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RESEARCH ARTICLE

History metal (Pb, Zn, and Cu) deposition and Pb isotope variability in multiple peatland sites in the northern Great Hinggan Mountains, Northeast China

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Abstract
Placer gold mining is important anthropogenic sources of dust and metals that can strongly influence the environmental quality of the surrounding ecosystem. However, scarce studies have focused on evaluating the influence of placer gold mining on historical metal deposition in the surrounding ecosystem in the northern Great Hinggan Mountains, which is located at northeast of China. To address this research gap, four peatland cores with different distances to a gold placer in the northern Great Hinggan Mountains were selected in this study. Based on the ^{210}Pb depth-age model, historical variations in the Pb isotope and deposition fluxes of Pb, Cu, and Zn were reconstructed. The results show that metal deposition in the northern Great Hinggan Mountains was mainly influenced by the placer gold mining around the 1900s when the gold placer started to produce gold, and placer gold mining more seriously influenced the western sites that were closer to the placer gold mining. With increasing global metal productions after 1930, the proportion of the metals from placer gold mining sources gradually decreased, and part of Pb were transported via the atmosphere from other regions (e.g., Europe, East Asia). With the implementation of environmentally friendly policies and the decreasing anthropogenic production of Pb, Cu, and Zn around the world, deposition fluxes of these metals in the northern Great Hinggan Mountain began to decrease after 2000.

Keywords Pb isotope · Metals · Great Hinggan Mountain · Placer gold mining · Peatland

Introduction

With increasing global human activity, large amounts of metals are being emitted into the atmosphere and are finally deposited into natural ecosystems (Gao et al. 2014a; Li et al. 2014). Anthropogenic sources (e.g., industry, transportation, residential living) have produced large amounts of metals and have seriously influenced the environmental quality of surrounding regions (Islam and Tanaka 2004; Zhou et al. 2015). The mining industry, such as gold mining, is an important anthropogenic source of metals and widely distributed around the world. Metals emitted from the mining industry could be transported through the atmosphere as aerosols accompanied by dust (Boamponsem et al. 2010). And surrounding soils, in particular, often exhibit being higher concentrations than those in other regions (Malm 1998; Ferreira da Silva et al. 2004). Furthermore, most gold placers are distributed in forest and river ecosystems, where the natural background concentrations of metals are low and concentrations are more easily increased by the deposition of external metals (Mol et al. 2001; Li et al. 2014). Thus, gold placers could increase the concentrations of metals in surrounding natural ecosystems and cause regional ecological risk in natural ecosystem (Tarras-Wahlberg et al. 2001).

Rivers in the northern Great Hinggan Mountains which locate in the upstream of Heilong River (Amur River) are rich in gold placers and are the important gold placer deposits in China. The gold placer deposits began to produce gold at the end of the 1880s in Yanzhigou, Mohe county (Chen et al. 2006; Yu et al. 2014). Metals such as

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Pb, Cu, and Zn are the associated metals in this gold placer (Chen et al. 2006), which could release and deposited in the surrounding ecosystem together with dust during gold mining (Egidarev and Simonov 2015; Martinez Cortizas et al. 2016). Thus, Yanzhigou gold placer was an important potential pollution source that potentially increases the historical deposition fluxes of these metals in the surrounding regions in the northern Great Hinggan Mountains. Except local metal sources, dust and metals emitted from other regions in northeastern China or even from other regions of the world (e.g., Europe, North America) could also be transported and deposited into the Great Hinggan Mountains via atmospheric processes (Bao et al. 2016; He et al. 2018). With increasing population and industrial development over the last 150 years, regional human activities also led to more metals being deposited in the Great Hinggan Mountains and been proofed in the middle of the Great Hinggan Mountains (Bao et al. 2012, 2015, 2016); however, scarce studies have focused on evaluating the historical influence of placer gold mining and regional human activities on metal accumulation process in the northern Great Hinggan Mountains.

Peatlands are an important ecosystem that widely distributed in the Great Hinggan Mountains (Xing et al. 2015). Because of its anaerobic environment and its ability to receive dust via atmospheric deposition, metals can be deposited and stored in peatlands for thousands of years (Gao et al. 2014b). Most of the metals in peatland are adsorb on organic particles and their associated microorganism community (Schaller et al. 2013). Because of a low diffusion rate of metals within peat soils, only the upper layer (mainly the diffusive boundary layer) potentially accumulates high amount of metals from the atmosphere. The migration process of metals from up to down layers and the loss of metals in peatland could be ignored (Martini et al. 2007). Therefore, peatlands were widely used to reconstruct historical metal deposition fluxes and sources around the world (Ohlson et al. 2006; Bao et al. 2012).

Not only the ratio of metals/conservative elements (i.e. enrichment factors, EF) sensitive to regional human activities (Cortizas et al. 2002; Shotyk et al. 2002), but also Pb isotope ratios could be used to identify anthropogenic and natural sources of Pb in natural ecosystems and atmospheric aerosols (Luck and Othman 2002; Shotyk et al. 2005; Bao et al. 2015). Therefore, these factors could evaluate the influence of placer gold mining on the environmental quality of surrounding peatland ecosystem (Boamponsem et al. 2010; Zakaria et al. 2015). To evaluate the influence of placer gold mining and regional human activities on historical metal deposition fluxes in the northern Great Hinggan Mountains, peat cores collected from four different peatlands in this region were selected for metal concentrations and Pb-isotope analysis. Based on ^{210}Pb dating, the historical deposition fluxes of Pb, Cu, and Zn in the studied region during the last 150 years were reconstructed by metal concentrations. And then, the impact of potential anthropogenic sources on the metal deposition fluxes in the northern Great Hinggan Mountains was revealed by EFs of metals and Pb isotope.

Materials and methods

Site description and sampling

The Great Hinggan Mountains have an elevation range of 180–2029 m above sea level, and are characterized by a dry, cold winter from November to April and a wet, hot summer from July to August (Bao et al. 2012). The Great Hinggan Mountains have a temperate continental monsoon climate, with a mean annual temperature of −4.3 to −2.8 °C and a mean annual precipitation of 400–600 mm. Detailed information about these mountains also has been provided elsewhere (Gao et al. 2018a). The studied peatlands are located in the northern region of the Great Hinggan Mountains in the northernmost part of China. These peatlands are located in a valley and have received metals through dry and wet deposition from nearby regions. The Yanzhigou gold placer was located at the westernmost region of the study region and began to produce gold in AD 1888 (Mohe government 2017). Four peat cores with different distances to the gold placer were selected in this study. Two peat cores were selected in the eastern region that are far from the gold placer (Hongtu, HT, N 51.62°, E 124.24°, 60 cm sampled; Huyuan HY, N 51.94°, E 123.63°, 40 cm sampled). Samples from these peatlands were collected in September 2014 (Gao et al. 2018b). The other two peatlands are selected with a short distance from the gold placer (Luoguhe, LGH, N 53.13°, E 122.06°, 42 cm sampled; Tuqiang, TQ, N 52.94°, E 122.85°, 77 cm sampled). These samples were cored and sampled in September 2016.

