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Historical trends of atmospheric black carbon on Sanjiang Plain as reconstructed from a 150-year peat record

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Black carbon (BC), one of the major components of atmosphere aerosol, could be the second dominant driver of climate change. We reconstructed historical trend of BC fluxes in Sanjiang Plain (Northeast China) through peat record to better understand its long-term trend and relationship of this atmosphere aerosol with intensity of human activities. The BC fluxes in peatland were higher than other sedimentary archives. Although global biomass burning decreased in last 150 years, regional large scale reclaiming caused BC fluxes of the Sanjiang Plain increased dramatically between 1950s' and 1980s', most likely resulting from using fire to clearing dense pastures and forests for reclaiming. The BC fluxes have increased since 1900s with increasing of the population and the area of farmland; the increase trend has been more clearly since 1980s. Based on Generalized additive models (GAM), the proportional influence of regional anthropogenic impacts have increased and became dominant factors on BC deposition.

Black carbon (BC), produced by incomplete combustion of fossil fuels or biomass, is widespread in environment and influences the biogeochemical process of ecosystem[1]. Global black carbon was emitted by 50–270 C Tg/a as residues of vegetation fires[2], and 4.4 C Tg/a from fossil fuel consumption in 2000 AD with a liner increase trend[3]. Nearly 18.97% of the global BC emissions were from China with 1.88 C Tg/a in 2009 AD[4]. BC contents in atmosphere were generally high with 2 ug C.m^{-3} in Beijing and Shanghai (China), which was nearly 2.5 times of that in Korea Climate Observatory-Gosan[5]. High contents of BC in China aerosol could be transported via the atmosphere to other countries, likely including the United States[6]. As an important component of the atmospheric aerosol, BC is capable of changing the climate[7] and transporting of POPs[8]. After few days of staying in atmosphere[9], BC generally deposits into the soil and could be stored as soil carbon pool for several thousand years[10]. Cong et al.[11] found that BC in Nam Co Lake region (Tibetan Plateau, China) was most likely influenced by economic development in South Asia. In Changbai Mountain (Northeastern of China), local BC emission strongly influences the level and gradient of BC in the snowpack[12]. Peatland, with slow degradation in anaerobic environment[13] and continuous deposition process[14], is an ideal archive for recording the BC deposition history. Historical anthropogenic sources (e.g. residential, biomass burning) and natural sources (e.g. wildfire) caused more BC emitted and raised BC deposition fluxes[2,3]. While, land drained and smoldering might lead BC modified in peat soils[15,16]. It is necessary to study historical BC deposition progress and its influence factors in peatland, while, few studies had been reported.

Sanjiang Plain, located at the northeast of China with prolonged winter of low temperature, had been reclaimed since 1895 when the Qing government began to encourage people migration to this region and exploited the northeastern China[17]. Slash-and-burn of pastures and forests during reclamation and burning firewood or coal during winter potentially emitted large amounts of BC. The area of arable land was increased by 2.5 times from 1954 to 2005 and has become one of the most heavy land use for crops (55%) in China[18]. In addition, residents in this region primarily use coal for heating in cold winter, resulting in large amounts of BC produced. The population in Heilongjiang Province around this area increased by 30 times from 1.27 million in 1897[19] to 38.34 million in 2011[20], as a result, the human effects were violently prevalent in the last 150 years. With the industrial and transportation developing, BC emitted from urban were further increased. Sanjiang Plain is an
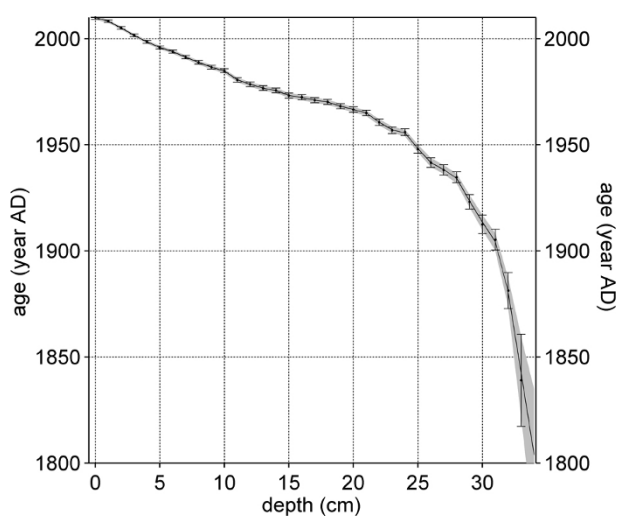
Figure 1 | Sediment dating results. Depth versus age plot calculated by the CRS model.

ideal study area for study the relationship of regional human activities with BC deposition because of its clear and great change of BC emitted from anthropogenic sources.

