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# Using magnetic susceptibility to evaluate pollution status of the sediment for a typical reservoir in northwestern China

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#### Abstract

Intensive anthropogenic activities may add to pollution risks to lakes and rivers, which can be revealed by the magnetic characteristics of sediments. The present study aims to assess the pollution status of the sediment of a typical reservoir in northwestern China by application of magnetic susceptibility. The values of magnetic susceptibility exhibited significant positive correlations with trace metals (Co, Cu, Mn, Ni, and V) and natural radionuclides  $(^{232}Th$  and  $^{40}K$ ). Multivariate statistical analysis indicated common sources and similar deposition characteristics of magnetic particles and trace metals. It was conformed that magnetic susceptibility could be used as an indicator to identify industrial sources of trace metals, but was not suitable to indicate the traffic or natural sources. Linear regression equations between the low-frequency magnetic susceptibility and the integrated pollution index as well as annual effective dose rate indicated a potential for using magnetic susceptibility in semi-quantitative assessment of trace metal pollution and radiological hazard in sediments. A three-step procedure is proposed for the use of magnetic susceptibility in pollution monitoring, which provides a fast and effective method for estimating the pollution extent and tracing the major sources of trace metals in the sediment of lakes and rivers.

Keywords Trace metal . Natural radionuclide . Pollution indicator . Source discrimination . Environmental magnetism method

# Introduction

Sediment is an important environmental medium in aquatic ecosystem, which is considered as the environmental host of the waste discharged by natural and artificial processes (El-Taher and Madkour [2011](#page-11-0)). Numerous harmful contaminants such as metals and radionuclides are detected in sediments, which constitutes a very useful tool for the water environment quality assessment (Szarlowicz et al. [2013](#page-12-0); Erenturk et al. [2014\)](#page-11-0). Trace metals including heavy metals and metalloid,

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such as nickel (Ni), copper (Cu), zinc (Zn), cadmium (Cd), lead (Pb), chromium (Cr), and arsenic (As), are the most frequently detected contaminants in sediments, which have adverse effects on aquatic organisms and account for potential health risks for human being (Suresh et al. [2012](#page-12-0); Acosta et al. [2015;](#page-10-0) Trujillo-González et al. [2016](#page-12-0)). These elements are precipitated or adsorbed on particulate material and accumulate in sediments and may influence the aquatic environment during the next few decades or even centuries because they are non-biodegradable (Doichinova et al. [2014,](#page-11-0) Peng et al. [2017\)](#page-12-0).

Human activities cause obvious enrichment of trace metals in the sediment, such as vehicle traffic, industrial and agricultural activities, and mining activities (Jordanova et al. [2013\)](#page-11-0). Therefore, much concern has been attracted to the monitoring and assessment of pollution status of the sediment (Erenturk et al. [2014,](#page-11-0) Jordanova et al. [2004](#page-11-0), Prajith et al. [2015](#page-12-0)). Geochemical methods are generally applied to monitor the occurrences of metals in sediments (Jordanova et al. [2013\)](#page-11-0). Considering the sampling procedures before analysis, the whole process is usually expensive and time consuming. Therefore, geophysical technologies which are generally time- and cost-efficient have been proposed to identify the occurrences and sources, and evaluate the risks of trace metals

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in soils and sediments (Hay et al. [1997;](#page-11-0) Strzyszcz and Magiera [1998;](#page-12-0) Lu et al. [2007;](#page-12-0) Wang et al. [2017](#page-13-0)).

Previous studies have acknowledged correlations between trace metal content and magnetic properties, and proposed that some magnetic properties might be used as a proxy for metal contents (Chen et al. [2013;](#page-11-0) Liu et al. [2013](#page-12-0); Dong et al. [2014](#page-11-0); Chaparro et al. [2017](#page-11-0)). In these methods, magnetic susceptibility has been widely applied as approximation of trace metal contamination because it is an easily measurable and concentration-dependent geophysical parameter (Bityukova et al. [1999;](#page-11-0) Canbay et al. [2010](#page-11-0); D'Emilio et al. [2012;](#page-11-0) Zhang et al. [2013](#page-13-0)). Furthermore, this method is especially useful in characterizing and screening of technogenic magnetic particles released by industrial and urban dust deposition derived from high-temperature technological processes (Magiera et al. [2011](#page-12-0); Basavaiah et al. [2012](#page-11-0); Rachwał et al. [2017\)](#page-12-0). Enrichment in trace metals identified by high magnetic susceptibility values of soils and sediments was observed in the surroundings of various industrial plants like coke plants, power plants, iron and steel works, and nonferrous metallurgical works (Lu et al. [2007](#page-12-0); Zhang et al. [2011;](#page-13-0) Liu et al. [2013;](#page-12-0) Rachwał et al. [2015](#page-12-0)). Therefore, magnetic measurements combined with geochemical analysis proved to be very helpful in identifying the trace metal pollution and tracing the sources of pollutants in soils and sediments (Wang et al. [2013;](#page-12-0) Szuszkiewicz et al. [2016\)](#page-12-0).

