b). On top of these maps the locations of 351 'cancer villages', as reported in Gong and Zhang (2013), were overlaid for reference and compiled by region (Table 1). Groundwater nitrate data from 52 of China's major groundwater basins, including shallow and deep aquifers (Fig. 2), was compiled and extracted from 71 sources as either NO_3 or $NO_3–N$ (Supplementary Fig. S1). All raw NO_3 concentrations were converted to $NO_3–N$, and the data was again divided into 'shallow' and 'deep' groundwater according to the classification in the original source, or if this was not specified, a cut-off depth of 100 m. Summary statistics, including median, 10th and 90th percentiles and inter-quartile ranges were calculated for each dataset and aggregated as a set of box-plots (Fig. 2). Nitrate isotope data ($\delta^{15}N_{NO3}$ and $\delta^{18}O_{NO3}$) were collected from 31 literature sources, covering ten groundwater systems across China (see Supplementary Table S1). These data

were compiled and plotted on a bivariate plot showing typical nitrate source isotopic compositions (after Kendall and McDonnell, 1998) and a series of box-plots showing ranges of isotopic values for different regions of China (Figs. 4 and 5).

## 3. Results: China's water pollution crisis – scale, extent and distribution

3.1. Overall water quality classification

China faces one of the most serious water shortage and pollution crises ever documented (Gleick, 2009; Tao and Xin, 2014). Based on data released by the Ministry of Environmental Protection (MEP), the State Oceanic Administration (SOA), and the Ministry of Land and Resources (MLR), we constructed pollution maps of the surface water and groundwater environments at the national scale (Fig. 1). Water quality in China is assessed according to a five or six-class ranking system under the Environmental Quality Standards GB383-2002, GB3097-1997 and GB/T 14848-93 for surface, marine, and groundwater, respectively (Table 2). These classifications are universally recognized indicators of water quality in China. The water quality class is determined by monitored levels of ~30 indicator pollutants and chemical indices, with the pollutant or index recorded at the highest concentration relative to guideline levels used to attribute water into one of the classes (Ministry of Environmental Protection, 2002). In general the indicators which most frequently exceed guideline levels by the highest amounts (and therefore determine water quality class) are ammonia-N, nitrate, nitrite, biological oxygen demand (BOD) and chemical oxygen demand (COD) (World Bank, 2006).

Fig. 1a shows water quality data from major river basins and near-shore coastal waters. Currently, out of 208,000 km of monitored river reaches in China, water quality in 31.4% reaches falls into class IV or worse, and thus is unfit for potable use or human contact. Water quality in 14.9% of river reaches is inferior to class V, indicating complete loss of potential for all consumptive uses or human contact. Of ten major watershed areas, only in the southwest and northwest is water quality in the majority of rivers rated as high to moderate (Classes I to III), while the major northern river systems – the Yellow, Liaohe, and Huaihe Rivers are rated as class IV or V, and the Haihe River as class VI (Fig. 1a). Water in six of nine major coastal bays in China is characterized as 'poor' or 'very poor' (Classes IV or V). In autumn 2014, the combined coastal area with water quality in class IV or V (unfit for human contact) covered 57,000 km^2. These areas are at the discharges of river systems that drain China's major industrial and agricultural regions, integrating numerous upstream pollution sources.

The quality of water in small tributaries feeding China's main river systems is generally recognized as being poorer than the main water courses themselves (Ebenstein, 2012; Yang and Zhuang, 2014). This is due to reduced dilution capacity of small streams compared to major rivers, and in some cases, poorer regulation of industrial wastewater discharge to small streams, which are typically in rural areas with poor environmental oversight. This means that surface water quality estimated at the national scale based on an assessment of water quality classes in major rivers (Fig. 1a) probably under-estimates the severity of surface water pollution, as smaller tributaries are excluded.

Fig. 1b shows the most detailed nation-wide data for groundwater pollution currently available in government statistics, using China's 5-class groundwater quality rating standard. Groundwater accounts for one-third of total water usage across the domestic, agricultural and industrial sectors in China, and approximately two-thirds of cities utilize groundwater as a major water supply. According to the latest Bulletin of Land and Resources of China
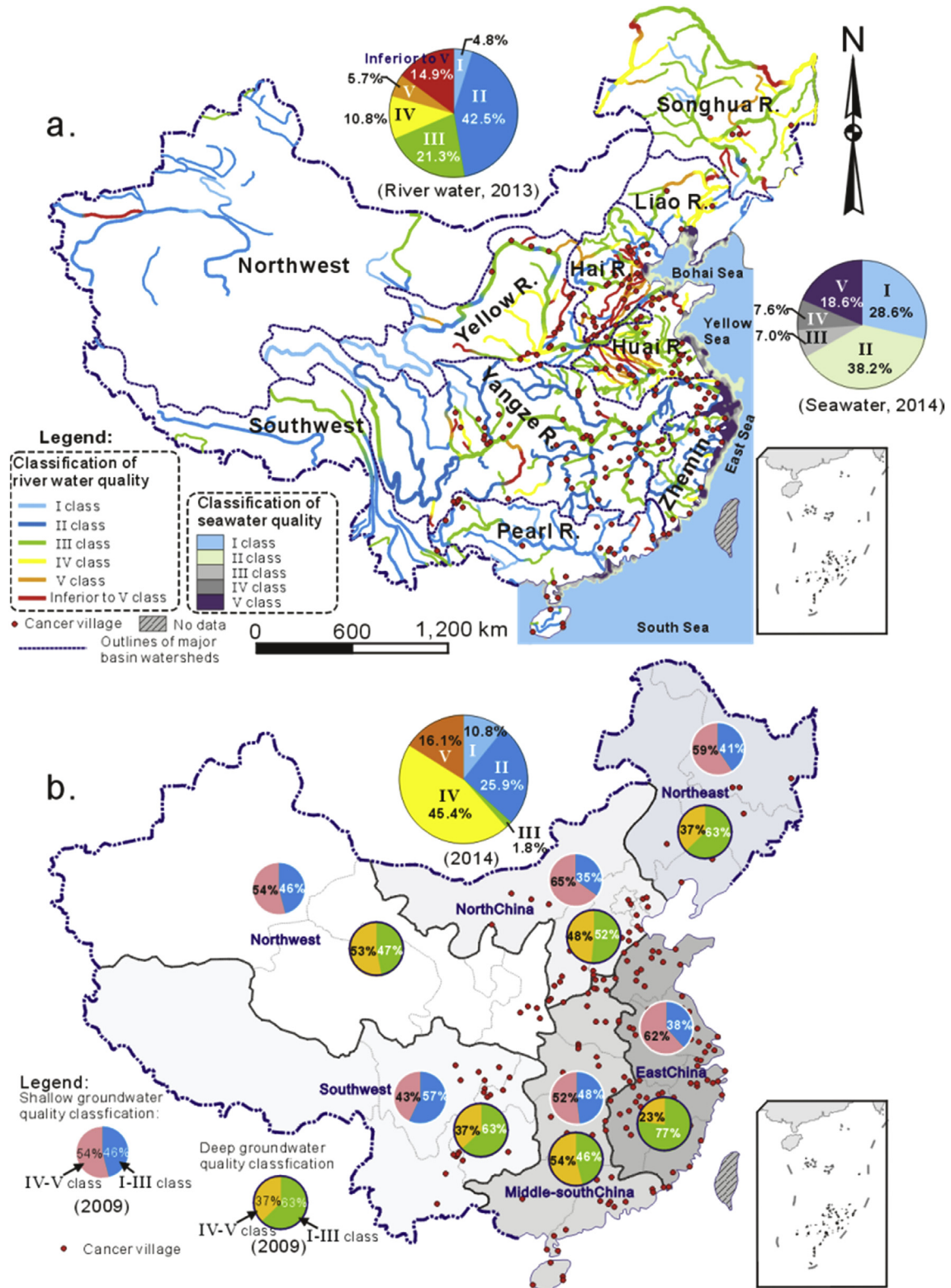
Fig. 1. Status of water pollution in China based on recent government statistics – A) Surface water (major rivers and seawater) ranked according to the 6-class water quality classification (GB 3838-2002 – see Table 1) and seawater quality of offshore areas ranked according to the 5-class classification (GB3097-1997); B) Groundwater ranked using the 5-class system (GB/T 14848-93) in 6 sub-areas of China, including shallow and deep groundwater. Overall percentages in each class for each water source in China are shown as the large pie-charts. Percentages shown in yellow and red on the smaller pie-charts on Fig. 1b indicate the proportion of samples in the lowest two classes (IV & V) for shallow and deep groundwater, respectively. Both maps (a & b) have been overlain with the locations of known 'cancer villages' (Gong and Zhang, 2013). For further detail on data sources and data processing see *Materials & Methods*.

