One-century atmospheric heavy metal pollution on the southeast Mongolia Plateau: implications for pollution trend in China

One-century sediment records of heavy metal pollution on the southeast Mongolia Plateau: implications for air pollution trends in China

Dejun Wan[a,b,*], Lei Song[a], Xin Mao[a], Jinsong Yang[a], Zhangdong Jin[b,c], Handong Yang[d,*]

[a] Institute of Hydrogeology and Environmental Geology, Chinese Academy of Geological Sciences, Shijiazhuang 050061, China

[b] State Key Laboratory of Loess and Quaternary Geology, Institute of Earth Environment, Chinese Academy of Sciences, Xi'an 710075, China

[c] Institute of Global Environmental Change, Xi'an Jiaotong University, Xi'an 710049, China

[d] Environmental Change Research Centre, University College London, London WC1E 6BT, UK

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One-century sediment records of atmospheric heavy metal pollution on the southeast Mongolia Plateau, North China

Abstract

Historical records of heavy metals in lake sediments from remote areas are important for assessing pollution trends of the regional atmosphere. Based on comparison analyses of heavy metals, Pb isotopes and total carbon in sediment cores from two relatively remote lakes (DL and ZGST) on the southeast Mongolia Plateau, the trends of atmospheric heavy metal pollution history during ~1900-2016 were reconstructed. Results suggest that the ZGST core is not suitable to reconstruct past pollutions owing to receiving lots of catchment in-wash of anthropogenic heavy metals via atmospheric deposition. The current anthropogenic fluxes of Zn, Cd and Pb in Lake DL are 11.7, 0.104 and 2.44 mg m^{-2} yr^{-1} respectively, close to those in Lake Sayram in West China, but lower than most other lakes in China. Both anthropogenic fluxes of Zn, Cd and Pb and $^{206}Pb/^{207}Pb$ ratios suggest that (1) before ~1950 atmospheric metal pollution was negligible; (2) since ~1950 the pollution became detectable but did not accelerate until ~1980, corresponded well with the beginning of social and economic development after the foundation of China in 1949 and the rapid development after the Reform and Opening-up in 1978; and (3) since ~2000 atmospheric Pb stopped to increase owing to phasing out of leaded gasoline. Moreover, combining the DL and ZGST records with other lake sediment records from remote regions, a two-stage evolution picture of atmospheric heavy metals over the last century in China was obtained by fitting analysis: (1) slight pollution in the 1950s-70s and (2) accelerated pollution since ~1980, although pollution levels vary among different regions. This suggests that 1980 can be recommended as the beginning of air pollution intensification over China, which is significant for defining the Anthropocene in China.

Keywords: heavy metal; air pollution; atmospheric deposition; sedimentary record; North China; Anthropocene

1. Introduction

Owing to rapid development of society and economy in China in recent decades, air pollution has become more and more serious and one of the most concerning environmental issues in recent years (Cai et al., 2017; Gao et al., 2017; Ma et al., 2016; Song et al., 2017). A typical characteristic of current air pollution is the increasing levels of atmospheric pollutants such as heavy metals of Zn, Cd, Pb and Hg (Gao et al., 2017; Tian et al., 2015; Pan and Wang, 2015). Long-term history of atmospheric heavy metals contributes to understanding pollution mechanisms, predicting the future trend, and setting up effective measures to control air pollution. However, observations on atmospheric heavy metals in China are mostly conducted in a few urban or rural sites and usually lasting for short periods such as few months or years (Chan and Yao, 2008; Pan and Wang, 2015). Therefore, it is necessary to seek other approaches like investigating geologic records to reveal long-term trends of atmospheric heavy metals in China.

In remote lakes, anthropogenic heavy metals are usually derived dominantly from atmospheric deposition. Hence sediment records from these lakes usually document atmospheric deposition changes in the past and thus often employed to reconstruct pollution histories (Catalan et al., 2013; Engels et al., 2018; Jones et al., 2015; Sarkar et al., 2015; Wan et al., 2016). In recent years, such kind investigations have been carried out in many lakes from relatively remote areas of China, such as the Tibetan Plateau, West China, the Yungui Plateau (e.g. Bing et al., 2016; Jin et al., 2010; Li et al., 2017; Lin et al., 2018; Wan et al., 2016; Yang et al., 2010; Zeng et al., 2014). These investigations provide valuable data for understanding long-term trends and background values of atmospheric heavy metals in China. However, most of these works often used only a single sediment core from a lake (e.g. Bing et al., 2016; Li et al., 2017; Wan et al., 2016; Zeng et al., 2014). In this case it may be difficult to differentiate some possible variations of heavy metals caused by natural processes or local pollutions from that by regional atmospheric deposition. Hence the reliability of such reconstructions needs to be further confirmed. Moreover, although the southeast Mongolia Plateau seems to be of relatively good air quality at present (Fig. 1a, $PM_{2.5}$, Ma et al., 2016), it may still receive considerable amount anthropogenic pollutants via long-range atmospheric transport because of its close distance from the heavily-polluted Jing-Jin-Ji region. However, to date little is known about long-term trend of air pullution in this region, owing to few direct observation or high-resolution geological record investigation. Therefore, it is necessary to recover multiple geological records and make comparing study to faithfully reconstruct air pollution history in this region.