Meanwhile, natural radionuclides such as  ${}^{40}$ K,  ${}^{238}$ U, and <sup>232</sup>Th are typically presented in sediments, which might lead to extra human exposure to radiation when the sediments are used as building material (Baeza et al. [2009](#page-10-0); Yii et al. [2009](#page-13-0); El-Taher and Madkour [2011](#page-11-0)). Significant correlations between magnetic susceptibility and the concentrations of  $232$ Th,  $238$ U, and  $40$ K have been reported (Ramasamy et al. [2014;](#page-12-0) Chaparro et al. [2017](#page-11-0)). Therefore, magnetism methods may also be used as additional tools in the assessment of radioactivity levels of sediment (Erenturk et al. [2014](#page-11-0)).

Wangjiaya reservoir is an important part of the Baoji Gorge Weibei Irrigation Project, which pumps water to supplement the water shortage of Baoji Gorge Irrigation Area and supplies artesian water to the original irrigation area. With long-term and large-scale industrial activities as well as rapid urbanization in Baoji city, the environmental quality of the reservoir has been potentially affected. In 2003, the establishment of Changqing industrial park upstream greatly increased the environmental risk to the reservoir. Since the intensity of agricultural practices and industrialization are considered to be the main sources of radioactivity and heavy metals in rivers (Saç et al. [2012](#page-12-0); Faweya et al. [2013\)](#page-11-0), considerable increase in radioactivity and trace metal levels of the sediment in this reservoir is expected (Krmar et al. [2009](#page-12-0); Chaparro et al. [2017\)](#page-11-0). Therefore, Wangjiaya reservoir represents an interesting case study for environmental contamination and ecological deterioration disturbed by intense anthropogenic activities.

The contamination levels of trace metals and natural radionuclides in sediments of this reservoir have already been reported in our previous studies (Lu et al. [2016;](#page-12-0) Zhu et al. [2016\)](#page-13-0). The results indicated potential ecological risks of anthropogenic pollutants in the reservoir (Shi et al. [2018](#page-12-0)). In this study, the magnetic susceptibility of the sediment in Wangjiaya reservoir was investigated. We focus on the study of the relationship between the magnetic susceptibility and the levels of trace metals and natural radionuclides. The main aim is to evaluate the applicability of using the magnetic susceptibility characteristics as environmental pollution proxies concerning the influence of pollution sources. In particular, this method will be available for rapid discriminating of hotspots with high pollution risks and for tracing the anthropogenic pollution sources in the sediment of lakes and rivers.

## Materials and methods

## Background of study area

The investigated area, Wangjiaya reservoir, is an artificial lake located in about 20 km northwest of Baoji city, NW China (Fig. [1\)](#page-2-0). The Qian River enters the reservoir from the north. The western and eastern parts of the reservoir are surrounded by farmlands, uncultivated lands, villages, and the main trunk of the traffic. The Changqing industrial park with zinc smelting, ironworks, coking, and coal-fired power generation is situated on the northeast side (Fig. [1\)](#page-2-0). Detailed information about the study area can be found in our previous studies (Lu et al. [2016](#page-12-0); Zhu et al. [2016;](#page-13-0) Shi et al. [2018\)](#page-12-0).

## Sampling and preparation

Sediment samples were collected in 34 sites of Wangjiaya reservoir (Fig. [1](#page-2-0)). At every sampling site, a surface sediment sample (0–20 cm) was collected by using a grab sampler and the longitudes and latitudes of each sampling site were recorded by a hand-held global positioning system (GPS) instrument. An uncontaminated sediment sample was collected from the upstream of Qian River as the reference in the study area for background study.

All samples were dried, crushed, and sieved through a 1.0 mm nylon sieve to remove stones, coarse debris, dead organisms, etc. Approximate 50 g sediment sample was separated and ground with vibration mill, and then sieved through a 0.075-mm nylon mesh for measuring metal concentration. Additionally, one half of the preliminarily sieved sediment samples were dried at 110 °C for 36–48 h in an oven and ground and then sieved through a 0.16-mm mesh. The homogenized samples were weighted and then packed in a gas-tight, radon impermeable, cylindrical polyethylene plastic container (7.0-cm height and 6.5-cm diameter). These containers are

<span id="page-2-0"></span>

Fig. 1 Study area and the location of sampling sites in Wangjiaya reservoir. CFPP coal-fired power plant, ZSP zinc smelting plant

hermetically shielded and also shielded externally to ensure that all of the daughter products of uranium and thorium, radon isotope  $(^{222}$ Rn and  $^{220}$ Rn) formed in particular, do not escape (Krishnamoorthy et al. [2014\)](#page-12-0). The sealed samples were stored for about 30 days to allow <sup>226</sup>Ra and <sup>224</sup>Ra to reach the secular equilibrium with their short-lived decay products (Alencar and Freitas [2005;](#page-10-0) Lu et al. [2014a\)](#page-12-0). After that period, the sediment samples were analyzed for natural radionuclide concentration using gamma ray spectrometry.

## Experimental methods

For measuring metal concentration, 4.0 g of milled sediment sample and 2.0 g of boric acid were put into the pressureinstrument (YYJ–260) mold and crushed into a 32 mm diameter pellet under 30 t pressures. The contents of Co, Cr, Cu, Mn, Ni, Pb, V, and Zn in sediment samples were determined by wavelength dispersive X-ray fluorescence spectrometry (XRF, PANalytical PW–2403 apparatus) (Lu et al. [2010,](#page-12-0) [2014b](#page-12-0), [2014c;](#page-12-0) Zhang et al. [2015](#page-13-0)). Duplicate samples and standard materials (GSS–1 and GSD–12) were used for quality control in the experiment, and the relative errors were less than 5% (Lu et al. [2014b,](#page-12-0) [2014c\)](#page-12-0).