Table 1

Cancer village distribution in China at provincial level, along with groundwater and river water quality classifications. CVN — cancer village number; CVD — cancer village density; CVN-r — number of cancer villages distributed in areas with river water quality inferior to grade III. The percentages of groundwater in grades IV & V is the ratio of wells with these grades to the total monitoring wells of the province. For river water quality, the percentage refers to the ratio of control sections monitored to the total control sections of the province in a given water quality grade. Data on cancer village distribution are from Gong and Zhang (2013).

Province	CVN	CVD	Groundwater IV—V class (%)	River water classification (%)				CVN-r
				I—III	IV—V	Interior to V	Interior to III	
HeiLongJiang	6	0.13	57.5	56.7	37.7	5.6	43.3	5
JiLin	1	0.05	27	67.5	24.4	8.1	32.5	0
LiaoNing	8	0.57	62	42.1	27.6	30.3	57.9	5
HeBei	40	2.21	63	48.6	21.4	30	51.4	34
BeiJing	1	0.63	43	52	7	41	48	1
TianJin	3	2.67	48.5	3.8	22	74.2	96.2	3
ShanDong	30	2.03	60	39.2	30.9	29.9	60.8	26
JiangSu	24	2.46	23	37.2	42.1	20.7	62.8	14
ShangHai	1	1.65	19	12.5	30.8	56.7	87.5	0
ZheJiang	20	2.01	30	66.2	18.9	14.9	33.8	2
FuJian	12	0.99	41	82.2	10.5	7.3	17.8	1
GuangDong	25	1.4	69	76	14	10	24	0
HaiNan	9	2.54	15	93.1	6.9	0	6.9	0
Inner Mongolia	9	0.08	55.4	40.4	40.5	19.1	59.6	1
ShanXi	15	0.99	51	48.5	24.2	27.3	51.5	13
HeNan	31	1.94	60	37.9	28	34.1	62.1	29
HuBei	17	0.94	46	77.4	11.1	11.5	22.6	1
HuNan	15	0.72	29	95.5	4.5	0	4.5	0
AnHui	19	1.4	90	70.5	17.8	11.7	29.5	16
JiangXi	18	1.09	66	91.2	4.2	4.6	8.8	0
GuangXi	2	0.08	35	92.5	7.3	0.2	7.5	0
ChongQing	11	1.37	69	74.7	25.3	0	25.3	1
Sichuan	6	0.13	57	83.2	12.7	4.1	16.8	2
ShaanXi	5	0.25	54	36.4	38.9	24.7	63.6	4
GuiZhou	3	0.17	48	74.4	3.9	21.7	25.6	1
YunNan	20	0.52	44	82.5	8.3	9.2	17.5	0
GanSu	0	0	50	63	12.3	24.7	37	0
NingXia	0	0	57	60	40	0	40	0
XinJiang	0	0	37	91.3	8.5	0.2	8.7	0
QingHai	0	0	—	100	0	0	0	0
Tibet	0	0	43	100	0	0	0	0

(2014), groundwater from 61.5% of 4896 monitoring wells in 202 cities across China was characterized as poor (IV class) or very poor (V class) (Fig. 1b) (Ministry of Land and Resources, 2014). Additionally, according to a recently published monthly groundwater status report of the Ministry of Water Resources, 80% of groundwater samples taken from more than 2000 shallow groundwater monitoring wells in China's northern basins falls into classes IV and V (Ministry of Water Resources, 2016). According to the national groundwater quality standard (GB/T 14848-93), water at or below Class IV is unfit for domestic or agricultural uses. The main pollutants above safe levels in these assessments, and which therefore determine the water quality classes, are the three types of nitrogen (NO_3—N, NO_2—N, NH_4—N), phenol, heavy metals and COD.

These data, maps and comparisons with previous surveys conducted by the Ministry of Land and Resources (Fig. 3) reveal two trends. One is a gradual expansion of the scale of groundwater pollution. The percentage of IV—V class water increased from 55.1% in 2011 to 61.5% in 2014; while an earlier assessment between 2000 and 2002 using fewer monitoring stations found the proportion in the lowest two classes was 37% (Shen, 2015). The second trend is increasing numbers of contaminants detected, signifying more complex pollution mixtures. However, as yet, individual data points and chemical concentrations are not disclosed in government data; the data is aggregated and reported as region-wide percentages (Fig. 1b), or numbers of monitoring stations falling into the various classes.