However, the information and knowledge of historical trend of BC fluxes of the Sanjiang Plain are very scarce. Peatland in this area serves as ideal archives for exploring environment changes. In this study, we presented the first data on BC deposition fluxes in the last 150 years in temperate peat soils in Northeast China obtained with the acid dichromate oxidation method. Based on variability of BC fluxes in last 150 years, we aim to reveal its links with regional anthropogenic emission through statistical analysis. Generalized additive models (GAM) were used to identify the effect of regional economic development (Heilongjiang Province) on BC deposition fluxes in Sanjiang Plain.

Results and Discussion

Peat chronology and BC contents. For the SJD peatland, ages and sedimentation rates were calculated using the CRSModel[21] in the MATLAB 2012a software. The uncertainty for the time scale were returns 95% confidence intervals from 1000 chronologies using sediment ages selected at random, based on the estimated age and standard deviation[21]. The sedimentary record covered about 150 years to reach back to 1850s (Fig. 1). The reliability of the profile dating was acceptable based on a constant decline in ^{210}Pb with increasing depth. Slight variations of density with depth were observed for profiles (0.27 ± 0.06 g/cm^3). The mean sedimentary rate and peat accumulation rate were 2.1 mm.a^{-1} and 0.13 g.cm^{-2}.a^{-1}, respectively. Sedimentary rates were higher in recent years, especially after 1950s with 4.6 mm.a^{-1}. The mean content of BC in the SJD peat profile was 24.3 mg C/g with a large variation between the maximum (61.2 mg C/g) and minimum (3.2 mg C/g). The range of BC content

in SJD peatland was similar with that in Switzerland fen (<1 to 93 mg C/g) which measured with Differential Scanning Calorimeter[15]. Compared to other soil types (e.g. forest soils, sediments, loess), contents of BC in peat soils were the highest, with up to hundred times higher than those in the sediments (Table 1). The BC content in peat soils was higher than that in other types of soils, and was an important stable component of the global soil carbon pool.

BC deposition flux and its historical trends. Compared to other deposition archives (e.g. lake sediments, loess), mean deposition flux of BC in SJD peatland was the highest, with up to hundred times higher than those in the other deposition archives (Table 1). Relatively similar deposition fluxes were found in forest soils in Amazon, where the frequent occurrence of fire could happen[22]. However, the difference between peatland and other deposition archives was that wildfire may occur on the surface of peatland because of high organic carbon content. Charcoal research showed that most of charcoal produced by surface wildfire was deposited locally, and little could be transmitted to other places through atmosphere[27], thus could result in the higher BC deposition fluxes in peatland than that in other archives. In addition to local fire, the short distance between urban and study area may be another factor that caused the mean fluxes of BC was higher. BC fluxes in same deposition archives with difference of distance from cities were different. For lake sediments, the BC fluxes in Taihu Lake (surrounded by a number of large cities) are higher than that in Nam Co Lake (far away from cities)[11,24]. The previous results indicate that BC produced by human actives could greatly affect the fluxes in archives (e.g. BC in Nam Co Lake region was most likely transported from South Asia)[11]. In the present study, the nearby city Jiamusi (a major city of Heilongjiang Province), many surrounding villages and a close by major railway could greatly influence the BC fluxes in the profile; combined influences of anthropogenic sources and natural wildfires likely increased the BC deposition fluxes in SJD peatland and resulted in the higher rates than those of other studies.