Vaccinium uliginosum is the dominant plant species in all four peatlands, accompanied by *Ass. Ledum palustre var. angustum*, *Carex schmidtii*, and *Alnus hirsuta*. The locations of all cores and gold placer were determined using a portable global positioning system (GPS) and are shown in Fig. 1. Peat cores were sectioned into 1-cm intervals with a stainless steel knife for ^{210}Pb dating and other analyses. The samples were stored in polyethylene plastic bags and subsequently brought to the laboratory for further analysis. These samples were then loosely disaggregated to facilitate air-drying at 20 °C.

Chronology

Reconstructions of the last 150 years of peat deposition are frequently based on ^{210}Pb data (Turetsky et al. 2004) and the constant rate of supply (CRS) model (Binford 1990). Peat samples were analyzed for their ^{210}Pb ages at 1-cm intervals

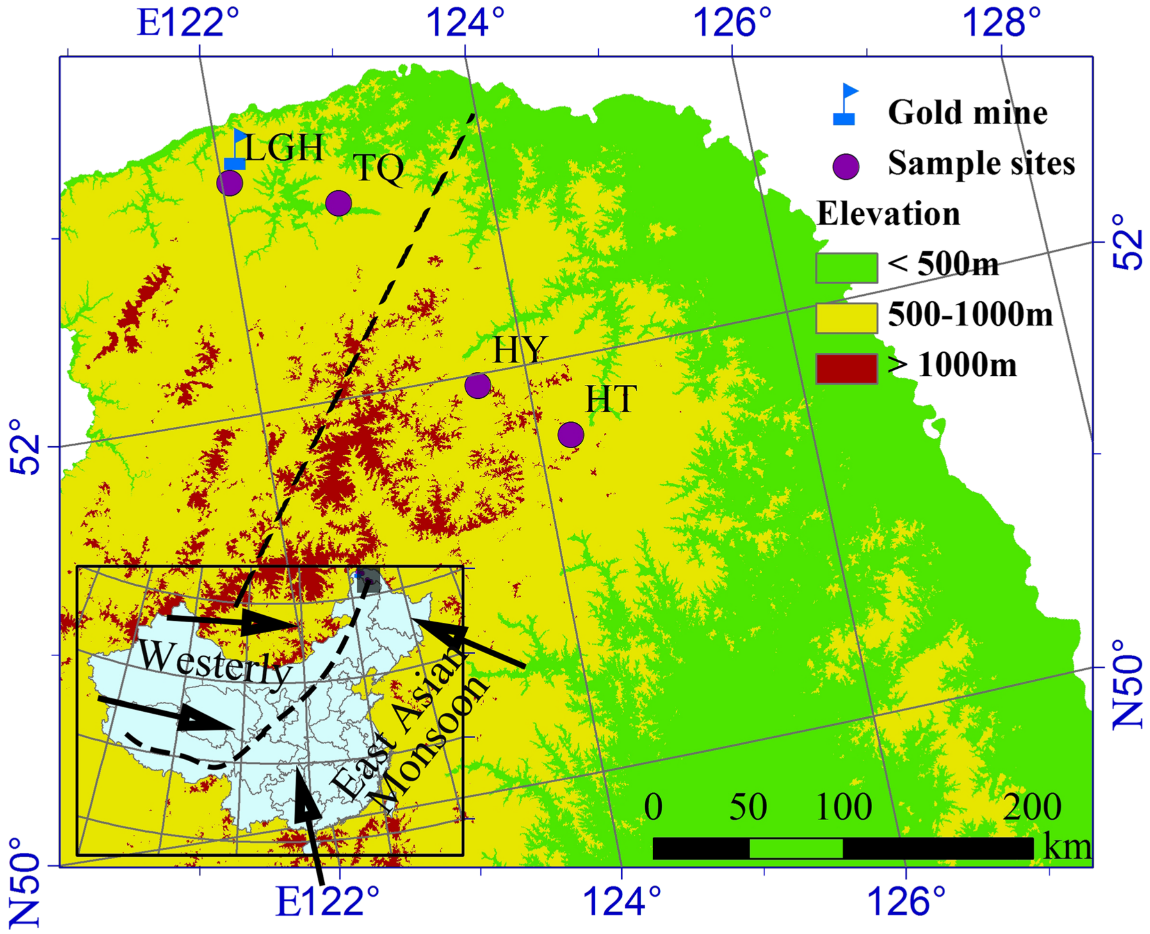

Fig. 1 Locations of the LGH, TQ, HY, and HT peatlands in the northern Great Hinggan Mountains in the northernmost part of China (gray rectangle). The flag is the Yanzhigou gold mine, whereas the dashed line represents the approximate limit of the summer monsoon (Chen et al. 2015)

by measuring the gamma ray emissions of the samples using a highly pure germanium semiconductor and a low-background gamma spectrometer (OTEC Instruments Ltd., USA). Soil samples in centrifuge tube with known activities were regarded as the standard radioactive sources and supplied by the National Institute of Metrology, China (Nr. 4NSG/0612). More than 30 samples in each peat core were selected for chronologic analyses at the Analysis and Test Center of the Northeast Institute of Geography and Agroecology, Chinese Academy of Sciences. The dry bulk densities of the samples were determined by oven drying at 105 °C for 12 h (Gao et al. 2016). Peat accumulation rates were calculated from the depth-age model and the dry bulk density at each depth within the peat cores.

Geochemical analysis

Based on ^{210}Pb age-depth model, all samples accumulated within 150 years in the four peat cores were analyzed for their Ti, Pb, Zn, and Cu concentrations, stable Pb isotopes, and total carbon and nitrogen contents. For the analyses of selected metals and stable Pb isotopes, the samples were digested using the same procedure as that used for the metal analysis in Bai et al. (2011) (i.e., total digestion using a concentrated mixture of $HNO_3/HClO_4/HF$). The analyses of total metal contents were performed using atomic emission spectrometry with inductively coupled plasma (ICPS-7500), and the recovery rates for metals in the State Standard Reference Materials (GBW07401) were approximately 93–109%. The stable Pb isotopes (^{204}Pb, ^{206}Pb, ^{207}Pb, ^{208}Pb) were measured using a Quadrupole Inductively Coupled Plasma Mass Spectrometry (NeXion 350D ICP-MS Spectrometer, PerkinElmer, USA), and the precision of stable Pb isotopes method for reference material (Multi analyte custom grade solution, ZZBIO-68) was approximately 10% at the 95% confidence level. The total carbon (TC) concentrations of the samples were calculated using LOI and bulk density values (multiplying the organic matter content by 0.5) (Lamarre et al. 2012). The total nitrogen contents of peat soil were measured after digestion (concentrated H_2SO_4 and catalytic) using a Flow Continuous Chemistry Analyzer. All the geochemical analyses were running at the Analysis and Test Center of the Northeast Institute of Geography and Agroecology, Chinese Academy of Sciences.

Enrichment factors

The variations in Pb, Zn, and Cu concentrations throughout the peat cores were analyzed to establish environmental oscillations and anthropogenic influence. For this purpose, the enrichment factor (EF) of a particular element Me was calculated as:

$$\mathrm{EF} = ([\mathrm{Me}]/[\mathrm{Ti}])_{\mathrm{peat}}/([\mathrm{Me}]/[\mathrm{Ti}])_{\mathrm{reference}}$$

where [Me] is the concentration of a metal (i.e., Pb, Cu, Zn). To calculate the EF of a particular element, Ti is commonly used as the conservation element because Ti in soil dust is resistant to chemical weathering (Cortizas et al. 2002; Shotyk et al. 2002; Weiss et al. 2002). Therefore, the EFs of Pb, Zn, and Cu were calculated by normalizing their concentrations to Ti in this study.