A key concern arising from these data is extensive pollution of deep groundwater, contained in semi-confined and confined aquifers, generally at depths greater than 100 m. This poses a challenge for future groundwater remediation efforts, as these aquifers (unlike shallow unconfined aquifers) are typically isolated from the rapid, surficial water cycle and require extremely long time periods — on the order of thousands of years — for natural flushing (Alley et al., 1999). Another concern is that while groundwater pollution does affect particular regions more or less seriously than others, both deep and shallow groundwater pollution is ubiquitous nation-wide in China. The most seriously affected regions are the densely populated North China Plain, where drinkable groundwater (I—III class) was found to occupy only 22.1% of shallow groundwater and 26.4% of deep groundwater (Duan and Gao, 2013); similar to the data in the more recent Ministry of Water Resources survey (Ministry of Water Resources, 2016). However, the more sparsely populated areas of northwest China are also not immune from serious groundwater pollution in both shallow and deep aquifers (Fig. 1b).

3.2. Nitrate concentrations and isotopic data across China

In addition to the government statistics reporting water quality classes at the national level, we compiled data from 71 studies of groundwater quality from the research literature, documenting concentration ranges of nitrate-N in 52 groundwater systems, including shallow (unconfined) and deep (confined) aquifers. We also compiled data for karst aquifers, where there is rapid vertical connectivity between different depths (Fig. 2). These data reveal a more detailed picture of the distribution of groundwater pollution by location and depth, basin-by-basin across China. Nitrate was selected for this purpose as it is an ideal 'gross' indicator of anthropogenic impact on groundwater, for the following reasons:
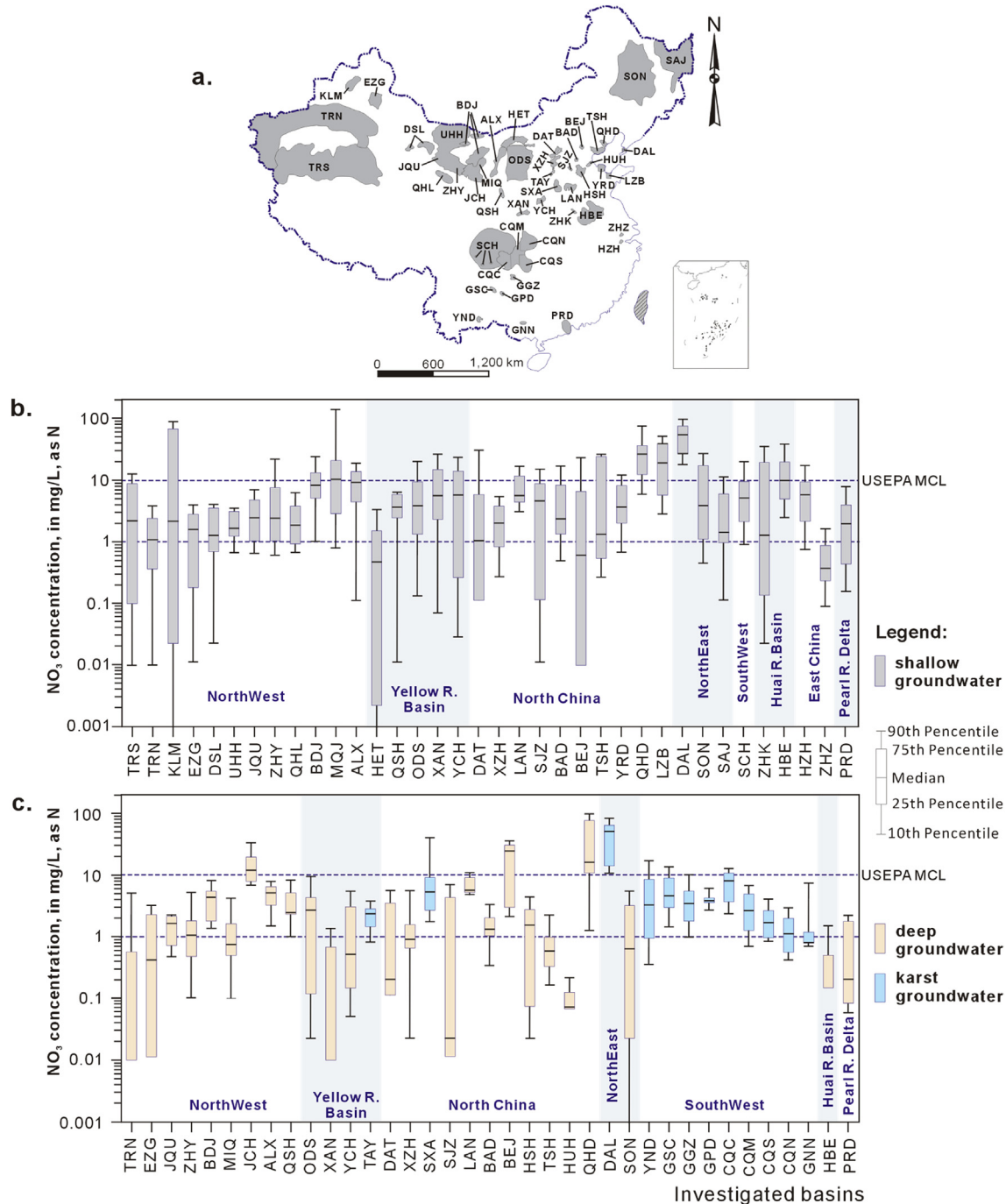
Fig. 2. Nitrate concentrations in groundwater from major groundwater systems in China: a) Location map of the 52 total study areas from which data were compiled; Boxplot distributions of nitrate concentrations (as N) in shallow groundwater (b) and deep or karst groundwater (c) throughout China. Boxplots show median, inter-quartile range and 10th and 90th percentile values. Data is compared to the United States Environmental Protection Agency maximum contaminant level (10 mg/L) and a background concentration of 1 mg/L, according to Burow et al. (2010). The data reveal endemic nitrate pollution in deep and shallow groundwater throughout China. For the full key to the location map and all data sources, refer to Supplementary fig. S1 and accompanying references.

1. Unlike many inorganic contaminants (e.g. arsenic or fluoride), nitrate rarely occurs as a natural constituent in groundwater from geo-genic sources (although naturally elevated levels can occur in desert areas, due to fixation by arid-zone plants) (Heaton, 1986).
2. It is one of the most readily analyzed contaminants and a large amount of data is available.
3. Nitrate has a range of agricultural, urban and industrial sources (e.g. fertilizers, sewage & animal waste, municipal, domestic and

industrial wastewater discharge), and it is highly soluble. Therefore, it is likely to be one of the most common (if not the most common) contaminants, serving as an index pollutant for many different pollution sources, mechanisms and processes.