The concentrations of natural radionuclides  $^{226}$ Ra,  $^{232}$ Th, and  $^{40}$ K in the sediment samples were determined by using a 3  $\times$  3-in. NaI (Tl) gamma ray spectrometric system with excel 8% energy resolution  $(^{137}Cs$  661.6 keV). The detector, housed in a lead cylindrical shield with a thickness of 10.5 cm and a height of 38 cm, was coupled to a 1024 microcomputer multi-channel pulse height analyzer and the system was calibrated for the  $\gamma$ energy range 50 keV to 3.2 MeV (Lu et al. [2012](#page-12-0)). The activity of <sup>232</sup>Th was determined by 238.6 keV and 2614 keV gamma rays emitted from <sup>212</sup>Pb and <sup>208</sup>Tl, respectively. The activity of <sup>226</sup>Ra was measured by 609.3 keV and 1764.5 keV gamma rays emitted from <sup>214</sup>Bi, and <sup>40</sup>K activity was measured directly through its gamma ray energy peak of 1460.8 keV (El-Taher et al. [2010\)](#page-11-0). All samples were counted for 18,000 s (Yang et al. [2013\)](#page-13-0).

The low- and high-frequency magnetic susceptibility (lowfrequency magnetic susceptibility  $\chi_{LF}$  and high-frequency magnetic susceptibility  $\chi$ <sub>HF</sub>) was measured on 10 g samples by using a Bartington MS2 Magnetic Susceptibility Meter at frequencies of 0.47 and 4.7 kHz (Wang et al. [2015\)](#page-12-0). The frequencydependent magnetic susceptibility ( $\chi_{FD}$ %) was defined as

$$
\chi_{FD}(\%) = \frac{\chi_{LF}^-\chi_{HF}}{\chi_{LF}} \times 100\%.
$$

# Assessment of trace metal contamination and radiological hazard

To explore the indicative function of magnetic susceptibility to trace metal contamination and radiological hazard, the pollution indexes of trace metals (single pollution index (PI) and integrated pollution index (IPI)) and the hazard index of radionuclides (annual effective dose rate (AED)) were calculated to assess the extents of trace metal contamination and radiological hazard. The PI was defined as the ratio of element concentration to the background content of the corresponding element (the control sample in this study). The PI of each element was classified as either low (PI  $\leq$  1), middle (1 < PI  $\leq$ 3), or high (PI > 3). The IPI of all measured elements for each sample was defined as the average value of the element's P and was then classified as low (IPI  $\leq$  1), middle (1 < IPI  $\leq$  2), or high (IPI > 2) (Chen et al.  $2005$ ; Lu et al.  $2009$ ). The AED of each sediment sample was calculated to evaluate the radiological hazard due to gamma ray emission from  $^{232}$ Th,  $^{226}$ Ra, and 40K (Gonzalez-Fernandez et al. [2012;](#page-11-0) Saç et al. [2012](#page-12-0); Faweya et al. [2013;](#page-11-0) Lu et al. [2014a](#page-12-0)).

Radium equivalent activity  $(Ra_{eq})$  is defined according to the estimation that 1 Bq kg<sup>-1</sup> of <sup>226</sup>Ra, 0.7 Bq kg<sup>-1</sup> of <sup>232</sup>Th, and 13 Bq  $kg^{-1}$  of <sup>40</sup>K produce the same gamma radiation dose rate and it is calculated through the following relation:

$$
Ra_{eq}=C_{Ra}+1.43C_{Th}+0.07C_K
$$

In this equation, the  $C_{\text{Ra}}$ ,  $C_{\text{Th}}$ , and  $C_{\text{K}}$  are the activity concentrations of <sup>226</sup>Ra, <sup>232</sup>Th, and <sup>40</sup>K in Bq kg<sup>-1</sup>, respectively. To avoid radiation hazards, materials whose  $Ra_{eq}$  is greater than 370 Bq  $kg^{-1}$  should not be used for residential building constructions (Faweya et al. [2013](#page-11-0)).

Health effects of exposure to radiation are not determined by just the specific activity of the radiation source but also by the radiation energy and the dose absorbed by individuals (Faweya et al.  $2013$ ). Indoor air absorbed dose rate  $(D_{in})$  due to the gamma radiation from activity concentrations of natural radionuclides was calculated by the following equation:

$$
D_{in}=0.908C_{Ra}+1.06C_{Th}+0.0767C_{K}\\
$$

where  $C_{\text{Ra}}$ ,  $C_{\text{Th}}$ , and  $C_K$  are the activity concentrations of <sup>226</sup>Ra, <sup>232</sup>Th, and <sup>40</sup>K in Bq kg<sup>-1</sup>, respectively.

The annual effective dose rate (AED) was calculated from the following equation:

$$
E = T \times Q \times D_{\text{in}} \times 10^{-6}
$$

where  $T$  is the time for 1 year and  $Q$  is the conversion factor of  $0.7 \text{ Sv } \text{Gy}^{-1}$ , which converts the absorbed dose rate in air into human-effective dose received, while indoor occupancy factor was given as 0.8 (because most people spend a high percentage of their time indoors) (Faweya et al. [2013\)](#page-11-0).