Because the normalization of data requires a reference (baseline), it is necessary (albeit difficult) to determine a period without human activities. Thus, the minimum values of the ratios $([\mathrm{Me}]\ /\ [\mathrm{Ti}])_{\mathrm{lithogenic}}$ (e.g., Me = Pb, Zn, or Cu) were selected to represent pre-anthropogenic inorganic geochemical background (Gao et al. 2014a). Furthermore, in order to distinguish between anthropogenic and lithogenic inputs of Pb, Zn, and Cu, we followed the approach proposed by a previous study (Gallego et al. 2013), in which these inputs are calculated as follows:

$$[Me]_{lithogenic} = [Ti]_{sample}{}^{*}([Me]/[Ti])_{lithogenic}$$
$$[Me]_{anthropogenic} = [Me]_{total} - [Me]_{lithogenic}$$

where the unit of $[\mathrm{Me}]_{\mathrm{lithogenic}}$ is in mg/kg, and [Ti] refers to the concentration of Ti at each depth in the four peat cores.

Results

Peat chronology

Based on ^{210}Pb data, the depth-age model and the sedimentary rates of the four peatlands were calculated using the CRS model with MATLAB 2012a software (Higuera 2010). The uncertainties of the ages at each peat depth were estimated by the standard deviations of 1000 randomly selected chronologies, and the mean ages were used as the peat ages at each depth in the four peatlands (Fig. 2). The peat accumulation rates at site LGH were slowest in all four peat cores in this study, where 10 cm of peat was accumulated from 1910 to the present. The peat accumulation rates in HT and HY were faster than those in the other two peat cores, as nearly 40 cm and 34 cm of peat accumulated over the last 150 years, respectively. The peat accumulation rates in the four peat cores decreased from the eastern site (HT) to the western site (LGH), and the peat accumulation rates in TQ were similar with the average peat accumulation rates of the four peat cores in this study (Fig. 2).

TC, TN, and C/N mass ratios

The concentrations of TC and TN, along with C/N mass ratios in the four peatlands during different periods, are shown in Table 1. Over the last 150 years, the average concentration of TC in HY was 367 ± 38 mg/g, which was the lowest of all of the four peat cores. The highest TC concentration of 465 ± 17 mg/g occurred at site LGH. The highest concentration of TC from 1900 to 1950 was 472 ± 14 mg/g, which occurred at site LGH. The average concentrations of TN in the four peat cores ranged from 10.3 ± 2.9 to 16.3 ± 5.8 mg/g. The highest

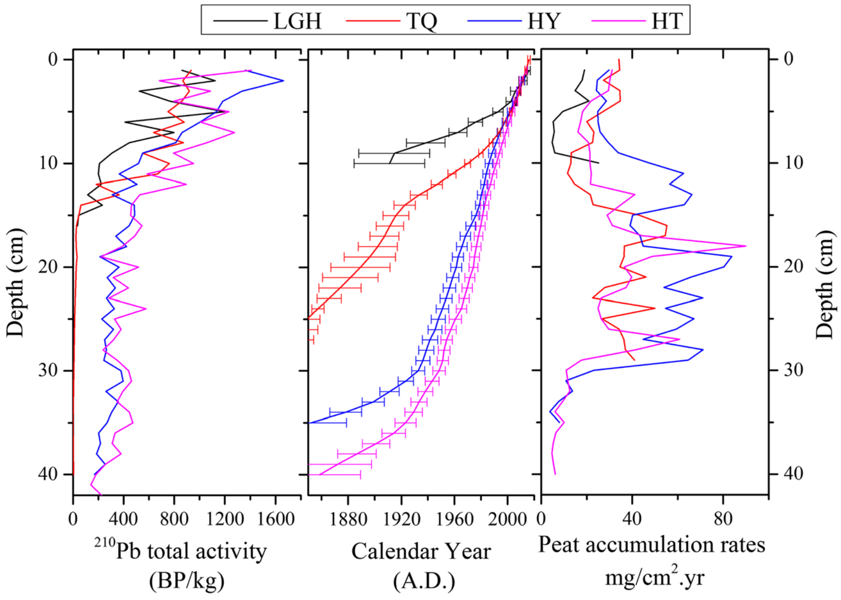

Fig. 2 Variations in total ^{210}Pb activities, ^{210}Pb-CRS-based chronology with 95% confidence interval calculated used data from the cores, and peat accumulation rates in the LGH, TQ, HY, and HT peat cores

Table 1 Average concentrations and standard deviations of total carbon (TC), nitrogen (TN), Ti, Cu, Pb, and Zn in four peat sites during four different periods (before 1900, 1900–1950, 1950–1980, and after 1980)