Nitrate is also an important contaminant from a human health perspective, as it has been linked to chronic illness of the digestive system and increased incidence of digestive cancers, which are widespread in parts of China with severe water pollution (World

Table 2
Surface water quality standard 6-class rating system.

Grade	Classification/applicable uses
I	Pristine water sources (e.g. river headwaters and protected natural catchment areas)
II	Class A water source protection areas for centralized drinking supply
III	Class B water source protection areas for drinking supply and recreation
IV	Industrial water supply and recreational water with no direct human contact
V	Limited agricultural water supply
VI	Essentially useless

Source: Ministry of Environmental Protection (2002).
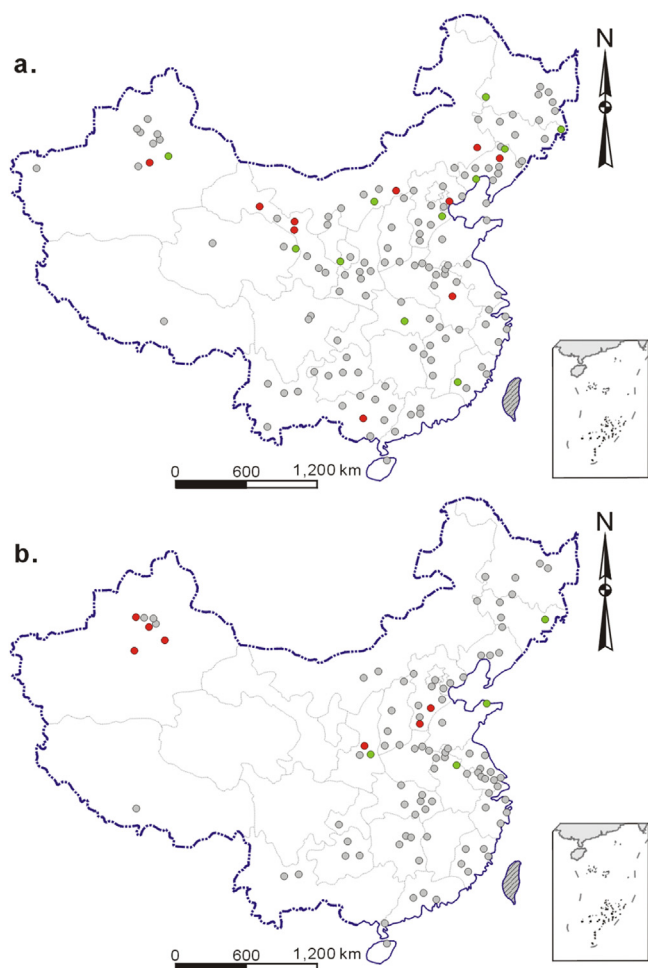
Fig. 3. Distribution of monitored sites where changes in water quality grade occurred between 2008 and 2009 for a) shallow groundwater and b) deep groundwater. Green = grade improved; grey = grade stable; red = grade deteriorated. Modified from Ministry of Land and Resources (2010). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Bank and State Environmental Protection Administration, 2007; Ebenstein, 2012).

Boxplots of NO_3–N data from the groundwater basins across China are shown in Fig. 2 along with the US EPA maximum contaminant level (MCL) of 10 mg/L, which is approximately equivalent to the World Health Organization standard of 50 mg/L nitrate as NO_3. According to the data, nitrate contamination is ubiquitous in China's groundwater. The MCL level is exceeded by the 90th percentile NO_3–N value observed in the majority of China's shallow groundwater basins (25 out of 36 study areas) as

well as many deep and karst aquifers (10 out of 37 study areas). The MCL is also exceeded by the median nitrate-N concentration in five shallow groundwater systems and four deep groundwater systems (all in northern China). The worst affected region is the coastal area adjoining the Bohai Sea, including Dalian (DAL); Laizhou Bay (LZB) and Qinhuangdao (QHD) (Fig. 2). All of these sub-areas showed median nitrate concentrations above the MCL, with the highest median being 55 mg/L NO_3–N at Dalian (5.5 times the MCL). This area is where a number of northern China's rivers drain to the ocean, and where regional groundwater flows converge and discharge at the coast. Areas of karst groundwater are highlighted in blue on Fig. 2c. In these aquifers, it is known that circulation times are relatively short (due to many preferential flow pathways) and thus shallow and deep groundwater systems are typically in rapid connection. Most of the karst systems show nitrate levels with median values below the MCL (Fig. 2c).

As is typically expected where nitrate is derived from surface sources such as agriculture, sewage or wastewater discharge, nitrate pollution is more serious in shallow groundwater (NO_3–N concentration ranges from 0.1 to 1819.5 mg/L, n = 627, median value = 8.0) compared to deep groundwater (NO_3–N concentration ranges from 0.1 to 90.3 mg/L, n = 118, median value = 2.6) (Table S1). However, numerous samples from deep aquifers also show levels of nitrate that indicate anthropogenic contamination, including many samples above the MCL (Fig. 2c).

Figs. 4 and 5 show compiled isotopic compositions of nitrate, including $\delta^{15}N_{NO3}$ and $\delta^{18}O_{NO3}$, for groundwater in ten major basins for which data were available (the raw data are contained in Table S1). Overall, the values range from −14.9 to 35.5‰ (n = 595, mean = 9.5‰, median = 8.6‰) for $\delta^{15}N_{NO3}$ and −8.1–51.0‰ (n = 255, mean = 10.2‰, median = 8.2‰) for $\delta^{18}O_{NO3}$ in shallow groundwater. In deep groundwater, the isotopic values range from −8.0 to 14.4‰ (n = 89, mean = 6.7‰, median = 6.8‰) for $\delta^{15}N_{NO3}$ and −2.3–39.6‰ (n = 67, mean = 10.1‰, median = 7.5‰) for $\delta^{18}O_{NO3}$. As shown in Fig. 5 the $\delta^{15}N_{NO3}$ values are generally lower in samples from the northwest of China and lower Yangtze River basin, while $\delta^{18}O_{NO3}$ values show similar ranges (with medians between 6.2 and 9.0‰) in the majority of basins, except for the northwest, which showed a higher median value (25.9‰). The wide ranges in observed isotopic values of nitrate indicate that nitrate in China's groundwater derives from multiple sources as opposed to one single, dominant source.