## Multivariate statistical methods

Multivariate techniques were widely used to study the association between trace metals and magnetic variables (Lu et al. [2010;](#page-12-0) Chaparro et al. [2011;](#page-11-0) Chaparro et al. [2012](#page-11-0)). Pearson correlation analysis, cluster analysis (CA), principal component analysis (PCA), and K-means cluster analysis were conducted in this study. Pearson correlation analysis was performed to detect the relationships between magnetic susceptibility, trace metals, and radionuclides. The methods of CA and PCA were applied to discriminate the potential sources of the pollutants in the sediments by defining geochemical groups with approximately the same geochemical pattern (Gallego et al. [2002;](#page-11-0) Wang et al. [2015](#page-12-0)). K-means cluster analysis was used to recognize the difference of sampling sites affected by different sources. All statistical analyses were performed by using SPSS 20.0 and SAS JMP Statistical 13.0.

# Results and discussion

## Distribution characteristics of magnetic susceptibility

The statistic values of mass-specific magnetic susceptibility in sediments from Wangjiaya reservoir are presented in Table 1. The low-frequency magnetic susceptibility  $(\chi_{LF})$  varies from 49.9 to  $102.0 \times 10^{-8}$  m<sup>3</sup> kg<sup>-1</sup> with a median of 91.2 ×  $10^{-8}$  m<sup>3</sup> kg<sup>-1</sup> and high-frequency magnetic susceptibility ( $\chi$ <sub>HF</sub>) varies from 47.0 to 92.7 × 10<sup>-8</sup> m<sup>3</sup> kg<sup>-1</sup> with a median of  $82.4 \times 10^{-8}$  m<sup>3</sup> kg<sup>-1</sup>, respectively. Although the median values of  $\chi$ <sub>LF</sub> and  $\chi$ <sub>HF</sub> are very close to those in the control sample (91.7 and  $83.8 \times 10^{-8}$  m<sup>3</sup> kg<sup>-1</sup>), the 50% for  $\chi$ <sub>LF</sub> and 44% for  $\chi$ <sub>HF</sub> of the sampling sites are higher than those of the control sample. The relatively high  $\chi$ <sub>LF</sub> values are frequently distributed in both sides of the reservoir middle and upper

Table 1 Descriptive statistics of magnetic susceptibility (× 10−<sup>8</sup> m3 kg−<sup>1</sup> ) in the sediments of Wangjiaya reservoir

	$\chi$ LF	XHF	XFD%
Min	49.9	47.0	3.3
Max	102.0	92.7	11.1
Mean	87.1	79.3	8.8
Median	91.2	82.4	9.1
<b>SD</b>	13.4	11.5	1.4
$CV(\%)$	15.4	14.5	16.0
Kurtosis	1.88	1.80	6.33
<b>Skewness</b>	$-1.56$	$-1.53$	$-2.02$
Control sample	91.7	83.8	8.6

Notes: Min, minimum; Max, maximum; Mean, arithmetic mean; SD, standard deviation;  $CV(\%)$ , coefficient of variance;  $\chi_{LF}$ , low-frequency magnetic susceptibility;  $\chi_{HF}$ , high-frequency magnetic susceptibility;  $\chi_{FD}\%$ , frequency-dependent magnetic susceptibility

reaches, such as S23, S24, S25, S26, S27, S28, S29, S30, S31, and S32 (Fig. 2a). The previous study suggested that the magnetic susceptibility of the sediment was controlled by anthropogenic sources like emission from industrial and fossil fuel combustion, particles from vehicles, fertilizers, pesticides, etc. (Yang et al. [2007\)](#page-13-0). Additionally, the magnetic susceptibility enhancement in these sediment samples might greatly indicate the anthropogenic input from long-distant sources, including industrial sources in Changqing industrial park and traffic sources (Chlupáčová et al. [2010;](#page-11-0) Rachwał et al. [2017\)](#page-12-0).

The calculated frequency-dependence magnetic susceptibility ( $\chi_{FD}$ %) ranges from 3.3 to 11.0% with a median of 9.1%. About 65% of sampling sites show higher  $\chi_{FD}$ % than that of the control sample (8.6%), which are generally distributed in the middle to downstream of the reservoir (Fig. 2b). Frequency-dependence magnetic susceptibility is considered to reflect the presence of ultra-fine super paramagnetic ferrimagnetic grains particularly those grains between 0.010 and  $0.025 \mu m$  (Shilton et al. [2005](#page-12-0)). Thus, the relatively higher  $\chi_{FD}$ % values indicate the obvious contribution of fine magnetic particles and the existence of super paramagnetic (SP) grains in the sediment (Wang et al. [2015](#page-12-0)), which is probably due to anthropogenic activities (Ng et al. [2003;](#page-12-0) Chlupáčová et al. [2010](#page-11-0)). Through field investigation, in addition to the industrial emission from the Changqing industrial park upstream, vehicular traffic, agriculture sewage discharge, and domestic sewage discharge from surrounding villages are potential origins of target pollutants in the reservoir.