Historical trend of BC deposition fluxes in the SJD peatland was increased in the last 150 years with several peaks. At the beginning of 1900s and 1930s, small peak values (1.6 mg C.cm^{-2}.a^{-1} and 2.9 mg C.cm^{-2}.a^{-1}, respectively) appeared in those periods (Fig. 2). Dramatic changes in the BC fluxes occurred between 1950s and 1980s. During that period, the range of BC fluxes were from 0.7 to 11.1 mg C.cm^{-2}.a^{-1}, and considerably higher than the other periods. After 1980s, the BC fluxes were relatively stable, but consistently climbed from 0.4 to 4.1 mg C.cm^{-2}.a^{-1}. The background fluxes of BC which obtained from Charanalysis 1.1 could be considered as the trend of the regional BC deposition. By comparing the background of BC fluxes with different years smooth, the results obtained from the 10 years smooth were sensitive and influenced by each peak values in original trend, and trend obtained from the 20 years was smooth and could not be used to reflect historical events that caused BC fluxes changed dramatic. The results obtained from the 15 years smooth were better than other smooth years to reflect the regional BC emission change. Overall, the trend of the regional BC deposition fluxes

Table 1 | The Black Carbon Contents (mg C·g^{-1}) and Deposition Fluxes (mg C·cm^{-2}·a^{-1}) for different deposition archives

| Types	Description	Contents (mg C/g)	Fluxes (mg C/cm².a)	Methods
Peatland	Sanjiang Plain, China	3.2–61.2	0.4–11.2	Acid Dichromate Oxidation
Forest soils[22]	Amazon, Brazilian	1–27	0.3–9	Acid Dichromate Oxidation
Loess[23]	Loess Plateau, China	1–5	0.01–0.3	Acid Dichromate Oxidation
Lake sediments[24] (near urban)	Taihu, China	0.43–1.95	0.12–0.69	TOT/R
Lake sediments[11]	Tibetan Plateau, China	0.49–1.09	0.012–0.044	TOT/R
continental shelf[25] (near urban)	Palos Verdes Shelf, American	1.2	0.7–1	CTO375
continental shelf[25]	Gulf of Marine, England	0.3–1.7	0.1–0.2	CTO375
Pelagic sediments[26]	Global	0.02–1	0.00012	infrared spectroscopy
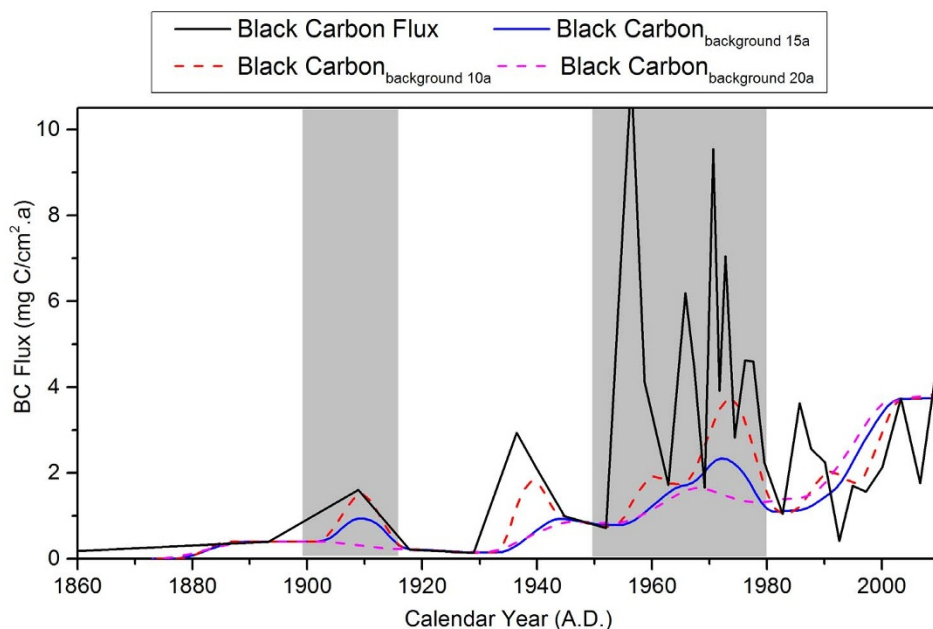
Figure 2 | Variation of BC fluxes (black line) and background fluxes with different smooth years (10 years, red dash; 15 years, blue line; 20 years, magenta dash) from 1850 to 2010 at SJD.

increased in the last 150 years, and was stable in recent years with the high values (Fig. 2).