In this study sediment cores were recovered from two lakes (Lake Dali (DL, the second largest lake in Inner Mongolia) and Lake Zhagesitai (ZGST)) in relatively remote areas of the southeast Mongolia Plateau (Fig. 1b). The sediment profiles of heavy metals as well as Pb isotopes, Ti and total carbon (TC) contents from these two lakes were set up. The heavy metal records that can faithfully reflect regional air pollutions were employed to reconstruct atmospheric pollution trends of heavy metals during ~1900-2016, and to discuss their evolutions and mechanisms in the southeast Mongolia Plateau. By comparison and fitting analyses with lake sediment records from the other four regions, to obtain a rough picture of atmospheric metal pollution over the last century in China. This study is significant for understanding long-term evolutions and mechanisms of heavy metals in atmosphere of the Mongolia Plateau and even in China as a whole. Additionally, as heavy metals are one of the most important and widespread pollutants of current human activities, their evolutions in the atmosphere can provide evidence for defining the Anthropocene in China.

2. Materials and methods

2.1. Site description

DL and ZGST, located on the southeast Mongolia Plateau of North China (Fig. 1b), are both inland, closed-basin lakes. DL (43°13′–43°23′N, 116°29′–116°45′E, ~1230 m above the sea level) is the second largest lake in Inner Mongolia, with a water-surface area of 238 km^2 and a maximum water depth of 11 m (Xiao et al., 2009). There are two permanent rivers from the northeast and two intermittent streams from the southwest entering the lake. The lake is covered with 0.5-1-meter ice from November to April (Xiao et al., 2009). ZGST lies about 50 km to the northeast of DL (Fig. 1b). It is a small shallow lake, with a water-surface area of ~2 km^2 and a maximum water depth of ~2 m. There are only small intermittent streams entering the lake. In the DL area, the average annual precipitation is 350-400 mm and ~70% of the annual precipitation occurs in June–August. The average annual evaporation is about 1287 mm. The average annual temperature is 1-2 ºC with a January average of -17 to -24 ºC and a July average of 16-18 ºC (Xiao et al., 2009). As ZGST is a small lake, we did not find any climatic data in the lake area, but the climate here may be similar to DL owing to short distance (~50 km) between them and similar natural environment.

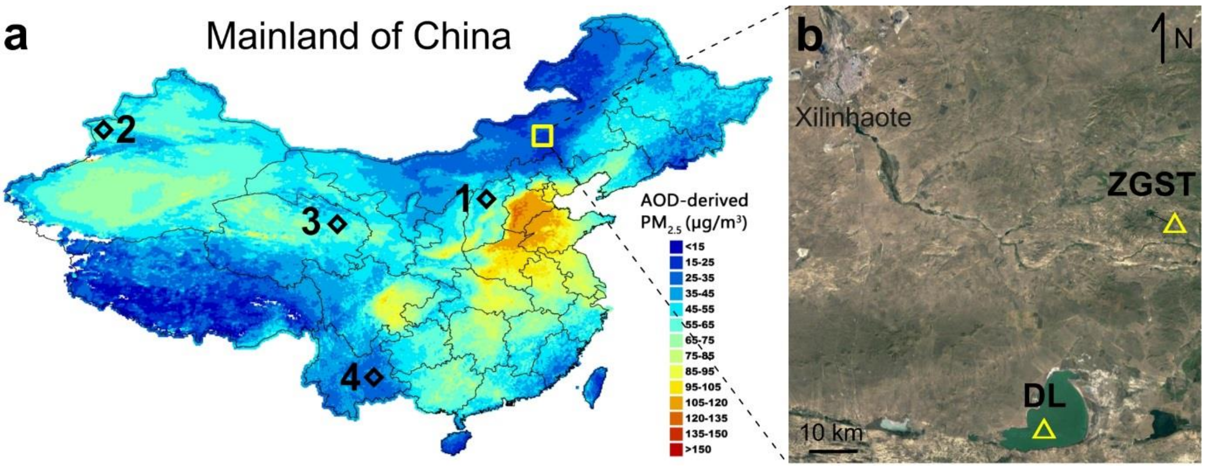

Fig. 1 (**a**) The study area in the Mainland of China. Colors represent spatial distributions of 10-year (2004-2013) mean $PM_{2.5}$ levels over China (Ma et al., 2016). 1, 2, 3, 4 and 5 represent locations of Lake Gonghai (Wan et al., 2016), Lake Sayram (Zeng et al., 2014), Lake Qinghai (Jin et al., 2010) and Lake Fuxian (Liu et al., 2013). (**b**) Sampling sites of sediment cores in Lake Dali (DL) and Lake Zhagesitai (ZGST) on the southeast Mongolia Plateau.