## Distribution patterns and pollution assessment of trace metals

Trace metal concentrations in the sediments of Wangjiaya reservoir have been reported in the previous study (Shi et al. [2018\)](#page-12-0). According to the classification reported by Wilding [\(1985\)](#page-13-0), the seven elements (Co, Cr, Cu, Mn, Ni, Pb, and V) show low variability ( $CV \leq 15\%$ ), and Zn shows a high variability  $(CV > 36\%)$  in spatial distribution in this study. The high CV of Zn might be due to the extremely high values of Zn in the northeast bank, which indicates potential pollution sources around the area. To illustrate the spatial distribution of these metals, the concentrations of individual metal and the reference levels of these trace metals in each sampling site are illustrated in Fig. [3](#page-5-0).

Generally, metal concentrations in the middle of the reservoir (S1–S18) record relatively higher values compared to other sampling sites, especially Co, Cr, Cu, Ni, and V. High values of Mn are mostly concentrated in the west bank of the reservoir (S27–S34). While high concentrations of Pb and Zn are more likely to be observed in the east bank of the reservoir (S19–S26), which might be due to traffic sources originating from vehicular emissions and the wear and tear of vehicle parts (tires, brakes, etc.) (Van der Gon and Appelman [2009,](#page-12-0) Dong et al. [2017\)](#page-11-0). The higher concentrations of Zn in the northeast bank might also be related to the zinc smelting plant in the north (Shilton et al. [2005](#page-12-0)). Such spatial variation of these metals might be due to the distinct distances between the sampling sites and the emission sources, the variation in the water flow rate, the nature of the sediment particles, and potential emissions, as well as metal characteristics (Bastami et al. [2014;](#page-11-0) Pan et al. [2017a](#page-12-0)). It is noteworthy that most of the upper outliers are located near the bank of the reservoir, which might indicate the presence of potentially polluted sediments generated by punctual and diffuse pollution source (D'Emilio et al. [2012](#page-11-0)).

The single pollution index (PI) and the integrated pollution index (IPI) are calculated to estimate the pollution levels of trace metals in sediments (Table [2](#page-6-0)). The IPIs and the most PIs of individual elements are in the range of 1 to 2, indicating a moderate level contamination of trace metals in sediments.



Fig. 2 The low-frequency magnetic susceptibility  $(\chi_{LF})$  (a) and the frequency-dependence magnetic susceptibility ( $\chi_{\text{FD}}\%$ ) (b) of the sediments in Wangjiaya reservoir

<span id="page-5-0"></span>Fig. 3 Concentrations of Co, Cr, Cu, Mn, Ni, Pb, V, and Zn in the sediment of Wangjiaya reservoir



<span id="page-6-0"></span>Table 2 Summary of the single pollution index (PI) and the integrated pollution index (IPI) in the sediments

	PI- Co	PI- <b>Cr</b>	PI- Cu	PI- Mn	PI- Ni	PI- Pb	PI- V	PI- Zn	IPI
Min	0.95	0.99	1.06	1.74	1.01	1.09	1.15	1.06	1.19
Max	1.51	1.42	2.05	2.85	1.89	1.61	1.56	5.45	1.94
Mean	1.32	1.10	1.65	2.41	1.59	1.33	1.43	1.66	1.56
Median	1.32	1.09	1.66	2.48	1.61	1.34	1.44	1.52	1.57

## Radiation levels and radiological hazard in sediments

Activity concentration of natural radionuclide and associated radiation hazard indices in the sediments were shown in our previous study (Lu et al. [2016](#page-12-0)). The relatively high concentrations of  $^{232}$ Th,  $^{226}$ Ra, and  $^{40}$ K are mostly observed in the downstream of the middle of the reservoir (e.g., S2–S6) with a few higher values scattered along the bank (e.g., S21, S26, and S33). This phenomenon might suggest that the variation in activity concentrations of  $^{232}$ Th,  $^{226}$ Ra, and  $^{40}$ K might be due to agriculture activities surrounding these sites (Gonzalez-Fernandez et al. [2010](#page-11-0); Saç et al. [2012](#page-12-0)). The previous studies have reported that the higher concentrations of  $^{226}$ Ra and  $^{40}$ K may be associated with the extensive exploitation of phosphate and potassium fertilizers in soils surrounding agricultural areas (Akhtar et al. [2005](#page-10-0); Bikit et al. [2005](#page-11-0); Krishnamoorthy et al. [2014](#page-12-0)). Although the specific levels of radionuclide activity in sediments are mostly related to the types of rocks or soils from which the sediments originate, the transport and mobility of natural radionuclide in the aqueous system is another important factor influencing the content of radionuclide in sediments at a given place (Tzortzis et al. [2004](#page-12-0); Krmar et al. [2009](#page-12-0); Strakhovenko et al. [2010](#page-12-0)). Thus, physicochemical conditions and water input from river runoff should be taken into consideration (Zorer et al. [2008](#page-13-0); Suresh et al. [2011\)](#page-12-0).