Anthropogenic factors influenced the BC deposition. Many factors which potentially affect BC deposition fluxes might be divided into two main categories (i.e. natural and anthropogenic factors). With the climate changing, the frequency and intensity of wildfire might alter, and the BC deposition fluxes likely different[28]. Anthropogenic

factors were affected by the intensity of human activities; emission inventory was derived from different sources (i.e. industry, biomass open burning, power plant, transportation, and emission from people's daily life)[4]. The development of industry and transportation were coupled with increased energy consumption. In Heilongjiang Province, total coal consumption were 22.42 million tons, 1.84 million tons for gasoline and 0.05 million tons for diesel oil in 1995. 26.39 million tons of coal consumption, 3.19
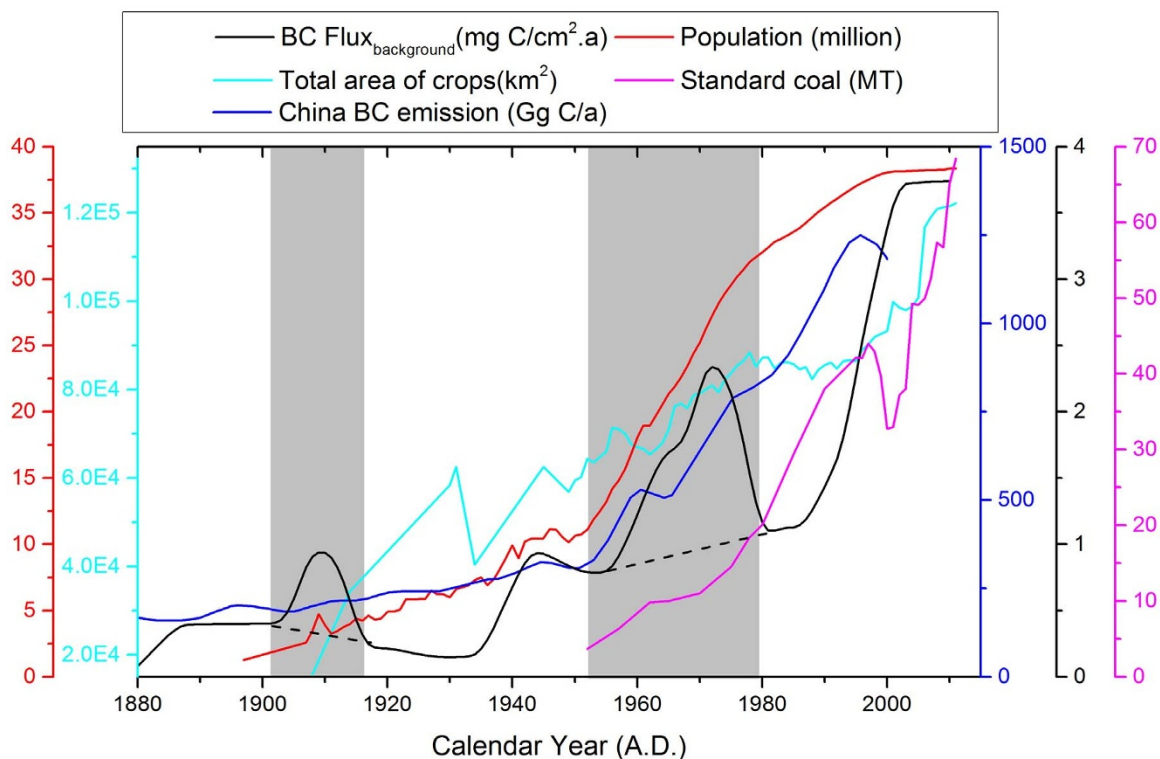