Sites	Periods	TC mg/g	TN mg/g	C/N	Ti mg/kg	Cu mg/kg	Pb mg/kg	Zn mg/kg
HT	Before 1900 *n* = 5	449 ± 18 A	12.4 ± 1.1 A	36.3 ± 2.6 A	380 ± 55 A	42.8 ± 10.7AB	12.8 ± 1.4 A	13.9 ± 1.9 A
	1900–1950 *n* = 8	430 ± 15AB	12.2 ± 1.0 A	35.6 ± 3.3 A	594 ± 195B	57.4 ± 15.3 A	17.4 ± 3.9 A	29.4 ± 10.9A
	1950–1980 *n* = 13	420 ± 15 B	15.4 ± 2.0 B	27.7 ± 4.6 B	494 ± 129AB	55.7 ± 28.0 A	28.6 ± 10.6B	43.7 ± 52.3A
	After 1980 *n* = 15	434 ± 20AB	13.4 ± 1.7AB	33.0 ± 4.6AB	316 ± 160A	18.1 ± 18.7B	9.75 ± 4.18A	56.3 ± 69.4A
	Total *n* = 41	431 ± 19 a	13.7 ± 2.0 a	32.2 ± 5.3 a	434 ± 181a	40.7 ± 27.1a	17.6 ± 10.4a	41.9 ± 52.3a
HY	Before 1900 *n* = 2	344 ± 22 A	15.1 ± 0.1A	22.8 ± 1.6A	1364 ± 27A	26.8 ± 4.0	43.3 ± 6.8A	51.0 ± 13.6A
	1900–1950 *n* = 9	329 ± 16 A	15.1 ± 1.0A	21.9 ± 2.1A	1591 ± 127A	20.7 ± 4.5A	35.9 ± 4.7A	55.7 ± 26.0A
	1950–1980 *n* = 11	357 ± 23 A	14.2 ± 1.4AB	25.5 ± 3.5A	1369 ± 307A	28.5 ± 24.2A	34.9 ± 15.7A	73.3 ± 57.4A
	After 1980 *n* = 12	408 ± 23 B	11.4 ± 2.2B	36.8 ± 6.5B	456 ± 175B	15.7 ± 21.9A	27.6 ± 9.9A	68.2 ± 60.6A
	Total *n* = 34	367 ± 38 b	13.5 ± 2.2a	28.4 ± 7.8a	1106 ± 536b	21.8 ± 19.4ab	33.1 ± 11.6b	65.5 ± 49.5a
TQ	Before 1900 *n* = 10	406 ± 24 A	20.7 ± 1.7 A	19.7 ± 2.0 A	408 ± 151A	12.1 ± 8.8 A	12.5 ± 7.6 A	18.0 ± 8.4 A
	1900–1950 *n* = 7	414 ± 16 A	20.9 ± 3.1 A	20.4 ± 4.8 A	292 ± 54 A	16.5 ± 11.3A	10.9 ± 6.9 A	18.6 ± 6.8 A
	1950–1980 *n* = 3	444 ± 35 A	10.7 ± 1.7 B	41.7 ± 3.8 B	249 ± 188A	5.19 ± 1.12A	7.94 ± 4.87A	41.1 ± 31.0A
	After 1980 *n* = 9	433 ± 38 A	9.60 ± 0.77B	45.3 ± 5.0 B	323 ± 218A	19.7 ± 40.2A	17.6 ± 13.6A	55.4 ± 47.4A
	Total *n* = 29	420 ± 30 a	16.3 ± 5.8 a	30.1 ± 12.9a	337 ± 165a	14.8 ± 23.1b	13.2 ± 9.71a	31.5 ± 31.8a
LGH	1900–1950 *n* = 3	472 ± 14 A	12.4 ± 4.6 A	41.7 ± 14.2A	94.1 ± 68.8A	31.5 ± 40.8A	8.43 ± 8.28A	48.4 ± 37.0A
	1950–1980 *n* = 2	471 ± 20 A	8.07 ± 1.11A	58.8 ± 5.7 A	163.4 ± 85.0A	3.52 ± 1.22A	2.62 ± 0.32A	25.5 ± 0.2 A
	After 1980 *n* = 5	459 ± 19 A	10.0 ± 1.4 A	46.7 ± 8.8 A	229.3 ± 84.5A	8.55 ± 3.30A	12.6 ± 5.5 A	36.5 ± 4.6 A
	Total *n* = 10	465 ± 17 c	10.3 ± 2.9 b	47.6 ± 11.1b	176 ± 94a	14.4 ± 22.8b	9.36 ± 6.72a	37.9 ± 19.6a

a, b, c: groups of four sites were divided by one-way ANOVA analysis and Tukey's honestly significant difference (Tukey-HSD) test

A, B: groups of four periods were divided by one-way ANOVA analysis and Tukey's honestly significant difference (Tukey-HSD) test

average concentration of TN from 1900 to 1950 was 20.9 ± 3.1 mg/g, which occurred at site TQ. In LGH peat core, the average C/N mass ratio was 47.6 ± 11.1 over the last 150 years; and the highest value of the C/N mass ratio was 58.8 ± 5.7, which appeared in 1950–1980. Unlike the LGH peat core, the average C/N mass ratios at the other three sites were approximately 30, as they ranged from 28.4 ± 7.8 to 32.2 ± 5.3.

Metal concentrations and deposition fluxes

The highest average concentration of Ti was 1106 ± 536 mg/kg in HY core (Table 1); this value was nearly six times as large as that in LGH core, which was the lowest of the four peat cores (176 ± 94 mg/kg). Unlike Ti concentrations, which exhibited clear differences between the four cores, the ranges of Pb, Cu, and Zn concentrations were similar. In addition, the concentrations of these three metals in the four cores all decreased from east to west, especially for Cu, which decreased from 40.7 ± 27.1 to 14.4 ± 22.8 mg/kg. The concentrations of Pb, Cu, and Zn in the four cores during most periods increased from the beginning of the 1900s (before 1900) to the present (after 1980). However, in HT and LGH, high concentrations of Cu appeared at the beginning of 1900 and decreased after 1980.

The differences in Ti fluxes between the western and eastern sites in the northern Great Hinggan Mountains indicate that dust deposition in these two regions differed (Fig. 3). In the western sites (i.e., TQ and LGH), high Ti fluxes appeared at the beginning of the 1900s and around the 2000s. High Ti fluxes in the eastern sites appeared in the middle of the twentieth century (i.e., 1940s–1980s). Before 1900, the Ti fluxes in TQ were slightly higher than those in the other peatlands, and the metal fluxes were the highest of all four peatlands. The Pb and Cu fluxes in the TQ peatland were around 10 mg/m^2·year and 20 mg/m^2·year respectively. Highest Pb and Cu fluxes were also appeared in the 1910s and then gradually decreased. After 1930, the Pb and Cu fluxes in HY and HT increased markedly and were higher than those in LGH and TQ. Especially, from 1950 to 1980, the fluxes of Pb and Cu increased more markedly in the eastern two sites and much higher than in the western sites. The fluxes of Pb and Cu in both four sites in the northern Great Hinggan Mountains gradually started to decrease at the 1980s. After 2000, the Pb and Cu fluxes in the four peatlands were similar and around 3 mg/m^2·yr and 2 mg/m^2·year, respectively. Both metal fluxes in the four sites were lower than those when the local placer gold began mining around the 1900s.

Stable Pb isotopes

The variations in different stable Pb isotope ratios with depth in the four peat cores are shown in Fig. 4. The average ^{208}Pb/^{204}Pb ratio in LGH was 39.3 and higher than those