## Relationships between magnetic susceptibility and trace metals and natural radionuclides

#### Correlation analysis

The Pearson correlation coefficients  $(R)$  between pairs of the variables are listed in Table [3](#page-7-0). Significant correlations are observed between magnetic susceptibility indexes ( $\chi$ <sub>LF</sub>,  $\chi$ <sub>HF</sub>, and  $\chi_{FD}\%$ ) and the levels of most of detected metals and natural radionuclides.  $\chi$ <sub>LF</sub> is used to discuss the correlation between magnetic susceptibility and trace metals and radionuclides owing to significant positive correlations among the pairs of  $\chi$ <sub>LF</sub>,  $\chi$ <sub>HF</sub>, and  $\chi$ <sub>FD</sub>%.

As showed in Table [3,](#page-7-0) significant positive correlations are observed between  $\chi$ <sub>LF</sub> and Co (0.622,  $p$  < 0.01), Cu (0.442,  $p < 0.01$ ), Mn (0.483,  $p < 0.01$ ), Ni (0.485,  $p < 0.01$ ), and V  $(0.593, p < 0.01)$ . The previous studies reported that the significant correlations between the magnetic susceptibility and trace metals can indicate common sources (industrial pollution, parent rocks or pedogenic processes, vehicle-derived emissions, etc.), mutual dependence, and identical behavior of the magnetic particles and trace metals during the transport (Chaparro et al. [2010](#page-11-0); Chlupáčová et al. [2010](#page-11-0); Suresh et al. [2011](#page-12-0); Castañeda Miranda et al. [2016](#page-11-0)). Trace metal pollution is in many cases accompanied by emission of ferrimagnetic particles and Co, Ni, Cu, and Mn are more likely to concentrated in the magnetic fraction (Kukier et al. [2003;](#page-12-0) Rachwał et al. [2017\)](#page-12-0), which well explains the strong relationship between  $\chi$ <sub>LF</sub> and these elements. On the contrary, no correlations are observed between  $\chi$ <sub>LF</sub> and Cr, Pb, and Zn in this study (Table [3\)](#page-7-0). The absence of correlation between  $\chi_{LF}$  and the three elements might suggest variations in the sources and specific circumstances (Lu et al. [2007](#page-12-0); Zorer et al. [2008;](#page-13-0) Canbay et al. [2010](#page-11-0)). This result also indicates that magnetic susceptibility has not been affected significantly by anthropogenic activities which enhanced Pb and Zn concentrations in the sediment (Karimi et al. [2017\)](#page-11-0). In addition, it should be noticed that the IPIs show significant correlations with  $\chi_{LF}$  $(R > 0.500, p < 0.01)$ , which confirms that variations of ferrimagnetic mineral concentrations are consistent with the degree of trace metal pollution (Jordanova et al. [2004](#page-11-0); Wang et al. [2015](#page-12-0)).

The obvious difference was found in correlation analysis results of magnetic susceptibility and natural radionuclides (Table [3](#page-7-0)). The positive correlation are observed between  $\chi_{LF}$ and <sup>232</sup>Th (0.478,  $p < 0.01$ ) and <sup>40</sup>K (0.378,  $p < 0.05$ ), which are consistent with previous studies (Krishnamoorthy et al. [2014;](#page-12-0) Ramasamy et al. [2014](#page-12-0)) and might be the result of the absorption of magnetic minerals by lower grain sized fractions. In addition, significant positive correlations are observed between radioactive elements  $(^{232}Th$  and  $^{40}K$ ) and the five metals (Co, Cu, Mn, Ni, and V), which might be due to the common sources (e.g., the local agricultural activities and coal-fired power plant) and similar geochemical be-havior of the radionuclides and these metals (Lu et al. [2014a;](#page-12-0) Özmen et al. [2004](#page-12-0)).

Overall, the correlation analysis can suggest a potential for indicating anthropogenic pollution by the intensity of magnetic susceptibility, which are easy, prompt, and cost-effective to perform (Chaparro et al. [2017](#page-11-0)). To determine the quantitative relationship between magnetic susceptibility indexes and the pollution levels of trace metals and radionuclides, linear



<span id="page-7-0"></span>Table 3 Pearson correlation coefficients between magnetic susceptibility and the levels of heavy metals and natural radionuclides  $(n = 34)$ . Italic values in the table represent the significant positive **Table 3** Pearson correlation coefficients between magnetic susceptibility and the levels of heavy metals and natural radionuclides  $(n = 34)$ . Italic values in the table represent the significant positive

\*Correlation is significant at the 0.05 level (two-tailed) \*Correlation is significant at the 0.05 level (two-tailed)

\*\*Correlation is significant at the 0.01 level (two-tailed) \*\*Correlation is significant at the 0.01 level (two-tailed)

regression analysis was performed between magnetic susceptibility parameters and the IPI and AED. Figure 4 illustrates that a good linear relationship between the magnetic properties and the IPI and AED, which reflects the indication role of magnetic susceptibility in identifying the pollution of trace metals and natural radionuclides in the sediments (Chaparro et al. [2011;](#page-11-0) Yang et al. [2007](#page-13-0); Jaffar et al. [2017](#page-11-0)). Furthermore, the linear regression equations between MS and IPI, AED were established to semi-quantitatively assess trace metal pollution and radiological hazard.