Figure 3 | Variation of BC background fluxes (black line) from 1850 to 2010 at SJD, Population (million) in HLJ province (red line)[19,31], total area of crop lands (km²) in HLJ province (cyan line)[17,31,32], coal consumption (millions of tons of standard coal) in HLJ province (magenta line)[20], and BC emission(Gg C/a) in China(blue line)[3].
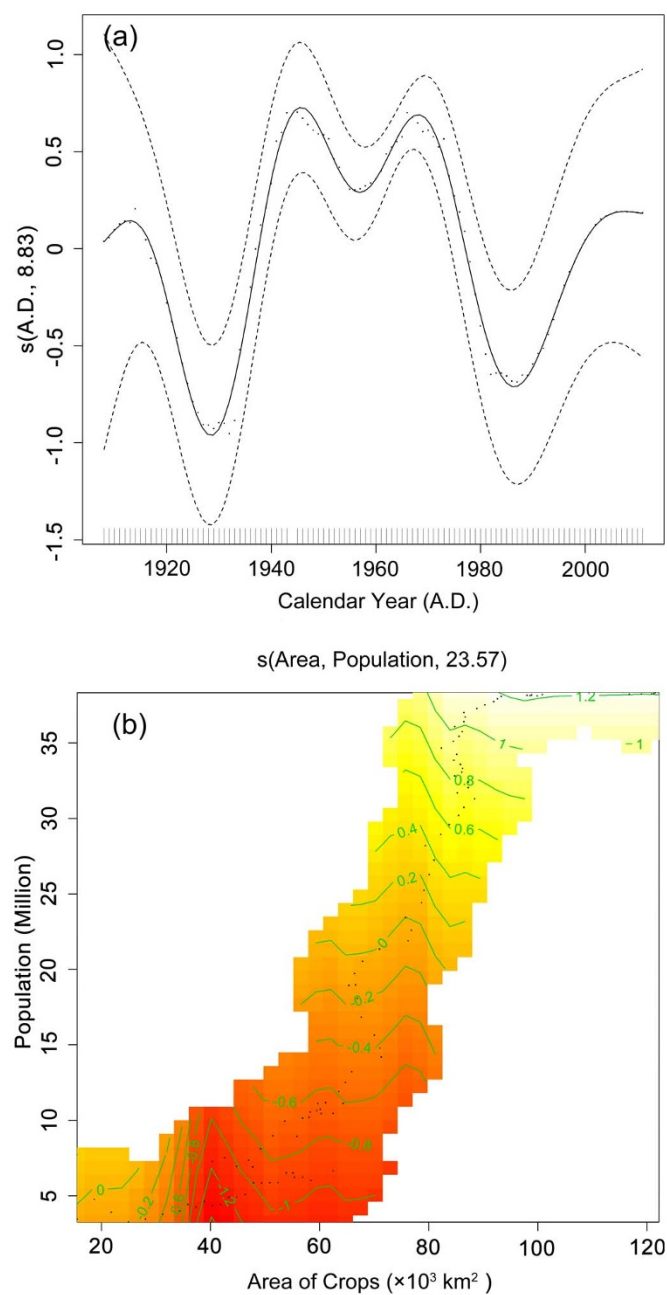
Figure 4 | **Estimated smooth terms of BC background fluxes for the GAM model.** (a) The smooth function of calendar years. The y-axis represents natural effects on BC background fluxes. The dashed lines are estimated 95% confidence intervals; (b) The smooth function of population and the area of crops. The color represents anthropogenic effects on BC background fluxes, buff were represent higher effects and red were represent lower. Green represents the contour lines of effects.

million tons for gasoline and 5.12 million tons for diesel oil in 2009[29]. Coal consumption was much higher than other types of energy consumption in Heilongjiang Province[20]; as a result, the factor of BC emission from coal consumption (raw coal 0.32 g/kg) has been much higher than any other energy consumption(crude oil 0.0396 g/kg; gases 0.001 g/kg)[4]. In this region, the coal consumption could be used as an indicator of the degree of BC emitted from industry and transportation. Biomass open burning was affected by the yield of crops[4], thus the area of crops could be treated as an indirect indicator of open biomass burning sources. In addition, the population in Heilongjiang Province was used to indicate the change of BC

emitted from residential. The background values of BC fluxes obtained from Charanalysis 1.1 were considered as regional BC fluxes changes[30]. It is better to separately reflect the historical changes of BC emission from natural and anthropogenic sources. Therefore, the area of arable land, population and coal consumption in Heilongjiang province were used to assess the anthropogenic sources change. The relationship of anthropogenic sources and background deposition fluxes of BC is shown in Figure 3.