2.2. Sampling and analysis

In July 2016, two sediment cores were recovered using a gravity corer from relatively deep water areas of lakes DL and ZGST (Fig. 1b). The cores were sectioned at 1.0-cm intervals, and dried at -25 °C with a vacuum-freezing dryer. Their subsamples were ground to powder smaller than 63 μm for chemical analysis.

For chemical measurements, each ground sample was weighted for ~0.125 g and hot-digested with mixed acids of $HClO_4$, HNO_3 and HCl (Wan et al., 2016). Concentrations of Ti and Zn were measured by a Leeman Labs Profile inductively coupled plasma atomic emission spectrometry (ICP-AES) and other trace metals such as Cd, Pb, and Pb isotopic ratios by an Agilent 7700x inductively coupled plasma mass spectrometry (ICP-MS). The measurements show high precisions with most relative standard deviations less than 5%.

Activities of ^{210}Pb, ^{137}Cs and ^{226}Ra in the core were detected by a low-background germanium detector (EG and GOrtec Gamma Spectrometry) at the State Key Laboratory of Lake Sciences and Environment. ^{210}Pb activities were determined at 46.5 keV and ^{137}Cs activities were at 662 keV. ^{226}Ra activities were detected at 295 keV and 352 keV γ-rays emitted by its daughter isotope of ^{214}Pb (Wan et al., 2016). Unsupported ^{210}Pb activities ($^{210}Pb_{ex}$) were calculated by subtracting ^{226}Ra activities from the total ^{210}Pb activities. Standard errors (1σ) have been calculated from the counting statistics.

3. Results and discussion

3.1. Core dating

Although sediment cores were taken from lakes of DL and ZGST, only the DL core was dated by using ^{210}Pb and ^{137}Cs activities. This is because that the DL core is more suitable to reconstruct regional atmospheric heavy metal pollution history than the core from ZGST after comparison analysis on the heavy metal profiles in these two cores (see 3.2).

Equilibrium of total ^{210}Pb activity with supported ^{210}Pb activity is at a depth of ~30 cm of the DL core. Unsupported ^{210}Pb activities in the core decline more or less following an exponential trend with depth. The core was dated using the constant rate of ^{210}Pb supply (CRS) model (Appleby, 2001). The CRS dating result suggests that the 1963 is at the depth of 17.5 cm. This agrees with the ^{137}Cs record, which shows that the 1963 ^{137}Cs peak is likely to be at 15-20 cm depth. However, there might be a big uncertainty for the CRS chronologies at the bottom around 25-30 cm of the core, due to low $^{210}Pb_{ex}$ activities with relatively big errors at this section. So we extrapolated ages for the section according to the average sediment rate (0.0769 g cm^{-2} yr^{-1}) in the central (11-20 cm) section of the core. There may be still some uncertainty for the ages, but it is of limited influence on reconstructing past heavy metal pollutions which occurred mainly in the top ~20-cm section of the core.

3.2. Heavy metal concentrations in the sediment cores from DL and ZGST

Heavy metal concentrations in sediment cores from DL and ZGST are shown in Fig. 2. For easy comparison of metals in the two cores, their X axes are set at the same range of 3/4*Average[C_i] $\leq$ X $\leq$ 5/4*Average[C_i] ([Ci] is the concentration of element Ci). Concentrations of Zn, Cd and Pb in the DL core range 55.6-68.7, 0.139-0.276 and 16.6-20.1, with averages at 63.1, 0.206 and 18.4, respectively; while those in the ZGST core range 45.3-66.4, 0.114-0.272 and 12.7-18.5, with averages at 55.8, 0.193 and 15.6, respectively. The profiles of the heavy metal concentrations show that concentrations in the bottom 5 cm sections of the DL and ZGST core are relatively low and stable, which can be considered as the background, and this is followed by a general increase trend towards the sediment surfaces.

Compared with their backgrounds, the present concentrations (top 5 cm) of Zn, Cd and Pb in the DL core increased by 21%, 82% and 18%, respectively, while in the ZGST core increased by 32%, 109% and 25%, respectively. The figures suggest smaller increases in heavy metal concentrations in the DL core compared to those in ZGST. The difference is partly due to relatively higher background values in DL than ZGST, but the more important cause is that the ZGST core may receive more catchment in-wash of anthropogenic heavy metals via atmospheric deposition owing to its location closer to the lakeshore relative to the DL core (Fig. 1). Therefore, the DL is more suitable to reveal past regional atmosphere metal pollutions compared to ZGST. Hence in the following only the DL core is employed to reconstruct past pollution.