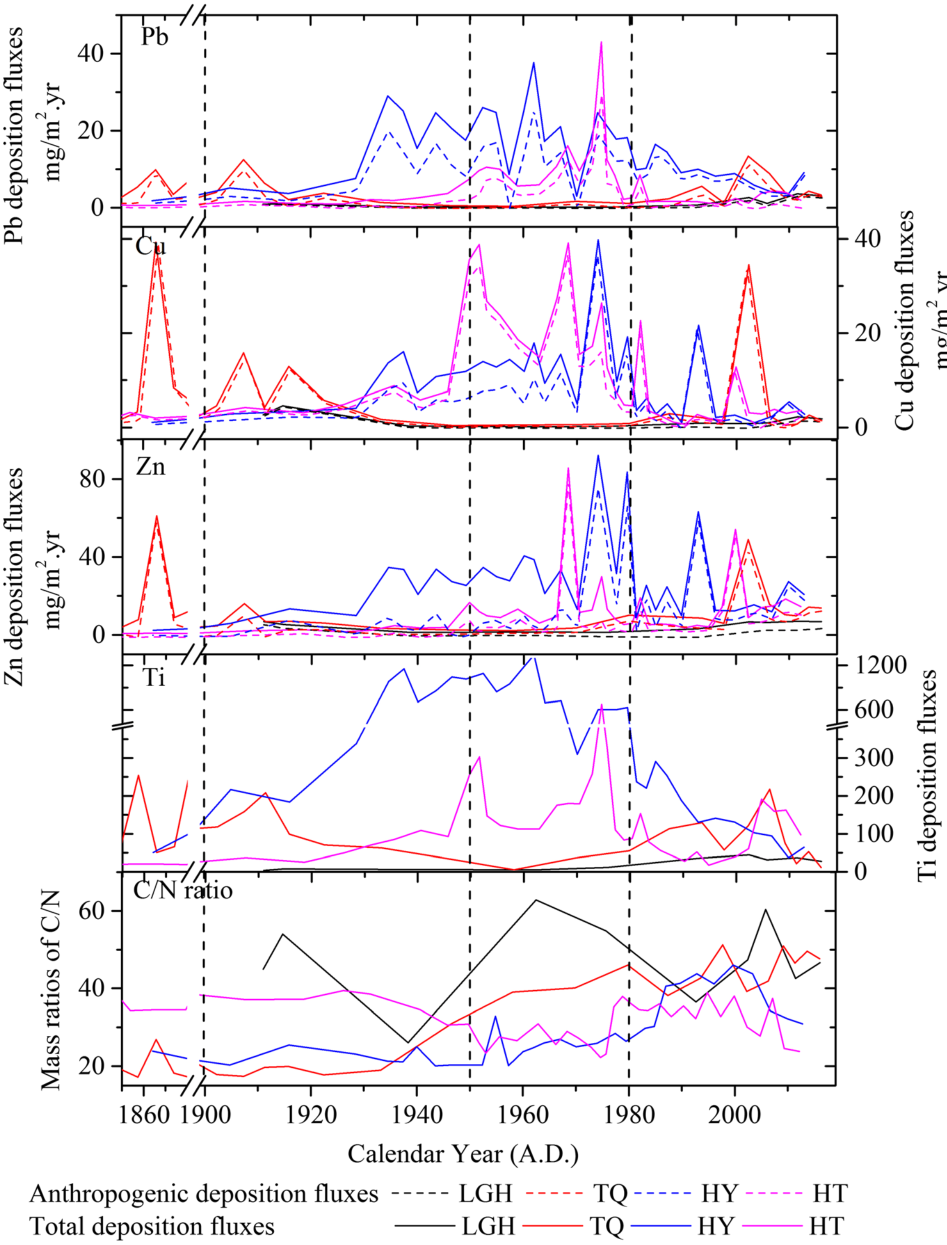

Fig. 3 Total accumulation rates and anthropogenic accumulation rates of Pb, Cu, Zn, Ti, and C/N mass ratios in the LGH, TQ, HY, and HT peatlands over the last 150 years

observed in the top 20-cm peat soils in the other peat cores where the average ratio was between 38.5 and 38.9. The ratios of $^{208}Pb/^{204}Pb$ in TQ clearly increased with increasing depth, and similar trend also occurred in the core from HY between a depth of 20 cm to the bottom of the core. In the top 20 cm of all four peat cores, the ratios of $^{208}Pb/^{206}Pb$ markedly decreased with the depth increasing. Especially in LGH, the ratios of $^{208}Pb/^{206}Pb$ decreased from 2.14 to 2.10. Peak values of the $^{208}Pb/^{206}Pb$ ratio were existed at the bottom of the other three peat cores (e.g., 2.14 in TQ ca. 24 cm). Unlike the $^{208}Pb/^{206}Pb$ ratios, the $^{208}Pb/^{207}Pb$ ratios fluctuated but slightly increased with increasing depth in TQ and HY. The $^{208}Pb/^{207}Pb$ values in the four peat cores ranged from 2.42 to 2.46. With increasing depth in HT, an increasing trend existed around 10 cm, where the ratios of $^{208}Pb/^{207}Pb$ increased from 2.42 to 2.45. However, the $^{208}Pb/^{207}Pb$ ratios in HT peat core consistently decreased from 2.45 to 2.43 as the depth of the peat layers increased from 10 to 40 cm.

Discussion

Chronology of metal pollution in the northern Great Hinggan Mountain

Increases in the mass ratios of metals and conservative elements are typically caused by increasing proportions of anthropogenic metals, thus, EFs of metals could be used to

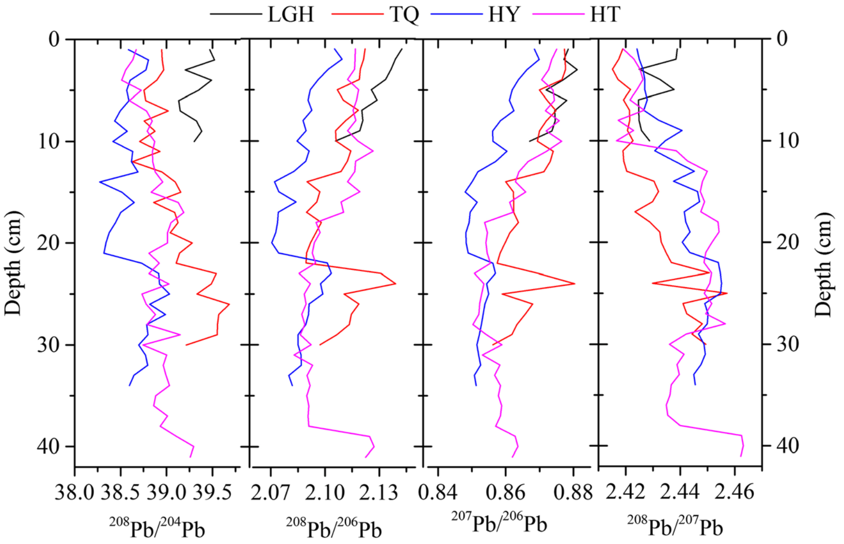

Fig. 4 Variations in the $^{208}Pb/^{204}Pb$, $^{208}Pb/^{206}Pb$, $^{207}Pb/^{206}Pb$, and $^{208}Pb/^{207}Pb$ ratios in the LGH, TQ, HY, and HT peat cores

evaluate the influence of human activities on the concentrations of metals in soils (Shotyk et al. 2001, 2002). In this study, Ti was selected as conservative element to calculate the EF of metals (i.e., Pb, Cu, and Zn), and the historical variations in the EF of these three metals in the four peatlands over the last 150 years were reconstructed (Fig. 5). Ti deposition fluxes were selected to reconstruct historical dust deposition and evaluate the natural metal deposition history. The total deposition fluxes of metals were selected to evaluate deposition history of metals, and anthropogenic deposition fluxes which were calculated by EFs were used to evaluate the anthropogenic metal deposition history (Fig. 3). Except for the metal deposition fluxes, the degree of peat decomposition is also an important factor that influences the accumulation process of metals in peatlands (Biester et al. 2012). Thus, the C/N ratio was selected as peat decomposition indicator to evaluate the impact of peat decomposition on the metal deposition fluxes in each peatland (Fig. 3).