The background BC fluxes were dramatic increased in the past 150 years. Overall, there were two periods where the fluxes of BC increased and then declined after several years later in these periods. The first period occurred at the beginning of 1900s when large number of people from south of China immigrated to this area and some natural lands were changed to farmland. By the end of this period (A.D. 1908), the area of farmlands in northeast China were four times more than before[17]. Fire was a useful tool for changing natural lands to farmland and widely used during reclaiming, which could be the main reason that caused the BC fluxes increased in this period. Furthermore, 2811.5*10³ km² (25.8% of total area) of arable land developed in Sanjiang Plain from 1954 to 1986[18] similarly caused the fluxes of BC increased clearly in the second period. Compared the area of farmland and the BC fluxes, the increase in farmland always accompanied with higher BC fluxes than other periods (e.g. 1900s to 1910s, 1950s to 1980s), especially before 1980s. After the large-scale land reclamation (1954 to 1986), the deposition fluxes of BC increased were primarily resulted from the increased in the population and the coal consumption. If natural lands were not changed to farmland in these two periods, BC fluxes may had increased gradually like dash line in these two periods and shown in Fig. 3. In previous study, because the global expansion of intensive grazing, agriculture and fire management, biomass burned were reduced over the past 150 years[33]. Regional anthropogenic sources increased and large-scale land reclamation might the major reason that cause historical trend of BC fluxes in Sanjiang Plain and global biomass burned trend were opposite to each other. The trend of population was more closely related with BC fluxes than coal consumption probably due to that the local industry was not well developed and the anthropogenic BC emission was largely from open crop biomass burning and residential. The degree of BC emitted from residents was dominated by increasing population; thus, the BC fluxes trend were closer to population change. The total BC emitted in China in last 150 years greatly increased[3]. If there was no period of large scale reclaiming in the region, the deposition trend of BC in the SJD profile should had been similar to the overall BC emission. It suggested that the trend of BC deposition had a close relationship with human activities in last 150 years. The changes of BC emitted from increasing human activities may lead the BC fluxes change.

BC Fluxes Link with natural and anthropogenic emission. The above mentioned suggests that population and the area of arable land could be regarded as major indicators of the degree of anthropogenic BC emission. The change of calendar years might be regarded as natural factors (e.g. wildfire) and other uncertain factors (e.g. BC transported through global atmosphere circle and deposited into study area) that affected BC emission. The time, population and area of arable land were regarded as initial parameters to fit a preliminary base model. Missing data were obtained by linear interpolation and the preliminary model could be written as:

$$\log(BC) = s(time) + s(population) + s(area)$$

where time is the calendar year from 1900 to 2010, BC is the background value of BC deposition fluxes (mg C. cm^{-2}.a^{-1}). The units of population and area of arable land were million and square kilometer, respectively. Final model was obtained with the model based on these parameters that have the lowest AIC value. The final model with 97.7% deviance explained could be written as:
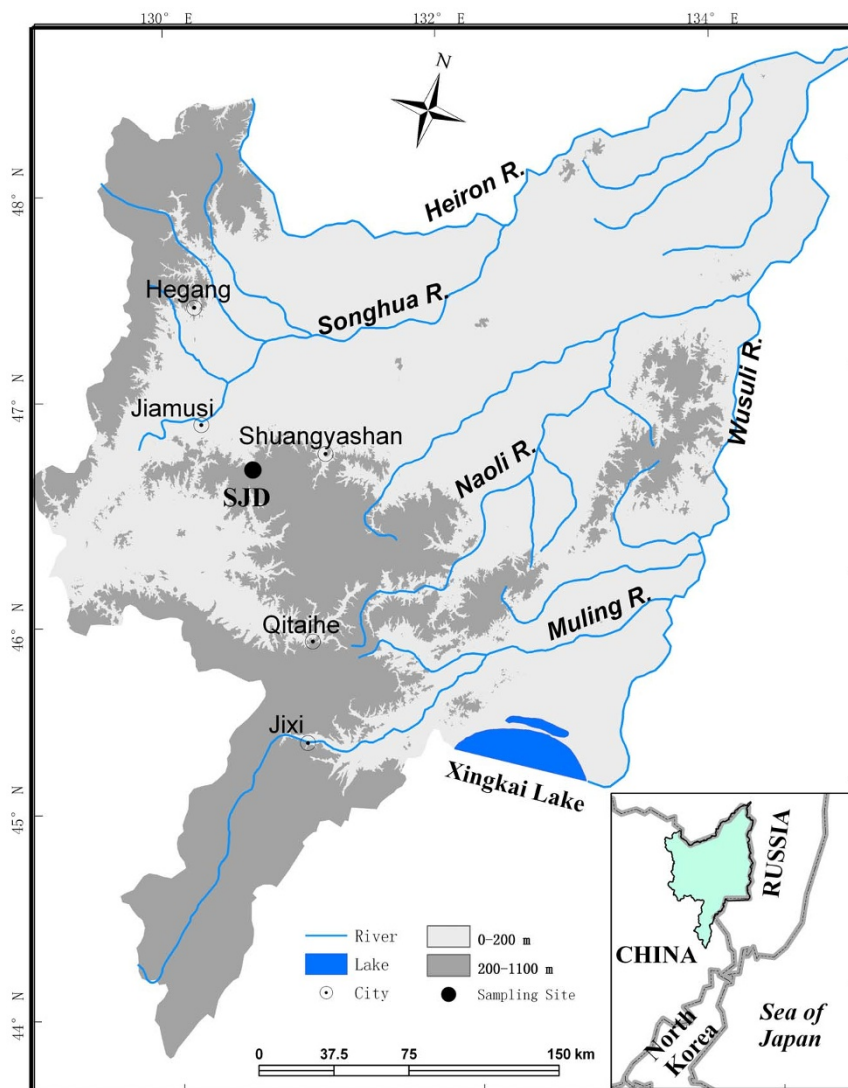
Figure 5 | The site of study area. The figure was created using the results of remote sensing interpretation[18], and generated by Chuanyu Gao, using ArcMap 10.0.

$$\log(BC) = s(time) + s(population + area)$$

No other refinements made any significant improvements to the models and the results obtained by GAM were shown in Fig. 4. Calendar year ($F = 118.314$, $P < 0.001$) and anthropogenic factors ($F = 5.407$, $P < 0.001$) were found to have significant effects on the deposition of BC. Curve of calendar years and its effects on background fluxes of BC was shown in Fig. 4a, which decreased from 1900 to 1930s, and increased in the follow 40 years, with slightly fluctuation around 1960s. At the beginning of 1970s, the curve was began to decreased and kept nearly 20 years. And then, from 1990s to the present, the curve was raised again. With the population and the area of arable land increasing, the effect on background fluxes of BC were increased clearly (Fig. 4b). From 1950s to 1970s, the annual sunshine hours were higher than other periods during 1950 to 2000s. Since the 1980s, the annuals sunshine hours have reduced clearly. What's more, there were three periods during last one hundred years with low precipitation (1920s, 1940s, and 1970s). After 1970s, the precipitation increased and lasted nearly 20 years. From the end of 1990s to the present, the precipitation reduced again[34]. Higher annual sunshine hours and lower precipitation might increase the probability of occurrence of wildfire and lead BC fluxes raised. Historical trend of

the meteorological data in northeast China was similar with the trend of calendar years effects on background BC fluxes. The trend of year effects on BC fluxes could be used to indicate the impact of climate change on the BC fluxes. Compared the curve trend of calendar years and its effects during last 100 years obtained from GAM and the original trend of BC background deposition fluxes (Fig. 3 and Fig. 4a), there were similar before 1960s, and became different after 1960s. With the pass of time, the difference between them became more clearly. This result showed that the impact of natural factors on BC deposition fluxes decreased; anthropogenic factors were gradually increased and became the dominant factor in the recent 30 years.

Methods

Site description and sampling. Peat samplings were carried out in peatlands of Sanjiang Plain, Northeast China (Fig. 5). Detail information of SJD peatland (46°34.864′ N; 130°39.873′ E) were shown in Gao et al.[35]. The present vegetation in the sampling sites includes *Betula fruticosa-Calamagrostis angustifolia* community. A nearest major city Jamusi is only 34 km away, thus human activities might greatly influence the peatland. In Nov 2010, Samples were taken by digging profile. In order to estimate influencing by post deposition progresses (land drained and smoldering), peat profile without drained and ash layer was chosen. Furthermore, samples were collected away from the road to minimize possibly immediate effects of local human activities near the profile. The types of soils were brown peat in upper soils. Samples were sliced into 0.01 m sections using stainless steel knife, stored at a sealed polyethylene plastic bag and brought to the lab for farther analysis. Samples were loosely disaggregated to facilitate air-drying at 20°C.