4. Discussion

4.1. Major sources of contamination and mechanisms of contaminant spread

Major sources of surface water and groundwater contamination in China include un-regulated sewage and municipal wastewater discharge, industrial wastewater discharge and agricultural fertilizers and pesticides (World Bank and State Environment Protection Agency, 2007; Liu and Yang, 2012; Tao and Xin, 2014). Comparisons of pollution load by source indicate that on aggregate, agricultural sector pollution outweighs urban and industrial sources in terms of major water quality indicators such as nitrogen (as nitrate, nitrite and ammonia) and phosphorus (Watts, 2010). However, both sectors are significant contributors to water pollution, and regional variation in the dominant source depends on the intensity of each sector and the degree of local pollutant discharge regulation. The widespread nitrate contamination documented in Fig. 2 is in large part related to agricultural non-point-source pollution, as has been documented in previous studies (e.g. Chen et al., 2005; Ju et al., 2007). Areas such as the northwest of China and North China Plain are intensively cultivated, and nitrate contamination from
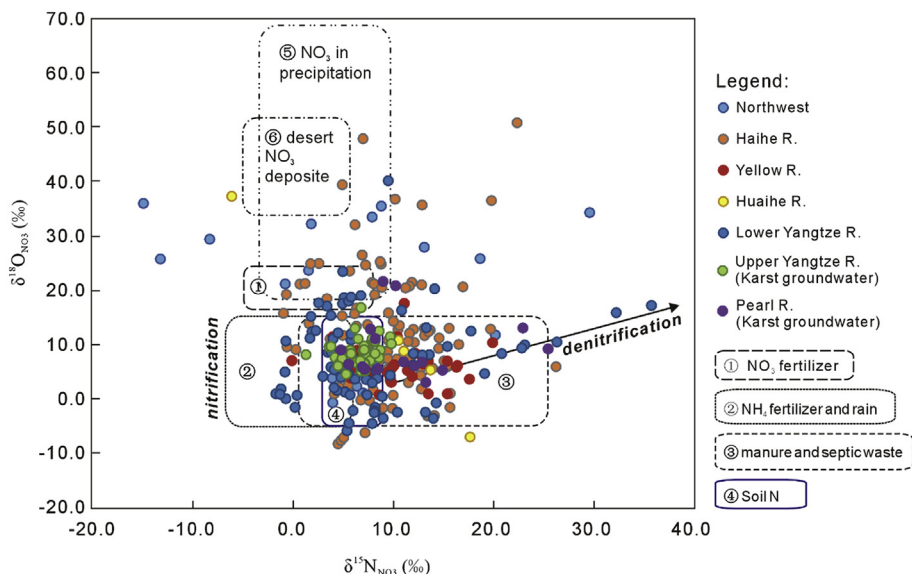
Fig. 4. $\delta^{15}N_{NO3}$ and $\delta^{18}O_{NO3}$ values of NO_3^- in groundwater samples from different watersheds.
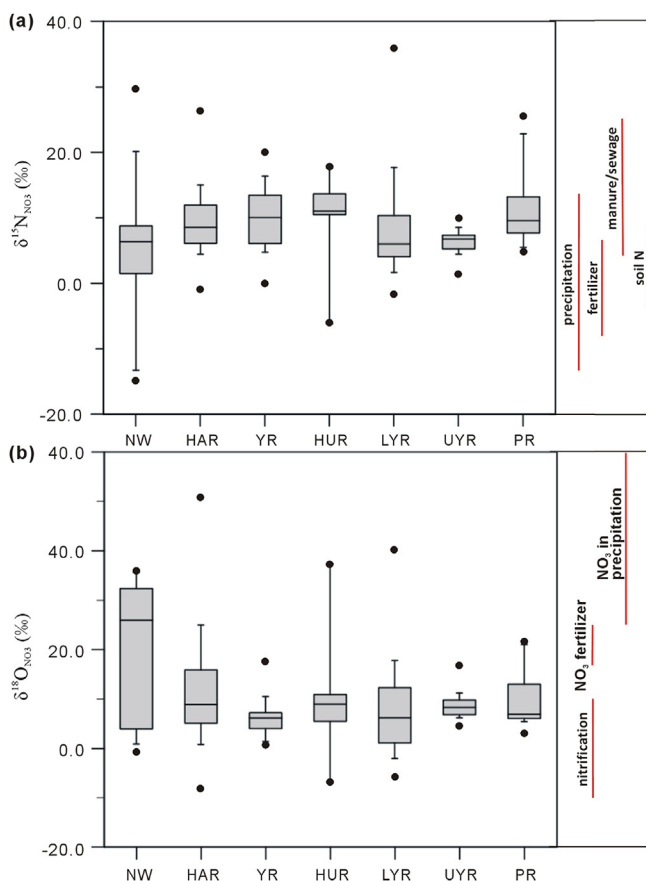

Fig. 5. Compositions of nitrogen and oxygen isotopes of nitrate in groundwater from different basins in China. The lower and upper edges of the box represent 25 and 75 percentiles of all data; the lower and upper bars represent 10 and 90 percentiles of all data; the dots outside of the box and bars represent the maximum and minimum values. NW- northwest basins (numbers of water samples: n = 17); HAR- Haihe River basin (n = 120); YR- Yellow River basin (n = 38); HUR- Huaihe River basin (n = 5); LYR-lower reach of Yangtze River basin (n = 80); UYR-upper reach of Yangtze River basin (Southwest karst groundwater) (n = 44); PR- Pearl River (Southeast karst ground-water) (n = 16). Datasets are included in Table S1. The range in isotopic values attributed to different sources are derived from Jin et al. (2015) and references therein.