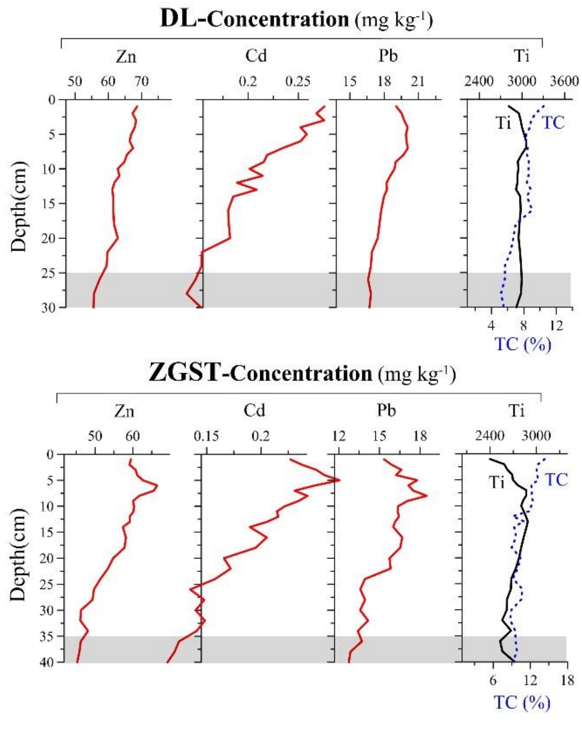

Fig. 2 Heavy metal concentrations, Ti, and TC in sediment cores from lakes of DL and ZGST. For comparison, their X axes are set at the same range of 3/4*Average[C_i ≤ X ≤ 5/4*Average[C_i].

3.3. Pb isotopes and sources of heavy metals in the sediment cores

Pb isotope results show that $^{206}Pb/^{207}Pb$ ratios decreased obviously in the top 0-~13 cm section (~1980-2016) of the DL core and in the top 0-~20 cm section of the ZGST core compared to their lower sections (light-blue shadow areas), respectively. The changes are approximately opposite to the increase trends of Pb concentrations as well as other heavy metals in both cores (Fig. 2 and 3). The $^{206}Pb/^{207}Pb$ ratios in the top sediments are close to those in the anthropogenic sources such as Chinese Pb ore and coal (Cheng and Hu, 2010) and urban aerosols from Beijing (Mukai et al., 2001) and Tianjin (Wang et al., 2006) (Fig. 3). Decline of the $^{206}Pb/^{207}Pb$ ratios indicates human-induced Pb, probably as well as other metals, showing pollution signals in the upper sections of the cores.

Human-induced heavy metals in lake sediment are usually derived from three main pathways: atmospheric wet and dry deposition, catchment erosion, and direct dumping (Chen et al., 2016). The lakes are relatively remote, there are few local pollution sources in their catchments, so direct release of anthropogenic heavy metals can be ignored. For catchment erosion, it brings dominantly natural-origin heavy metals into the lake, but it may also bring a certain amount of anthropogenic heavy metals previously deposited and stored in the catchments. These facts suggest that the human-induced heavy metals in the two lakes are dominantly derived from regional atmospheric deposition rather than local pollutions, and this is confirmed by similar evolution trends of heavy metals (especially Cd, Pb and Zn) and Pb isotopes in the two lakes (Fig. 3). Therefore, heavy metal variations in these cores have the potential to reconstruct atmospheric metal deposition history. However, absolute concentrations of heavy metals may be affected by lacustrine sediment processes such as changes of primary productivity and carbonate content (Duan et al., 2014; Wan et al., 2016), indicated by approximately mirror variations of Ti and TC in both the DL and ZGST cores (Fig. 3). Therefore, absolute metal concentrations cannot be directly employed to reconstruct past heavy metal pollutions.