Chronology. The reconstruction of recent (last 150 years) deposition is frequently based on ^{210}Pb data peat cores[14,36,37]. Profile samples were analyzed for ^{210}Pb by measuring the gamma ray emission of the samples on the high-pure germanium semiconductor and low-background gamma spectrometer (OTEC Instruments Ltd. USA). The sediment accumulation rates were calculated from ^{210}Pb profiles and physical properties of sediment. A constant rate of supply (CRS) model was used to determine dates based on ^{210}Pb counts[38]. Thirty two samples were selected for the analysis of chronology at the State Key Laboratory of Lake Science and Environment, Nanjing Institute of Geography and Limnology, CAS.

Black carbon. In this study, we used the dichromate oxidation method developed by Song *et al.*[39] to measure the BC in peat soils. The first step was to remove inorganic carbon; the predetermined amount (1 g) of a sample was treated for 20 h in 10 ml 1 mol/L HCl acid contained in plastic centrifuge bottles. After digestion, the contents were centrifuged and the residue was added with HCl (3 mol/L) + HF (22 mol/L) mixture (10 ml) at a volumetric ratio of 1 : 2 for 20 h. Then the samples were centrifuged and the residue was soaked in 1 mol/L HCl (10 ml) for 10 h. After the first step, the residue consists of organic matter, kerogen and BC. The second step was to remove NPOC (non-pyrogenic organic carbon) in residues. The 0.1 mol/L of NaOH (30 ml, 12 h, twice) was used to remove humic acid, and removed kerogen by $K_2Cr_2O_7$ (0.1 mol/L) and H_2SO_4 (2 mol/L) mixed solution (60 h, and keep mixture stay yellow). All steps were treated in 55°C bath[40]. Quantified the residual carbon as BC by using a continuous-flow isotope ratio mass spectrometer (CF-IRMS) at the Analysis and Test Center of Northeast Institute of Geography and Agroecology, Chinese Academy of Sciences. The CF-IRMS system consists of an EA (Flash 2000 series) coupled to a Finnigan MAT 253 mass spectrometer. The combustion temperature was set at 960°C. Standard samples with known carbon contents (IRMS certified reference: BN/132357) were used to calibrate the measurement and to monitor the working conditions. The content of BC in wood char[41] tested by new measurement (48.7 ~ 50.2%, n = 3) was similar to what (48.4 ~ 55.8%, n = 4) had been tested with original dichromate oxidation method in different laboratories[1].

Background values. CharAnalysis version 1.1 was used to identify the background deposition fluxes of BC[42,43]. The results of smooth from different modes were used to optimize background values, and to indicate regional fire events in reconstructing fire history through charcoal records[44]. Deposition fluxes of charcoal and BC could be affected by both local and regional emission. The trend of regional BC emission in last 150 years was derived from the background value of BC deposition fluxes. The appropriate reciprocal of dry bulk density was used to calculate the volume of samples. Moving modes with 1 year interpolate were used to estimate background value (low-frequency) of BC fluxes.

Generalized additive models (GAM). GAM are regression models where smoothing splines were used for covariates[45], and widely used in handling the complex non-linearity relationship[46]. In this study, GAM was used to identify the factor that may affect BC deposition fluxes in the SJD profiles. Smoothing parameters were selected by generalized cross-validation (GCV) in semi-parametric regression; the residual distribution was assumed to be Gaussian distribution[47]. This model was conducted by using the 'gam' function in the R environment for statistical computing[48] with the package 'mgcv'[46]. The variables in the final models were fitted to yield a lower Akaike's Information Criteria (AIC)[49].

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G.C. and W.G. conceived and conducted the experiments, G.C. and L.Q. designed and carried out the data analysis and co-wrote the paper together with Z.S., H.J. and L.X. All co-authors were involved in sample and provided helpful feedback for the paper.

Additional information

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