excess fertilizer appears to be one of the dominant sources in these areas. For example, in northwest China, the $\delta^{15}N_{NO3}$ values group near 0‰, and $\delta^{18}O_{NO3}$ values are between 0 and 25‰ (e.g. Fig. 5 and Supplementary Fig. S2). Nitrate in agricultural regions with higher $\delta^{15}N_{NO3}$ values (e.g. up to 10 or 15‰) may also be derived from excess fertilizer, with subsequent fractionation during de-nitrification having enriched the residual ^{15}N in the pool (Fig. 4; Mayer et al., 2002; Kendall et al., 2007). Over-usage of fertilizer and pesticide in China is well known, with average per unit area ap-plications on farmland 2.8 times and 3 times the world average, respectively (Ministry of Environmental Protection, 2011). Miner-alization of organic nitrate due to soil cultivation is also a well-documented process (e.g. Heaton, 1986), and many of the sam-ples compiled show isotopic values consistent with this source (Fig. 4). This is likely a result of the intensive cultivation practices which have emerged in China over recent decades.
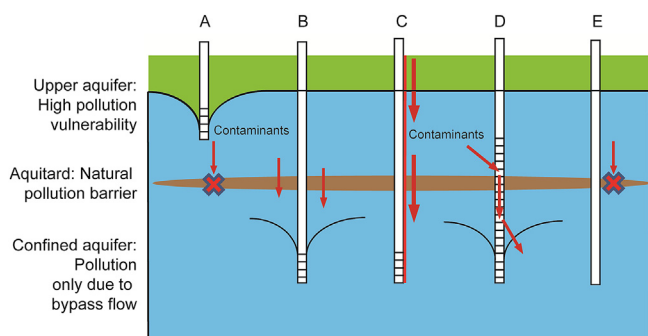
It also appears that septic or other sewage waste and/or animal manure are important sources of nitrate in the groundwater in many regions. Areas such as the Haihe River basin, Huaihe River basin and Pearl River Delta are heavily urbanized, and wastewater discharge to surface water is a well-documented problem (World Bank and State Environment Protection Agency, 2007; Yang and Zhuang, 2014). The nitrate isotope data in these three areas (me-dian $\delta^{15}N_{NO3}$ values of 8.6, 8.4 and 9.6‰, respectively) and possibly the Yellow River Basin near the city of Xi'an (median of 10.1‰) are all consistent with wastewater being a primary source (Fig. 5; Table S1).

Nitrate contamination shows a predictable trend of decreasing concentration with increasing sample depth, consistent with sur-face sources being attenuated during downward transport (by dilution and/or de-nitrification). Nitrate concentrations have also been shown in some studies to correlate with tritium contents in groundwater – an indicator of recent recharge (Liu and Chen, 2009; Han et al., 2011). These trends indicate that shallow aquifers are particularly vulnerable to be polluted by surface activities. A basin-scale process impacting groundwater quality in China has been the recent switch in the predominant recharge source from natural infiltration of rainfall and surface water to return-flow from agri-cultural irrigation (O'Dochartaigh et al., 2010; Currell et al., 2012). The process is due to a combination of extensive dam building in mountainous areas, which has prevented natural flooding over the

most productive groundwater recharge areas, along with intensive over-irrigation in agriculture. Irrigation return flow is generally high in salts, nitrate and agricultural chemicals, and so this switch in recharge mechanism has resulted in major diffuse groundwater pollution (Ju et al., 2007). This is a global problem in semi-arid regions (Scanlon et al., 2007), but it is particularly serious in China (Currell et al., 2012); the nitrate data in this study provide further evidence that the process is widespread. Our data also show that wastewater discharge is an important source of nitrate pollution in groundwater. Due to intensive pumping of groundwater, many surface water systems in northern China have become 'losing' (Cao et al., 2016) and thus waterways impacted by wastewater discharge can readily act as pollution sources to the underlying aquifers.

The data in Fig. 2 also reveal extensive contamination of deep groundwater by nitrate, particularly in the northern basins (Fig. 2c). This indicates that a pathway must exist which allows contaminants from the surface to reach deep aquifers (including those that are confined by low permeability aquitards). Groundwater age-dating studies carried out in the North China Plain and other basins indicate that much of the deep groundwater in confined systems was recharged 10s of thousands of years before the present, indicating circulation and replenishment on geological time-scales (Kreuzer et al., 2009; Currell et al., 2010, 2012). Therefore, the contamination of these deep aquifers by anthropogenic nitrate must be taking place due to 'short circuit' or 'bypass flow', such as rapid transport down faulty or abandoned wells, or those which are constructed with long open screened intervals, providing interconnection between shallow and deep groundwater bodies.

The boom in groundwater development in China over the past 3-4 decades has seen drilling of millions of wells across the nation — it is estimated that over 4.5 million wells have been drilled in northern China alone (Shen, 2015). Many of these wells are known to be screened across multiple aquifers (Wang et al., 2007). These wells create a large number of potential conduits between shallow and deeper aquifers (Fig. 6). This opens the possibility that deeper aquifers containing high quality water, which would naturally have much lower vulnerability to pollution from surface activities, may not be isolated from polluting surface activities (Fig. 6). Due to the general absence of well licensing and maintenance systems, the condition and integrity of all deep wells can't be guaranteed, and cross contamination is in some regards inevitable (Shen, 2015). This

type of short-circuiting is the only way to explain the observed high nitrate concentrations at hundreds of meters depth in some cases. Currell et al. (2010) documented groundwater samples confined groundwater in the Yuncheng Basin as deep as 180 m, with nitrate-N concentrations of more than 45 mg/L. The observation of tritium (an indicator of modern recharge) in selected samples of deep, nitrate contaminated water (Han et al., 2014) is further evidence of this 'short-circuiting' mechanism.

While the nitrate data do tend to confirm findings from previous assessments that water pollution from agriculture is widespread, urban sources are also significant drivers of water quality degradation in China. The Bulletin of the Ministry of Land and Resources data (Fig. 1b) are mostly sourced from wells in urban areas, and these generally show relatively high levels of ammonia, COD and organic contaminants, indicating predominantly industrial pollution sources. Unfortunately, detailed results of surveys of organic water pollutants and heavy metals in aquifers in China are rare; meaning detailed attribution of sources and mechanisms is more difficult than for agricultural pollution. However, a 2013 investigation revealed a large discrepancy (over 1×10^{11} tons) in the amount of water supplied to China's industries and the amounts of reported industrial wastewater discharge, leaving a large volume of potentially polluted water unaccounted for. This has led to the conclusion that industries have been engaging in illegal discharge of wastes to surface water through unmonitored discharge points, and groundwater, through secret disposal wells (Ministry of Environmental Protection, 2011). Further publication of site-specific data on pollutant concentrations in urban areas in future would greatly assist future pollution control and clean-up efforts.

The rapid urbanization currently occurring in China also should not be ignored in the discussion of pollution sources and mechanisms. Rapid urbanization is creating many new pollution challenges, such as the need to collect and treat increasing volumes of urban sewage, and the loss of arable land through urban expansion and soil pollution (Chen, 2007).