Since 1895, people from other regions began to migrate to (e.g., Southern China) or invade (e.g., Russia and Japan) Northeast China and exploit its forest and mineral resources (Zhang et al. 2006). The increasing human activities caused

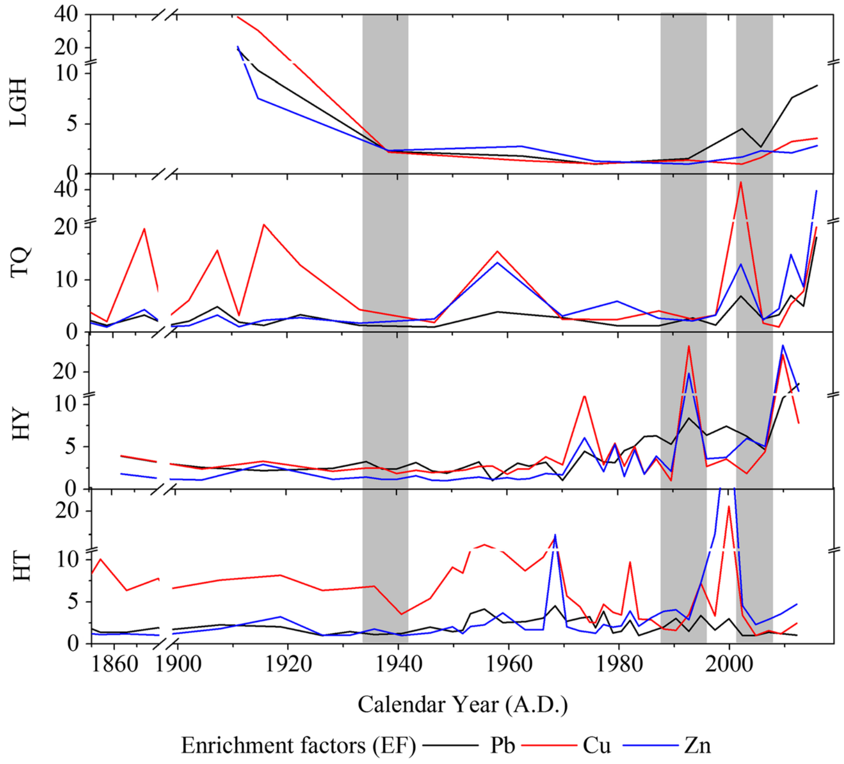

Fig. 5 Historical variations in the enrichment factors of Pb, Cu, and Zn in the LGH, TQ, HY, and HT peatlands over the last 150 years. The gray shadow indicates the enrichment factors of Cu, Pb, and Zn in part peatlands obviously higher than nearby periods

the deposition fluxes of metals and black carbon obvious increased in other regions in Northeast China had been proofed by previous studies (Gao et al. 2014a, 2016). Since that time, the Great Hinggan Mountains also had been exploited by humans without any protection policies, and regional anthropogenic metal productions in Northeast China also started to influence the metal deposition fluxes in the northern Great Hinggan Mountains. The gold placer (Yanzhigou) was started mining at the end of the nineteenth century (Wang and Sun 1997). At the beginning of the 1900s, the EFs of metals at site LGH were higher than those in other periods; the Cu_{EF} value was approximately 40. In the TQ peatland, the values of Pb_{EF} and Zn_{EF} were not markedly higher than they were during other periods, and they were slightly higher than 1 in this period. However, the Cu_{EF} values exhibited several peak values and reaching a maximum value of approximately 20 before 1920. Metals such as Cu and Pb are associated with gold in the Yanzhigou gold placer (Chen et al. 2006); both could be released to the surrounding ecosystem through water and the atmosphere as dust during the mining process. The placer gold mining which caused Cu pollution was more serious than those of the other two metals in the northern Great Hinggan Mountains, especially in western regions around the 1900s. The gold placer is located approximately 20 km to the north of the LGH peatland. The short distance between the LGH peatland and the gold placer led more metals from the gold placer to accumulate in LGH peatland than other sites. In the eastern sites (i.e., HY and HT), the EFs of metals at the beginning of the 1900s were stable and slightly higher than they were during other periods. During this period, the Cu_{EF} values in HT were slightly lower than 10 and much higher than the EFs of other metals in these two peatlands, which were almost lower than 5. The low EFs of metals in the eastern sites means that metal deposition in this area was less influenced by the placer gold mining than those in the western sites. This result is also supported by an increase in the anthropogenic proportions of these metals when gold mining began. Because of low anthropogenic metal production in other regions globally (Nriagu 1996), we speculate that pollutants produced by the placer gold mining and residents living near the mine were the major anthropogenic sources in the studied region in this period. Gold production around the 1900s only caused the Cu_{EF} values in the western sites (especially LGH) to be markedly higher than they were during the preceding periods. With increasing distance from the peatland to the gold placer, the decreasing effect from the placer gold mining causes the EFs of metals in the peatlands to gradually decrease.

During the 1920s, as the Pb and Cu fluxes in the TQ and LGH peatlands decreased, the Pb and Cu fluxes were similar to the eastern sites, and natural sources of metal fluxes contributed to most of the total metal fluxes. The EFs of metals in the LGH and TQ peatlands decreased from 1930 to 1050, and the minimum EF values appeared in the 1940s. After 1930, the Pb and Cu fluxes in HY and HT increased markedly and were higher than those in western sites. Similar to the Ti fluxes, high pollution metal fluxes also appeared in both sites during these periods, and more markedly in eastern region. With the increasing of local human activities, more forest fires occurred in the north of Great Hinggan Mountain and more dust were released to the surrounding natural ecosystems after the 1900s (Gao et al. 2018b). The increasing dust deposited into peatland was speculated as major factor that increased Ti fluxes during this period. In the HY and HT peatlands, the increasing Cu and Pb fluxes were accompanied by large amounts of Ti deposited in this region, and means local forest fire was an important metal source in the northern Great Hinggan Mountains after the 1900s. Because the Great Hinggan Mountains are located along the margin of the summer monsoon, the eastern sites of the Great Hinggan Mountains are also influenced by a stronger Asian summer monsoon and weaker westerlies than the western sites (Chen et al. 2015; Gao et al. 2018a). Japan and Russia invaded and exploited the natural resources in northeastern China at the beginning of the twentieth century, and several wars (e.g., the Second World War) also occurred from 1920 to 1950 (Zhang et al. 2006). High degree of pollutants produced by anthropogenic sources in northeastern China could be transported by the Asian summer monsoon and more easily deposited into the eastern regions of Great Hinggan Mountains, and finally caused Cu and Pb deposition fluxes in HT and HY peatland increased markedly from 1900 to 1950.

After the People's Republic of China was established in 1949, the Korean War and industrial development without any environmentally friendly policies and high frequency of fire still produced limited amounts of pollutants from 1950 to 1980 (Gao et al. 2018b). Mining processes in Yanzhigou gold placer were changed from manual to mechanical (e.g., mechanical gold dredger), more dust released to the surrounding ecosystem accompanied with increasing gold production. Similar with 1900–1950, regional industrial development, forest fire, and dust released from the gold placer were the important anthropogenic metal sources in the northern Great Hinggan Mountains. Unlike 1900–1950, the degree of anthropogenic pollutions in the northern Great Hinggan Mountains was stopped increasing from 1950 to 1980 and deposition fluxes of selected metals were markedly higher than previous period. And, the metal deposition fluxes in the eastern sites were markedly higher than those in the western sites. With the degree of regional human activities increasing, more anthropogenic metals deposited led several clear peaks of Cu_{EF} and Zn_{EF} appeared after 1960. Except LGH peatland, the EFs of metals in the other three peatlands increased markedly after 1960 and more pronounced in the eastern region.