#### Multivariate statistical analysis

In our previous study (Zhu et al. [2016\)](#page-13-0), three clusters of the trace metals have been identified by using the methods of cluster analysis (CA) and principal components analysis (PCA). Copper, Ni, Mn, V, and Co are identified a cluster and well correlated with each other. These five elements generally present high correlation with magnetic susceptibility. According to previous studies and the discussion above, this cluster is identified to be a mixed source, including natural sources, industrial sources, and local consumption residues (Wang et al. [2014;](#page-12-0) Zhu et al. [2016;](#page-13-0) Pan et al. [2017a](#page-12-0)). However, the elements of Pb, Zn, and Cr show weak or no correlation with the magnetic susceptibility. The significant positive correlation between Pb and Zn  $(0.503, p < 0.01)$  indicates similar sources, which might be mainly related to traffic sources (Van der Gon and Appelman [2009](#page-12-0); Pan et al. [2017b\)](#page-12-0), while Cr mainly originates from natural sources (Banerjee [2003;](#page-11-0) Zhu et al. [2016](#page-13-0)). Therefore, the trace metals originated

from industrial sources are more likely to present high correlation with magnetic susceptibility.

To explore the influence of different sources to contamination levels of sampling sites, K-means cluster analysis is carried out based on the variables of magnetic susceptibility, trace metals, and natural radionuclides. Three groups are well defined as illustrated by Fig. [5](#page-9-0). Most of the sample sites are included in group 1 (18 of 34) and group 3 (14 of 34), while S14 and S17 compose the group 2. The cluster results of the sample sites are clearly shown in Fig. [6](#page-9-0). Group 1 mainly consists of the sampling sites near the bank of the reservoir, which usually present higher values of certain variables determined in this study. This might be due to the point sources and untreated wastewater discharge along the bank (Chaparro et al. [2008;](#page-11-0) Yang et al. [2017](#page-13-0)). Group 2 only includes S14 and S17, which present lower values of the variables. Group 3 consists most of the sampling sites in the middle of the reservoir and the relatively low inter-site variability throughout the sites suggests that the sediment are mostly locally derived, well-mixed, and consistent in provenance (Zhang et al. [2011](#page-13-0)). The enhanced concentrations of the target pollutants in this group are taken to be a combination of industries and agricultures from upstream (Jiang et al. [2017\)](#page-11-0).

## Influence of different sources on magnetic susceptibility as an indicator

Previous studies have indicated that magnetic parameters offer potential as a proxy for trace metal levels in the sediments. However, the association of them might be complex, which is depended on the number and variety of emission sources, their

Fig. 4 Linear correlation analysis between magnetic susceptibility parameters and the integrated pollution index (IPI) and the annual effective dose rate (AED) in sediments ( $n = 34$ ). Shaded area shows the confidence limits for the expected value, while outside dotted lines display the confidence limits for an individual predicted value



<span id="page-9-0"></span>

Fig. 5 K-means cluster analysis of all sampling sites  $(n = 34)$  based on the variables of magnetic susceptibility, heavy metals, and natural radionuclides. The sites were classified into three groups, showing group 1 ( $\circ$ ), group 2 (+), and group 3 ( $\circ$ )

distance from the study area (Strzyszcz and Magiera [1998](#page-12-0); Suresh et al. [2011\)](#page-12-0). Except for human activities, natural processes including weathering of parent material or biologically mediated pedogenic processes can also influence the magnetic susceptibility of soils (Szuszkiewicz et al. [2016](#page-12-0)).

To evaluate the influence of different sources on the relationship between magnetic susceptibility and levels of trace metals and radionuclides, we analyzed the correlations of the three magnetic susceptibility parameters and levels of metals



Fig. 6 Groups of all sampling sites  $(n = 34)$  according to the K-means cluster analysis

and radionuclides of the sediments in the middle, southwest, and northeast of the reservoir, respectively (Table [4\)](#page-10-0). A significant improvement in correlation coefficients between magnetic susceptibility and the levels of trace metals and radionuclides are observed in the middle of the reservoir. In particular, the correlation coefficients between  $\chi$ <sub>LF</sub> and IPI is 0.856  $(p < 0.01)$  for samples from the middle of the reservoir, which is obviously higher than the value for samples from the overall reservoir  $(0.576, p < 0.01,$  Table [3\)](#page-7-0). Such difference can confirm that magnetic proxies provide a rapid means for detecting trace metal contamination caused by multi-anthropogenic pollution sources in a large-scale area and the sensitivity was controlled by pollution sources (Wang et al. [2015](#page-12-0)). Thus, magnetic susceptibility could be used as a first screening of the industrial pollution in the investigated areas.

# Magnetic detection of trace metal contamination in the sediment

Based on the correlation relationship between magnetic susceptibility and trace metals, a three-step procedure is proposed for using magnetic susceptibility in identification of areas and extents polluted by trace metals in this study.