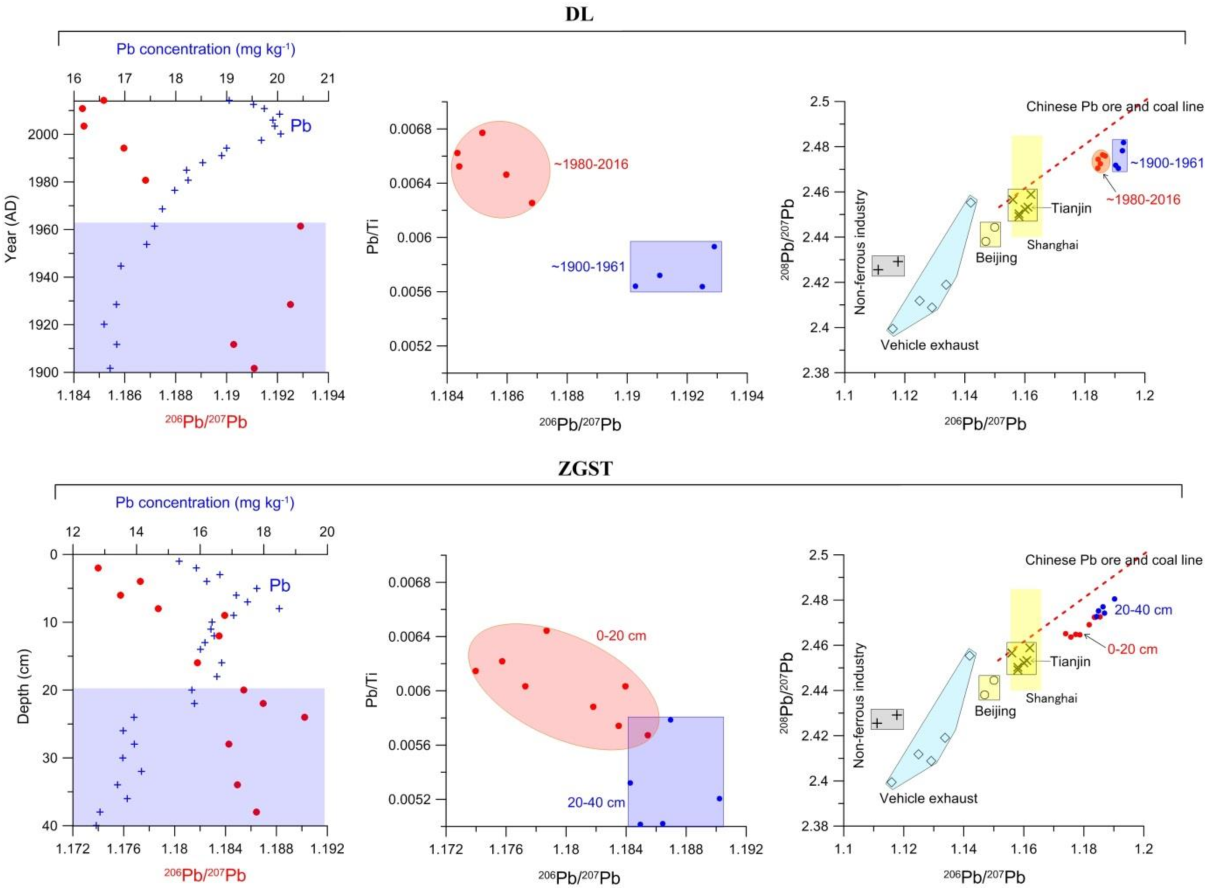

Fig. 3 Pb isotopes ($^{206}Pb/^{207}Pb$ and $^{208}Pb/^{207}Pb$), Pb concentrations, and Pb/Ti in sediment cores from DL and ZGST. Also shown are Pb isotopes in Chinese Pb ore and coal (Cheng and Hu, 2010), vehicle exhaust (Liu et al., 2004; Wang et al., 2002; Zheng et al., 2004), non-ferrous industry (Liu et al., 2004; Wang et al., 2002; Zheng et al., 2004), urban aerosols from Beijing (Mukai et al., 2001), Tianjin (Wang et al., 2006) and Shanghai (Chen et al., 2005).

3.4. Reconstruct natural and anthropogenic metal fluxes

To reconstruct atmospheric metal pollution histories, natural and anthropogenic fluxes of these metals were calculated in the DL sediment core, respectively. The anthropogenic and natural fluxes ($M[Flux_{anthropogenic}]$ and $M[Flux_{natural}]$, mg m^{-2} yr^{-1}) are calculated according to the following equations by using Ti as reference element (Jin et al., 2010; Kuwae et al., 2013; Wan et al., 2016; Zeng et al., 2014):

$$M[Flux_{anthropogenic}] = (M[C_{sample}] - Ti[C_{sample}] \times M[C_{background}]/Ti[C_{background}]) \times R \times \rho \times 10$$

$M[Flux_{natural}] = M[C_{sample}] \times R \times \rho \times 10 - M[Flux_{anthropogenic}]$

where $M[C_{sample}]$ and $M[C_{background}]$ represent bulk and background concentrations of metal M in sediment, respectively. $Ti[C_{sample}]$ and $Ti[C_{background}]$ represent the bulk and background concentrations of Ti in sediment, respectively. In this study, the average bulk metal concentrations in the bottom 5-cm section of the sediment core are considered as the background. R is the sedimentation rate (cm yr^{-1}) and ρ is the dry bulk density (g cm^{-3}) of the sediment.

The reconstructed natural and anthropogenic metal fluxes in Lake DL are shown in Fig. 4. For easily comparing changes between different metals, their X axes are set at the same range of $0 \leq X \leq 2$*Average[flux]. The results suggest minor changes in natural fluxes of these metals (Fig. 4), implying the reliability of the reconstructed results to some extent.