After 1976, the Chinese government began to implement forestry law and control the intensity of forest resource exploitation (Liu 2001). With the implementation of environmentally friendly policies in the 1980s, the frequency of forest fire decreased markedly (Gao et al. 2018b), and the productions of Pb and Cu produced by natural sources may also decreased. However, with the development of regional and global industries, more metals were produced from anthropogenic sources and the increasing trend of metal deposition fluxes was found in the middle of Great Hinggan Mountains around the 1970s (Bao et al. 2012, 2015, 2016). The largest peak of Cu_{EF} appeared in the 1990s in HY peatland, and the variations in Cu_{EF} were more pronounced than Pb_{EF}. After 1980, the Pb_{EF} values in LGH peatland also began to increase. Thus, similar with the middle of Great Hinggan Mountain, the greater amounts of metals produced from developing regional industry became the most important sources of these metals in the studied region after 1980. Increasing regional anthropogenic sources likely caused the anthropogenic proportions of these metals to increase in the Great Hinggan Mountains and more markedly in the eastern regions.

After 2000, the Zn_{EF} values increased faster than those of the other two metals and were the highest overall. Similar to Zn_{EF}, the Pb_{EF} values in the HT and TQ peatlands also increased markedly and became slightly higher than the Cu_{EF} values after 2000. The increasing Pb_{EF} values and high Zn_{EF} values observed in most of the selected sites indicated that more Pb and Zn were produced by regional or even global anthropogenic sources and that anthropogenic proportions gradually increased in recent years. Thus, high EF values of metals indicate that metals were mainly from anthropogenic sources in this period in the northern Great Hinggan Mountains. The widely use of gasoline without Pb in northeastern China was speculate as one of the important factors that caused the anthropogenic sources of Pb to gradually decrease and the total Pb fluxes four peat cores to markedly decrease. With the implementation of environmental friendly policies and gold production process improvement, dusts and pollutants which were produced from the placer gold mining were also decreased. As the emissions of local pollutants decreased, pollutants were mainly derived from regional or even global anthropogenic sources, and the differences in the metal fluxes between the western and eastern sites of the northern Great Hinggan Mountains gradually decreased. After 2000, the Pb and Cu fluxes in all four peatlands were similar and around 4 and 3 $mg/m^2 \cdot year$, respectively. Both of these metal fluxes were a bit lower than those when the local placer gold mining first began around 1900, and far lower than those when lots of regional pollutants produced from anthropogenic sources between 1950 and 1980.

Overall, the different locations of the four peatlands were the major reason why the historical variations in metal fluxes at these sites were different during the last 150 years. When the placer gold mining began to produce gold around the 1900s, the metal fluxes in the western peatland increased markedly. After 1900, regional wars and economic development without any environmentally friendly policies caused the metal fluxes in the eastern sites to increase markedly until 1950. From 1950 to 1980, metal fluxes in the eastern sites fluctuated and were obviously higher than those in the western sites. With the implementation of environmentally friendly policies and gold production process improvement, less pollutants were produced from local anthropogenic sources which led the metal fluxes to decrease the same with the four peatlands in the northern Great Hinggan Mountains.

Spatial and temporal change in Pb sources

The Pb deposition fluxes and Pb_{EF} values observed in the four peatlands indicate that the distance between the peatlands and the gold placer led to historical variations in the Pb deposition fluxes in these peatlands over the last 150 years. These changes were also due to changes in industrial activities within this region and to differences in the transport of atmospheric pollutants. Although Pb isotope ratios could not quantify the proportions of potential Pb sources the same with several Pb sources, obvious differences of Pb isotope ratios between most of the potential Pb sources lead Pb isotope ratios which have been widely used to identify historical sources of Pb and to evaluate the influence of the different anthropogenic sources of pollutants on natural ecosystems (Leorri et al. 2014; Walraven et al. 2014). In the gold placers, the $^{208}Pb/^{207}Pb$ ratios range from 2.07 to 2.11, with an average value of 2.08; the $^{208}Pb/^{206}Pb$ ratios range from 2.45 to 2.50, with an average value of 2.46 (summarized by Chen et al. 2006). To reveal the historical sources of Pb in northern Great Hinggan Mountains, we compared the Pb isotope ratios in the placer gold mining and those in different Pb sources around the world (Sangster et al. 2000; Bollhöfer and Rosman 2001; Komárek et al. 2008; Cheng and Hu 2010). Based on the history of placer gold mining and regional human activities in the northern Great Hinggan Mountains, the historical variations in Pb isotope ratios in the four peatlands could be divided into four different periods (i.e., before 1900, 1900 to 1950, 1950 to 1980, after 1980) over the last 150 years as shown in Fig. 6.

Coal and raw ore began to be produced and consumed in the nineteenth century in Germany and other developed countries in Europe; consumption increased markedly after 1900 (Lavanchy et al. 1999). Because little coal and raw ore were consumed in northeastern China around the 1900s, gold mining represented the major potential local source of Pb. In the LGH peatland, Pb isotopes in the deeper layers of the peat core are interpreted to have been deposited before 1900, and their Pb isotopes indicate that the main source of Pb was the surrounding gold placers around the 1900s (Fig. 6). Unlike the

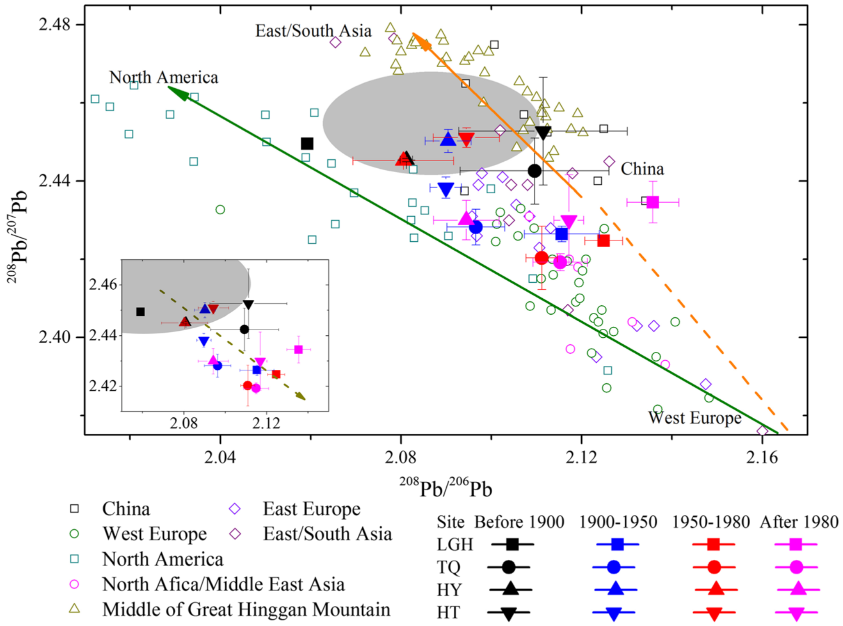

Fig. 6 Historical variations in the ^{208}Pb/^{206}Pb and ^{208}Pb/^{207}Pb ratios in the LGH, TQ, HY, and HT peatlands during different periods over the last 150 years, as well as ^{208}Pb/^{206}Pb and ^{208}Pb/^{207}Pb ratios in the middle of the Great Hinggan Mountains during different periods and atmospheric dust around the world (Bao et al. 2015; Bollhöfer and Rosman 2001; Cheng and Hu 2010; Komárek et al. 2008; Sangster et al. 2000). The error bars for data present standard deviation of all data points in each period. The gray ellipse indicates the range of ^{208}Pb/^{206}Pb and ^{208}Pb/^{207}Pb ratios of rock and placer in the Yanzhigou gold mine and its surrounding region (Chen et al. 2006)