The first step is a broad investigation of the distribution of magnetic susceptibility within a large area. Monitoring grids could be uniformly distributed in the study area, and 4 to 8 sampling sites per square kilometer are appropriate for longterm monitoring program according to our practice. Meanwhile, since the measurement of magnetic susceptibility is time- and cost-effective, more sampling sites could be included as required by special cases. High values of magnetic susceptibility would provide useful hints for preliminary screening of areas polluted by trace metals. Furthermore, the outliers and anomalies would provide information of potential sources. It has been suggested that this method might be more promising for areas where single source dominate the pollution pattern. Therefore, it is recommended that source identification should be fully explored for particular study areas (Wang et al. [2015\)](#page-12-0). This step offers facilities in optimizing monitoring campaigns in a large area. As second step, the field survey of magnetic susceptibility in situ is conducted in areas highlighted with high values of magnetic susceptibility. A field magnetic susceptibility meter with sensor coils or probes could be used in this step and the calibration of in situ magnetic susceptibility measurements with laboratory data should be conducted (Kapička et al. [1997;](#page-11-0) Gattacceca et al. [2004;](#page-11-0) Porsch et al. [2010\)](#page-12-0). The enhancement in magnetic susceptibility could indicate an increase of the concentration of heavy metals in contaminated sediments (Yoshida et al. [2003](#page-13-0); Boyko et al. [2004](#page-11-0)). Therefore, a detailed picture of the spatial distribution of trace metals can be obtained based on the result of magnetic susceptibility determination. These areas are supposed to be with trace metal pollution. Chemical analysis is

<span id="page-10-0"></span>Table 4 Pearson correlation coefficients between magnetic susceptibility and the levels of heavy metals and natural radionuclides in different locations of the Wangjiaya reservoir. Italic values in the table represent the significant positive correlation between the variables

	Middle $(S1-S18)$			Northeast (S19–S26)			Southwest (S27–S34)		
	$\chi_{\rm LF}$	$\chi_{\rm HF}$	XFD	$\chi_{\rm LF}$	XHF	$X_{\rm FD}$	$\chi_{\rm LF}$	$\chi_{\rm HF}$	XFD
Co	$0.882**$	$0.877**$	$0.841**$	0.046	0.053	$-0.222$	$0.883**$	$0.881**$	$0.809*$
Cr	$-0.285$	$-0.292$	$-0.279$	0.042	0.064	$-0.157$	$-0.754*$	$-0.761$ <sup>*</sup>	$-0.684$
Cu	$0.775**$	$0.766**$	$0.775**$	0.376	0.330	0.475	0.444	0.455	0.277
Mn	$0.713**$	$0.703***$	$0.747**$	0.612	0.560	0.667	$0.961**$	$0.957**$	$0.904**$
Ni	$0.775**$	$0.766***$	$0.792**$	0.111	0.071	0.266	$0.860**$	$0.866**$	$0.747*$
Pb	$0.698**$	$0.683***$	$0.745**$	0.559	0.528	0.416	$-0.538$	$-0.535$	$-0.641$
V	$0.877**$	$0.867***$	$0.850**$	0.690	0.660	0.552	$0.930**$	$0.932**$	$0.861**$
Zn	$0.580*$	$0.561^*$	$0.749**$	$-0.037$	$-0.016$	$-0.389$	$-0.889**$	$-0.894***$	$-0.851***$
IPI	$0.856**$	$0.842***$	$0.896**$	0.174	0.176	$-0.195$	0.599	0.598	0.471
$232$ Th	$0.641**$	$0.637**$	$0.607**$	$0.781*$	$0.780*$	0.579	$0.881**$	$0.885**$	$0.849**$
$^{226}Ra$	0.150	0.132	0.399	$-0.308$	$-0.298$	0.022	$-0.885**$	$-0.897**$	$-0.829*$
$^{40}\mathrm{K}$	$0.642**$	$0.649**$	0.461	0.094	0.087	0.174	$0.906**$	$0.913**$	$0.854**$

suggested in this step to identify the results and get accurate concentrations of trace metals. In the third step, a field survey based on chemical analysis in well-defined areas is performed and correlation between magnetic susceptibility and trace metals are to be analyzed. The correlation between magnetic susceptibility and environmental pollution should be further identified to establish a semi-quantitative method for longterm monitoring (Chaparro et al. [2015\)](#page-11-0).

In conclusion, we propose a monitoring procedure to use magnetic susceptibility in long-term pollution monitoring in sediment in lakes and rivers. The magnetic susceptibility could be used in discriminating potentially contaminated areas and chemical analysis subsequently could give detailed information of trace metal concentrations. Furthermore, interrelationship obtained by multivariate analysis will provide approximate estimations of trace metal concentrations by determination of magnetic susceptibility. This methodology gives a relatively fast and cost-effective tool for monitoring the pollution level by trace metals of the sediment. It is especially useful in tracing toxic metal pollution and repeat measurements within shorter intervals.

## Conclusions

This study revealed site-specific correlations between magnetic susceptibility and the levels of trace metal and natural radionuclides in the sediments. Significant enrichment of magnetic susceptibility is presented in those areas where concentrations of trace metals and natural radionuclides are higher. Because of the significant correlations between the variables,  $\chi$ <sub>LF</sub> is a good indicator for assessing the contamination levels of the sediment. Magnetic susceptibility is helpful in the rapid monitoring and discriminating different anthropogenic pollution. This provides a rapid and cost-effective methodology in assessment of trace metal pollution in sediment, especially those from industrial sources. A three-step procedure for the use of magnetic susceptibility in pollution monitoring is proposed. The environmental magnetism method is a helpful supplement to traditional geochemical methods in estimating the pollution levels and tracing the major sources of hazardous components in sediments. However, the nature of the relationship should be explored fully for particular environment and individual field settings before using magnetic susceptibility as a proxy for anthropogenic pollution.

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