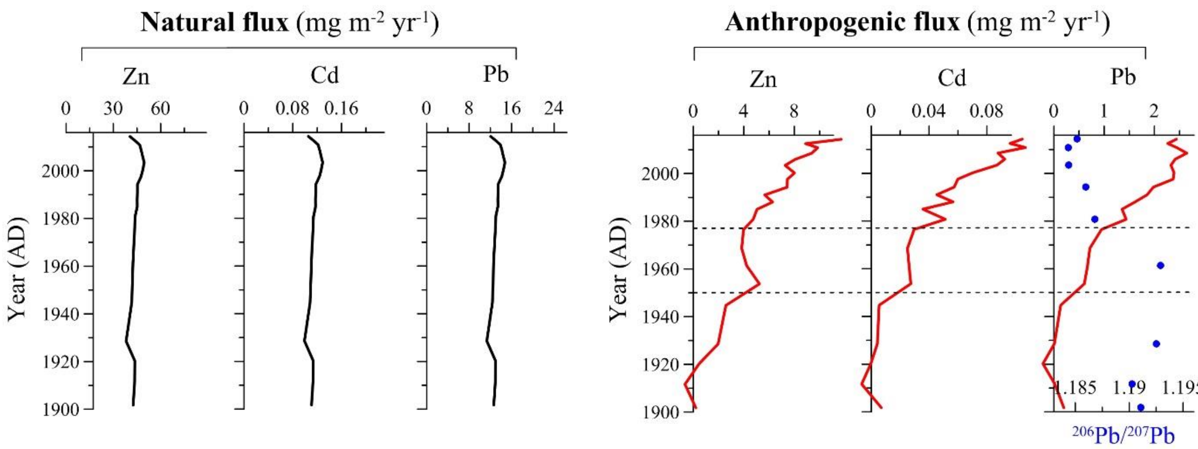

Fig. 4 Reconstructed natural and anthropogenic fluxes of Zn, Cd and Pb in Lake DL. For comparison, their X axes are all set at the same range of $0 \leq X \leq 2$*Average[flux].

3.5. Evolution of atmospheric heavy metals during ~1900-2016 on the southeast Mongolia Plateau

Before ~1950, anthropogenic fluxes of Zn, Cd and Pb were very low and relatively stable especially Cd and Pb (Fig. 4), implying negligible atmospheric metal pollution on the southeast Mongolia Plateau. This is confirmed by relatively high $^{206}Pb/^{207}Pb$ ratios (Fig. 3), further suggesting the reliability of the reconstructions. For Zn, there was a slight increase in its fluxes and concentrations from the 1920s to the 1940s (Fig. 4). This was likely related to local natural forces in Lake DL rather than pollution, indicated by no such change of Zn in the ZGST sediment core.

From ~1950 to ~1980, anthropogenic fluxes of Zn, Cd and Pb began to increase gradually, but the increase was not significant until ~1980 (Fig. 4). These changes imply that the atmosphere in the region started to be polluted by human-induced heavy metals noticeably since ~1950, but the pollution level was relatively low and did not increase rapidly until ~1980. On the whole, these two-stage changes corresponded well with the beginning of the development in socio-economy after the foundation of China in 1949 and the accelerated development of socio-economy after the Reform and Opening-up in 1978 in China, respectively (Cheremukhin et al., 2015; Zhang et al., 2017). Compared with heavy metal emissions, the anthropogenic metal fluxes show similar trends to the estimated annual atmospheric emissions in China from 1949-2012 (Fig. 5, Li et al., 2012; Tian et al., 2015). Further correlation analysis suggests that their correlation coefficients (R^2) are as high as 0.916 (Zn), 0.957 (Cd) and 0.634 (Pb), respectively. However, the anthropogenic metal fluxes show different trends of annual gross industrial production (GIP) and annual heavy industry production of Xilinguole District on the southeast Mongolia Plateau (Fig. 6). This may be due to (1) priority development of heavy industry in the early developing stage with limited pollution-controlling technology and equipment (Cui, 2012; Wang, 2015), (2) shrinkage of heavily polluting industries and rapid development of less polluting industries such as Information Tech, New Energy, Biology, and Finance after ~2000 (Duan and Tan, 2013; Yu and Chen, 2012), and (3) currency inflation in China especially in recent one or two decades (Chen, 2011).

In ~2000, the anthropogenic Pb flux stopped to increase and kept at around 2.5 mg m^{-2} yr^{-1} in the following years, whereas those of Zn and Cd still increased quickly (Fig. 4). The change of Pb was a result of the phasing out of leaded gasoline in ~2000 in China, which results in a sharp decline in atmospheric Pb emission (Fig. 5) (Duan and Tan, 2013; Li et al., 2012; Tian et al., 2015). Although DL is situated at a relatively remote area with few motor vehicles, the atmospheric Pb in this area also stopped to increase since ~2000, similar to other records in China such as Lake Gonghai (Wan et al., 2016) in central North China (Fig. 6). This suggests the effectiveness of ban on leaded gasoline to control atmospheric Pb in the whole country. However, although the atmospheric Pb emission decreased by a half after 2000 (Fig. 6), the anthropogenic Pb flux showed insignificant decrease (Fig. 4). This is likely due to (1) re-suspended old lead from ground-surface environments (Wan et al., 2016) and (2) increases in Pb emissions from other sources such as coal combustion and industry in recent years (Tian et al., 2015).