4.2. Implications and consequences of water pollution

The datasets compiled in this study, including government data on water quality classes at the river basin/regional level, groundwater nitrate data at the basin scale and nitrate isotopic data, show that groundwater pollution is ubiquitous throughout China, with no region unaffected. On the whole, the most serious contamination occurs in the north of China, where surface water pollution is also most severe (e.g. Fig. 1). It is widely recognized that the North China Plain, a focal point for both intensive agriculture and intensive heavy industry, is the location of China's biggest water and other environmental challenges (Shapiro, 2012). This is also the area where 'cancer villages' are most densely distributed in China (Table 1). Cancer villages are population centers where the incidence of cancer morbidity or mortality is significantly higher than national averages (Liu, 2010; Wan et al., 2011; Gong and Zhang, 2013; Yang and Zhuang, 2014). The first Chinese cancer village was documented in 1954, however by the end of 2011 this number had reached 351, including 186 arising between 2000 and 2009, causing some to call this China's 'decade of cancer' (Gong and Zhang, 2013). A seven-year study led by the former deputy director of China's Center for Disease Control and Prevention showed that residents of tributary areas of the Huaihe River basin died in significantly higher proportions due to digestive tract tumors than more distant control areas, and that rates of cancer morbidity rose from below national averages in 1973-74, to significantly higher than China's national average in the 2000s for liver, gastric and esophagal cancers (Yang and Zhuang, 2014). This agrees with previous epidemiological studies which have linked these cancer types

Fig. 6. Schematic diagram showing potential mechanisms of bypass flow leading to contamination of deep groundwater. Red arrows indicate transport of pollutants towards deep aquifer. A) Groundwater pumping only in shallow unconfined aquifer; low risk of pollution crossing aquitard; B) Intensive pumping of deep groundwater (from intact wells) results in slow leakage of shallow groundwater across aquitard; C) Faulty well annular sealing creates conduit for contaminants to bypass aquitard and pollute deep groundwater; D) Long-screened well allows contaminants to cross aquitard through the well; E) Cemented deep well prevents contamination crossing aquitard. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

to chronic exposure to water pollution (Ebenstein, 2012).

Perhaps the most significant issue illustrated by the data is the widespread pollution of groundwater in deep semi-confined and confined aquifers (Fig. 2). Contamination of these groundwater bodies is arguably more concerning than widespread shallow groundwater contamination, as the timescales of recharge and residence in deep aquifers are on the order of tens of thousands of years (for a compilation of groundwater ages determined using radiogenic isotope dating throughout major basins of northern China, see Currell et al., 2012). This means that there is limited capacity for natural flushing and dilution of contaminated groundwater on timescales of relevance to current human societies (e.g. Gleeson et al., 2016). Any future efforts to remediate these deep systems will therefore be costly, technically challenging and possibly of limited effectiveness. The Chinese government has also recently responded to concerns about high levels of shallow groundwater pollution in northern China by assuring the public that most drinking water supplies are derived from deep wells, which are naturally less vulnerable to contamination (Buckley and Piao, 2016). This indicates that 1) many people depend on deep groundwater for drinking water, and thus protection of its quality is critically important; 2) there is a belief that pollution pathways to these deep aquifers are limited. However, the data in this study indicate that this is not necessarily the case and that widespread pollution of deep aquifers is already underway. Given the evidence for bypass flow in the form of elevated nitrate in deep groundwater samples, it is likely that many other contaminants, including more recalcitrant and hazardous organic substances may have also been transported to deep levels, but that the available data is not yet detailed enough to document all such impacts. This is a major concern that should be addressed through targeted sampling of deep aquifers and analysis of a range of representative pollutants.

4.3. The 10-point water pollution action plan: the beginning of new era for China?

The *10-point Water Pollution Prevention and Control Action Plan of the Government of the People's Republic of China* was released in April of 2015 (Central People's Government of the People's Republic of China, 2015; Zheng, 2015). The major action points of the plan are:

1. Take control of and reduce pollutant discharge
2. Promote transformation of the economic structure (to lower pollution intensity)
3. Focus on protection of water resources through water saving
4. Strengthen support for science and technology
5. Allow market mechanisms to impact water and pollution levies
6. Strict environmental law enforcement & supervision
7. Strengthen the management of the overall hydrological cycle
8. Ensure security of water for ecological and environmental purposes
9. Confirm and implement the responsibilities for all parties involved
10. Strengthen public participation and social supervision

Major goals of this ten-point water plan are that by 2020, 70% of surface water quality in the seven major river basins will be of classes I — III, while no more than 15% of groundwater monitoring points will be of the lowest class (V). The plan has a focus on drinking water safety, and will ensure management and supervision of the water supply process 'from source to tap'. The national and regional water security situation for drinking water will be reported annually from 2016 onwards.

The plan introduces new incentives and penalties for water polluting industries, promising to periodically announce lists of poor performing enterprises, and giving out 'yellow cards' and 'red cards' based on annual performance. League tables of the worst and best performing 10 cities in each year will also be reported based on pollution control and water quality class improvements. A lack of positive or negative incentives for government officials to address water pollution has in the past been identified as a major barrier to the effective control of water pollution in China (Liu and Yang, 2012; Tao and Xin, 2014).

Several other aspects of the plan differ from previous approaches:

1. It lays stress on the philosophy of total water cycle health including groundwater, surface water and marine water, acknowledging their connections. It directly addresses seaport pollution control, which has been given less attention in past environmental policies.
2. It aims to give rise to the development of an extensive environmental protection industry and environmental protection services sector, creating a new area to boost sustainable economic growth. The implementation of the plan is expected to contribute to service sector growth as a share of GDP by 2.3%, including a cumulative increase in non-agricultural employment of 3.9 million people.
3. It clearly assigns monitoring and compliance responsibilities to particular agencies and responsible persons. This has been a major hurdle to successful water management policy in China in the past, as institutional overlap and lack of clarity on accountabilities have left many issues un-resolved (Liu and Yang, 2012; Liu, 2015; Shen, 2015). Under the new plan, agencies are being charged with particular responsibilities (Table 3) and will be held accountable through performance assessments.

The success of the plan will heavily depend on inter-agency co-operation and coordinated implementation at regional and local scales. The issue of non-cooperation between different agencies or levels of government and/or lack of clearly defined roles and responsibilities has hindered many previous attempts at improving environmental governance in China and it is still a major hurdle to effective groundwater management (Cai, 2008; Shen, 2015). This has caused cynicism among some Chinese citizens, who have coined sayings such as "The Ministry of Environmental Protection won't step into the water and the Ministry of Water Resources won't step ashore". Laws and regulations related to water resources have thus been characterized by weak enforcement and inconsequential penalties for non-compliance (Ma, 2004; Cai, 2008). On the other side, for many enterprises, costs of sewage treatment and pollution control technology have been prohibitive, and in combination with the weak regulatory regime, have fostered a culture which has long tolerated un-regulated discharge of pollution.