Pb isotope ratios in LGH, the ^{208}Pb/^{207}Pb ratios in TQ were lower than those in LGH at the beginning of the 1900s. They were similar to the lowest values of the ^{208}Pb/^{207}Pb ratios observed in the gold placers. The low ^{208}Pb/^{207}Pb ratios observed in TQ before 1900 indicated that the main source of Pb was the placer gold mining, although some of these Pb may have been derived from West Europe and China. Except for local placer gold mining, Pb produced from East/South Asia were also close to Pb isotope in the HT peatland, and this source is also assumed as a major source of Pb in the eastern regions before 1900. Due to the major sources of Pb in northern Great Hinggan Mountains which were from the gold placers and more metals were deposited into the surrounding sites (i.e., LGH and TQ), the Pb deposition fluxes and Pb$_{EF}$ in LGH and TQ were obviously higher than those in HT and HY before 1900 when the gold placers started to produce gold.

As the global consumption of fossil fuels increased during the periods of industrial development and the Second World War between 1900 and 1950, Pb produced from North America and West Europe not only caused Pb deposition fluxes in Europe and North America to markedly increase (Wieder et al. 1994; Novák et al. 2003), but also influenced the deposition fluxes of Pb globally. In the eastern region, the Pb isotope ratios in these two peatlands were closer to those of the Pb sources in North America than they were to those in West Europe during this period. While, the peatlands in the western regions (i.e., LGH and TQ) were more easily influenced by Pb produced from the western regions, and the Pb isotope ratios in the western regions were more close to those of West Europe than those in the eastern regions, such as the Pb isotope ratios in the LGH peatland became more closer to those of the sources in China and West Europe after 1900. Thus, besides the Pb produced from the local placer gold mining, Pb produced from Europe and North America became the important anthropogenic sources of Pb in the northern Great Hinggan Mountains between 1900 and 1950. In this period, anthropogenic sources of Pb from other regions around the world started to increase the Pb deposition fluxes in the northern Great Hinggan Mountains. At the same time, the natural productions of metals also increased which were caused by the high frequency of forest fire (Gao et al. 2018b); thus, the deposition fluxes of metals started to markedly increased in studied region, especially in HY and HT.

With the continued development of industry and economics in 1950–1980, Pb emissions from traffic in Europe continued to increase from 1950 to 1980 (Schwikowski et al. 2004). More Pb produced from Europe deposited in the northern Great Hinggan Mountains, and the Pb isotope ratios in TQ and LGH during this period were similar to those produced from West Europe. Because several mountain peaks (with altitudes of higher than 1000 m) exist between western and eastern regions, the Pb produced from Europe weakly influenced the Pb isotope ratios in the eastern sites. In addition to the influence of the placer gold mining, the values of Pb isotope ratios in HT and HY were also influenced by those produced from the North American Pb sources. Pb isotope ratios in eastern regions were similar to those in the gold placers and Pb sources in North America from 1950 to 1980. While, the differences in the Pb isotopes in the eastern and western sites were more clear than before 1950, which means that more Pb produced from Europe or North America caused the impact of Pb from these regions in the northern Great Hinggan

Mountains to be more serious between 1950 and 1980. Although the increasing frequency of forest fire increased the natural Pb production, the obvious increase of global anthropogenic Pb production leads the anthropogenic proportion of Pb in the northern Great Hinggan Mountains to increase and Pb deposition fluxes were obviously higher than other periods.

As Pb production was controlled after 1980 in Europe and North America, less Pb were produced from these regions and deposited in the natural ecosystem (Nriagu 1996). Since the 1980s up to the present, the ^{208}Pb/^{206}Pb ratios increased and ^{207}Pb/^{206}Pb ratios decreased in the HY peatland. The Pb isotope ratios in the HY peatland were clearly different with those in the gold placers, and they began to be influenced by Pb that was from Pb sources in South/East Asia. Similar variations in Pb isotope ratios were also found in site HT; while, the ratios in the HT peatland were closer to those from China, which indicated that Pb produced in China was a major source of Pb in the HT peatland. With the frequency of forest fire decreasing after 1980 (Gao et al. 2018b), the natural Pb production also started to decrease. Although the increasing of Pb_{EF} means that most of Pb were produced from anthropogenic sources, while, with global anthropogenic and natural Pb production decreasing, the Pb deposition fluxes in HT and HY peatland started to decrease after 1980. Pb isotope ratios in LGH and TQ were similar and both close to those of Pb produced from China or South/East Asia. The Asian summer monsoon could transport water and pollutants from southern China and South/East Asia to northeastern China (Liu et al. 2015). With increasing Pb produced from China and East/South Asia and decreasing Pb production from Europe after 1980, Pb produced from China and East/South Asia became the major Pb sources that influenced the deposition process in the northern Great Hinggan Mountains. Thus, more metals were produced from the developing industry in China and regional human activities were speculated as the major reason that increased the EFs of metals in LGH after 1980.

Shortly, except the local placer gold mining, Pb that was produced in other regions around the world and transported through the atmosphere was also an important source of Pb in the northern Great Hinggan Mountains. At the beginning of the 1900s, because the global Pb production was lower, Pb from the placer gold mining was the major sources of Pb in all four peatlands and caused the Pb fluxes in the western region to be higher than those in the eastern region. With regional or global Pb production increasing from 1900 to 1980, the proportion of Pb anthropogenic sources in the western and eastern sites was markedly different, and eastern sites were more easily influenced by global Pb productions, especially in West Europe and America. After 1980, the contribution of Pb from the placer gold mining was decreased and Pb production from China and South/East Asia gradually became the major sources of Pb in the northern Great Hinggan Mountains.

Conclusion

Based on Pb isotopes and metal deposition fluxes in four peatland cores, historical variations in deposition fluxes and the sources of metals in the northern Great Hinggan Mountains were reconstructed in this study. After placer gold mining began at the end of the nineteenth century, the metal deposition fluxes in the peatlands in the northern Great Hinggan Mountains began to be influenced by the placer gold mining. The placer gold mining caused the EFs and anthropogenic deposition fluxes of Pb and Cu in the western sites to be markedly higher than those in the eastern sites around the 1900s. With increasing distance between the gold placers and peatland cores, the impact of metals produced from the placer gold mining on peatland decreased. After 1900, the metals produced by regional and global anthropogenic sources increased and became major sources of metals in the northern Great Hinggan Mountains, which more strongly influenced the eastern two sites than others. Increasing sources of pollutants in China caused the metal deposition fluxes in the eastern sites to be higher than those in the western sites between 1950 and 1980. After 2000, with the gradual implementation of environmentally friendly policies around the world, global anthropogenic metal productions gradually decreased, which led the major sources of metals in the northern Great Hinggan Mountains to originate from China, and deposition fluxes of Cu, Pb, and Zn began to decrease after 2000.

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