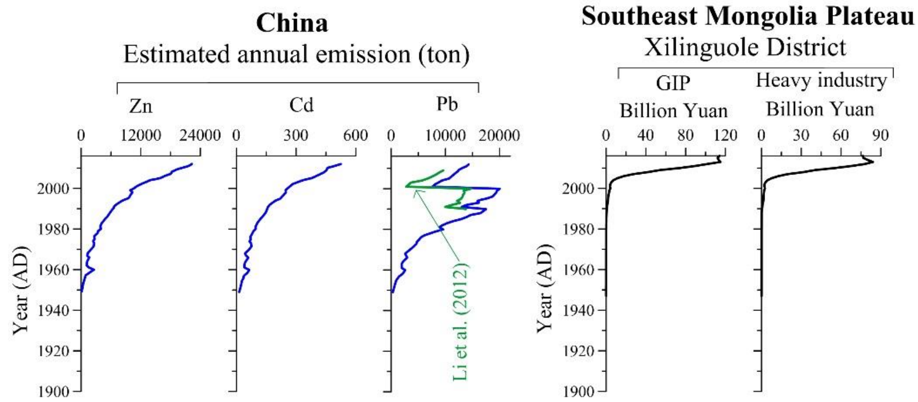

Fig. 5 Blue and green curves are estimated annual atmospheric emissions (ton) of heavy metals in China during 1949-2012 (Tian et al., 2015) and Pb emissions during 1990-2009 (Li et al., 2012). Black curves are annual gross industrial production (GIP) and annual heavy industry production in Xilinguole District during 1949-2016 (SBXD, 2017).

3.6. Atmospheric metal pollution trend over the last century in China

Compared with other sedimentary records (Fig. 6, and many others not shown in the figure), it can be found that the current anthropogenic fluxes of Zn, Cd and Pb in DL are close to Lake Sayram in West China but lower than most other lake sediment records in China, implying a low pollution level of atmospheric heavy metals on the southeast Mongolia. This is generally comparable to the air pollution distribution pattern over China as indicated by spatiotemporal trends in $PM_{2.5}$ concentrations in Fig. 1a (Ma et al., 2016). However, their evolution trends are similar to most other records from relatively remote regions of China, such as Lake Gonghai (Wan et al., 2016) in central North China, Lake Sayram (Zeng et al., 2014) in West China, Lake Qinghai (Jin et al., 2010) on Tibetan Plateau, and Lake Fuxian (Liu et al., 2013) in Southwest China. This may reflect the historical trends of anthropogenic heavy metals in China.

Considering these records are all from relatively remote areas of China, they have the potential to reflect heavy metal pollutions in the regional atmosphere. Therefore, they can be employed to represent a rough picture of atmospheric metal pollutions over the last century in inland China. Fitting analysis of all proxies (except $^{207}Pb/^{206}Pb$ in Lake Fuxian) from the five sedimentary records in Fig. 6 suggests a slight pollution trend atmospheric heavy metals in the 1950s-1970s and an accelerated pollution trend since ~1980 in China (Fig. 6, red curve), which is decades or even more than a century later than that in European and North American regions (Marx et al., 2016). These two trends corresponded well with the two developing stages of society and economy in China, i.e. the beginning development of socio-economy after the establishment of the People's Republic of China in 1949 and the rapid socio-economic development after the Reform and Opening-up in 1978 (Cheremukhin et al., 2015). Moreover, considering implications of heavy metal pollutions for evaluating the influence of recent human activities on the natural environment, the time of ~1980 can be recommended as the beginning of intensification in China's air pollution, which is significant for defining the Anthropocene in China.

However, there are some studies revealing slightly different atmospheric heavy metal pollution trends in China. For example, a lake sediment record from Lake Daihai in Inner Mongolia revealed a high pollution level of Pb in 1926 and 1945 but almost no pollution during 1946-1988 (Han et al., 2007), and an ice core from Guliya ice cap on west Tibetan Plateau revealed almost no pollution of Pb during ~1940-1970 but a high pollution level during ~1860-1940 (Sierra-Hernández et al., 2018). These differences may be due to (1) uncertainties of representativeness and dating in some sediment records (Yang and Turner, 2013) and (2) uneven and different start times of social-economic development over China (Liu et al., 2008). Therefore, in future it is necessary to recover more ideal sedimentary records to explore detailed spatial-temporal patterns of atmospheric pollution history in China.