The 10-point plan does address these key issues to an extent, through setting new incentives and penalties for officials and industries and 'naming and shaming' through the league tables. However the detailed workings of the incentive and deterrence measures, and the relationship between these and existing and/or conflicting incentives and performance targets are complicated. It is therefore difficult to predict how implementation of the plan will unfold in the coming years. One certainty is that without clear, accurate and transparent data on the scale and extent of pollution and its sources, the co-ordination between agencies and control and clean-up efforts will prove difficult (Qin, 2016). While our data compilation is a preliminary attempt to synthesize disparate datasets on water pollution, there are still many data gaps.

One area of particular importance which we have identified and

Table 3
Government agencies primarily involved in water management in China and their roles under the 10-point water pollution action plan (Central People's Government of the People's Republic of China, 2015).

Agency name	Primary responsibilities	Leading role(s) under the 10-point water plan
Ministry of Housing and Urban-Rural Development	Municipal water supply, drainage, construction of sewage treatment plants	*Points 1 and 3 of the plan.* Strengthen urban pollution control, strengthen the construction of supporting pipe networks, promote sludge disposal and recycled water use, strengthen urban water saving
Ministry of Agriculture	Management of agricultural pollution (non-point source); fisheries & wildlife conservation	*Points 1, 3 and 8 of the plan.* Promote agricultural rural pollution prevention, control agricultural non-point source pollution, adjust the planting structure and layout, promote the ecological health of aquaculture, develop agricultural water-saving
Ministry of Water Resources	Protection of water resources; flood control and drought relief; water withdrawal licences; *trans*-boundary water conflicts	*Point 3 of the plan.* Monitor and oversee the total quantity of water, improve water use efficiency, protect water resources scientifically, strengthen the management of rivers and lakes, determine ecological flow limits, strict control on groundwater exploitation, pay special attention to industrial water-saving, develop water-saving agriculture
Ministry of Land and Resources	Monitoring and protection of groundwater resources	*Point 3 of the plan.* Actively protect ecological space, strict control of groundwater exploitation
Ministry of Environment Protection	Pollution control, water quality monitoring; wastewater discharge monitoring; environmental impact assessment	*Points 1, 2, 6, 7, 8, 9 and 10 of the plan.* Vigorously promote the prevention and control of industrial pollution, carry out a special program to control pollution from ten key industry types, centralized management of industrial agglomeration, speed up comprehensive improvement of the rural environment, strict environmental law enforcement, ensure the eco-hydrological environmental security, confirm and implement the responsibilities for all parties, strengthen public participation and social supervision.
National Development and Reform Commission	Governance of major river basins	*Points 2, 4 and 5 of the plan.* Promote sustainable seawater desalination, develop the environmental protection industry, set the price of taxes and fees, establish incentive mechanisms, speed up the development of environmental services, improve levy policies, examination of results of water pollution prevention and control for relevant capital allocation, promote pluralistic financing of improvement projects, lead optimization of land-use and water network layout
State Oceanic Administration	Supervision, monitoring and control of marine pollution	*Point 8 of the plan.* Strengthen the near-shore environmental protection, protect marine ecological health
Ministry of Transport	Marine pollution control (shipping)	*Point 1 of the plan.* Strengthen port pollution control and prevention
Ministry of Industry and Information Technology	Prevention and control of industrial water pollution	*Point 2 of the plan.* Adjust the industrial structure, promote exit of polluting enterprises, pay special attention to the industrial water-saving, lead optimizing land-use zoning & urban growth layout

which is currently not addressed in the 10-point plan is ensuring the integrity of water (and other) wells, so that these do not act as pollution pathways between shallow and deep aquifers. A well-licensing, maintenance and decommissioning program at the appropriate jurisdictional level(s) should thus be an urgent priority. As noted above, statements from the Ministry of Water Resources suggest that this agency believes that sourcing drinking water supplies from deep aquifers ensures that these water supplies are isolated from surface pollution impacts; yet the data we have compiled indicate that this is not always the case, and that urgent action to cut pollution pathways is needed.

5. Conclusions and recommendations

A review and compilation of data on the water quality classes in groundwater and surface water at the regional and national scale shows that China faces a huge water pollution challenge in all parts of the country. As yet, the publicly available government data on water quality consists of aggregated regional indices, as opposed to detailed data on individual pollutants at the catchment or basin scale. Our compilation of groundwater nitrate data from the research literature for the first time enables a more detailed national assessment of the degree of water pollution. In particular, it highlights the issue of pollution of deep aquifers that are generally thought to be relatively isolated from the surficial hydrological cycle.

China's new 10-point Water Plan offers hope that China has reached a turning point with respect to water pollution. Based on

our analysis of the groundwater and surface water pollution data, and a reading of the 10-point plan in this context, we offer the following recommendations:

1. Apart from controlling pollution sources and reducing discharge of wastewater to streams and shallow aquifers (a focus of the plan), effort needs to be concentrated on the problem of groundwater well integrity, to protect deep groundwater aquifers from further pollution. Our analysis indicates by-pass flow from poorly constructed wells has caused widespread contamination of deep groundwater with nitrate, and therefore probably other contaminants. A program of well cementing and decommissioning aimed at cutting these pathways, and new standards for well licensing and construction are vital, as deep aquifers contain particularly high quality water and are almost impossible to remediate once contaminated.

2. Our analysis confirms that non-point-source pollution (with nitrate and related compounds) due to agriculture is a major issue affecting groundwater quality nation-wide (as well as wastewater discharge). There still exists no quantifiable standard in China to control non-point source pollution from fertilizers. A further step should thus be to establish standards for improving farming practices to effectively control application of chemical fertilizers and pesticides.

3. Water quality monitoring and data disclosure, particularly for groundwater, should be improved. To date, government agencies have only reported spatially aggregated data (rather than specific monitoring points or even catchments/sub-

catchments). This hampers efforts to encourage public participation in local pollution control and remediation action.

4.Due to increasingly complex arrays of pollutants being detected in recent years, the standards for surface- and groundwater quality should be updated. The quality standard for groundwater (GB/T 14848-93) is more than 20-years old and should be amended to include additional contaminants of potential concern such as persistent organic pollutants known to pose significant health and ecological risks (e.g., polycyclic aromatic hydrocarbons, polychlorinated biphenyls and organochlorine pesticides) as well as 'emerging' groundwater contaminants which are particularly important in urban and industrial areas − such as pharmaceuticals and their residues (e.g. Lapworth et al., 2012; Lapworth et al., 2015).

China's war on water pollution has just begun, and it will be fight that will take decades. If China can learn from international experiences, and build on the current momentum and political will, we believe great progress can be made under the new 10-point plan.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.envpol.2016.08.078.

The typical ranges of isotopic values for various sources are from Kendall and McDonnell (1998).

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