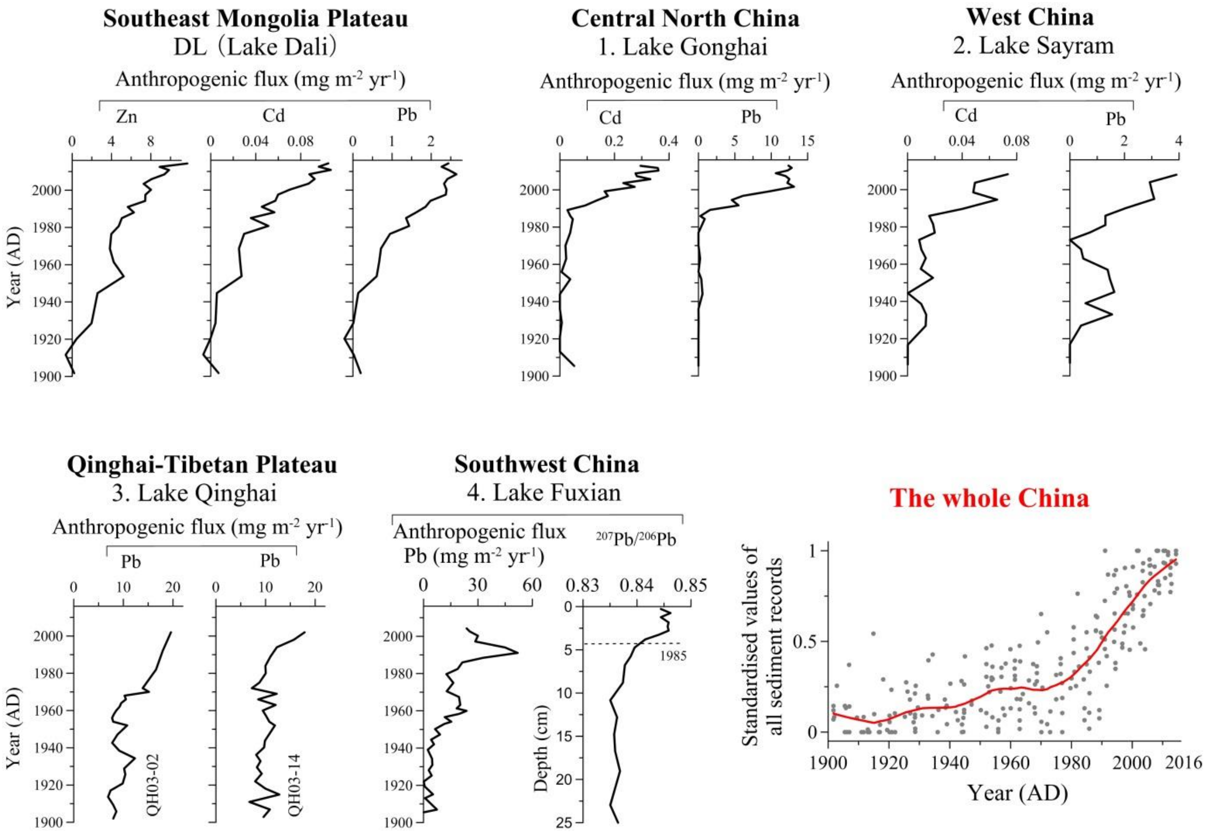

Fig. 6 Comparison with other sediment records of heavy metal pollutions from different regions of China (Fig. 1a), including Lake Gonghai (Wan et al., 2016), Lake Sayram (Zeng et al., 2014),

Lake Qinghai (Jin et al., 2010), and Lake Fuxian (Liu et al., 2013). In the last figure, red curve is the fitted trend of all the five sedimentary records in China, with dots showing standardised individual values of heavy metals in the five records.

4. Conclusions

In this study high-resolution sediment cores were taken from two lakes (DL and ZGST) from relatively remote areas of the southeast Mongolia Plateau in North China. Based on comparison analyses of elemental concentrations, Pb isotope ratios, and TC contents in the cores, atmospheric heavy metal pollution history during ~1900-2016 was reconstructed. Based on the reconstructed results, the following conclusions are drawn:

1. Before ~1950, the reconstructed anthropogenic deposition fluxes of atmospheric Zn, Cd and Pb in this region were close to zero and show minor changes, implying negligible atmospheric metal pollution.

2. From ~1950 to ~1980 these anthropogenic metal fluxes increased slightly and since ~1980 they became to increase rapidly, implying the atmospheric metal pollution was not intensified until ~1980.

3. In ~2000, the atmospheric Pb flux stopped to increase and kept at about 2.5 mg m^{-2} yr^{-1} in the following years, resulting from the phasing out of leaded gasoline in ~2000 in China.

4. Although atmospheric heavy metal pollution levels vary among different regions, the evolution trends of atmospheric heavy metals are comparable with most sediment records from other relatively remote areas of China, owing to roughly synchronous developing history of society and economy in China. Fitting analysis in sediment

records of heavy metals from five regions over China suggests that the ~1980 can be recommended as the beginning of intensification in China's air pollution, which is significant for defining the Anthropocene in China.

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Declarations of interest: None

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