Decoupling of Climatic Drying and Asian Dust Export During the Holocene

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Key Points:
- New proxy evidence shows an overall decline in Asian dust export with increased eolian activity
- The long-distant export of Asian dust was decoupled from climate drying during the Holocene
- The Siberian High played a central role on the magnitude of Asian dust export across the globe in the Holocene

Abstract Both paleoclimatic records and models suggest that the dust accumulation in ice cores and marine sediments, which is often regarded as a direct record of changing atmospheric dust loading and used to estimate its climatic impact, is strongly coupled to the aridity of dust source regions. However, the underlying association of this couple has not been tested directly because of the lack of continuous and well-dated independent records of eolian activity in dust source regions. Here we present a high-resolution multiproxy record of the Holocene eolian activity and climate changes from Lake Xiarinur in north China, a major Asian dust source region. Our data, together with the records of Asian dust accumulation in downwind areas, including the North Pacific Ocean and Greenland, suggest a decoupling between long-distant Asian dust export and climatic drying in the Holocene. Although the climate was humid and the eolian activity was weak in the source region during the early Holocene, the dust accumulation in the North Pacific Ocean and Greenland was relatively high. Similarly, while climatic drying and strong eolian activity occurred in north China during the late Holocene, the rate of dust accumulation in the downwind areas was low. The long-distant export of Asian dust is closely correlated with the strength of the Siberian High rather than the westerlies as widely believed before. The rate of dust accumulation in the GISP2 Greenland ice core is comparable in patterns with the intensity of the Siberian High forced eolian activity on both millennial and centennial time scales.

Plain Language Summary Scientific grounds: We bring new, uniquely indicative evidence to bear on the dynamics of long-distant Asian dust exports. By unifying the evidences of Holocene aeolian activity and climate changes in north China, dust accumulation in Northwest Pacific Ocean and Greenland ice, the atmospheric circulations dominated Asian dust export, we confirm a central role of the Siberian High in long-range Asian dust export, rather than climate drying and the westerlies as most previous studies suggested. These results have important implications for both accurately estimating aerosol climate forcing and predicting future impact of atmospheric dust concentration changes with climate changes. Popular appeal: Our work also provides important insight into the climatic and environmental impact of anthropogenic aerosols, an important issue of international broad interest. The central role of the atmospheric circulations in long-range Asian dust export, combined with comparisons between the environment with heavy and that without or little human interference in the Holocene, indicate that the anthropogenic activity would contribute little to the long-range Asian dust export without strong atmospheric circulations. Therefore, modern Asian anthropogenic pollutant aerosols would be poorly dispersed from the source regions, otherwise transported across the whole global.

1. Introduction

Mineral aerosol has been recognized as important global climate forcing via its effects on Earth’s radiation balance and the global hydrological and biogeochemical cycles (Booth et al., 2012; Evan et al., 2009; Sokolik & Toon, 1996). It is widely believed that climatic drying drives dust export by expanding dust source areas and increasing eolian activity in source regions (Biscaye et al., 1997; Guo et al., 2002; Rea & Leinen, 1988). This assumption is consistent with the observed concurrence of the production and export of African mineral dust with climatic drying (Middleton, 1985; Prospero & Lamb, 2003) and the coincidence of the high dust accumulation in Greenland ice cores and north Pacific sediments to the drying climate in Asian dust source regions on the glacial-interglacial time scale (Lambert et al., 2008; Porter, 2001). However, the dust accumulation in downwind ocean sediments and in ice cores does not always correspond to the degree of climatic drying.
in source regions (Lambert et al., 2008; Maher et al., 2010; Mahowald et al., 2007; Pye & Zhou, 1989), suggesting a possible overestimation of the role of climatic drying in driving dust emission and transport both regionally, and perhaps globally. The deserts of the Asian interior, including those in northern China, are among the two largest dust source regions on Earth, the dust from which accounts for ~25% of total global dust emissions (Ginoux et al., 2004) and is transported around the entire globe (Uno et al., 2009). Most importantly, the eolian deposition in the Chinese Loess Plateau is comparable with the dust accumulation in the high latitudes of both the Northern and Southern Hemispheres on orbital time scales (Lambert et al., 2008; Porter, 2001), indicating that understanding Asian dust export plays a significant role in unraveling mechanisms of global dust export. However, work on this topic has been limited due to the lack of continuous and well-dated independent records of eolian activity in the Asian dust source regions. Here we present a multiproxy lake sediment record of Holocene eolian activity and climatic changes from a major Asian dust source region. Our data, together with records of dust accumulation in downwind areas, the Northwest Pacific Ocean and Greenland, provide new insights into the dynamics of Asian dust export.

2. Materials and Methods

2.1. Samples and Methods

Lake Xiarinur (42.62°N, 115.47°E, and 1,230 m above sea level) is located in the central area of the Qtindag Sandyland (Figure 1). The lake was hydrologically closed during the studied interval indicated by the geographic characteristics. Furthermore, the lake is sensitive to the changes of both eolian activity and climate due to its location on the southern margin of present eastern Asian dust source and the northern limit of the East Asian summer monsoon (EASM).

One core (XN-A) was drilled in Lake Xiarinur in the spring of 2006. The length of the core is 286 cm. After the core was retrieved, the water left in the tube was removed by a syringe. Then, the core was cut into three segments, sealed, and transported vertically from the field to the laboratory where it was sampled. In the laboratory, the cores were split, photographed, and described on the spot. Then, the core was spliced continuously with 0.5 cm space. Sediment samples were freeze-dried, lightly disaggregated using a plastic pestle and mortar.

The grain-size distributions, dissolved salt, and authigenic carbonate oxygen isotope compositions of the lake sediments were measured for reconstructing the Holocene eolian activity and climate changes. Grain-size distribution of the samples was determined with a Malvern Mastersizer 2000 analyzer with a measurement range of 0.02–2,000 μm. The samples were pretreated with 10–20 mL of 30% H2O2 to remove organic matter and then with 10 mL of 10% HCl to remove carbonates. About 200 mL of deionized water was added, and the sample solution was kept for ~24 h to rinse acidic ions. The sample residues were finally treated with 10 mL of 0.05 M (NaPO3)6 on an ultrasonic vibrator for 10 min to facilitate dispersion before grain-size analysis. The Mastersizer 2000 automatically yields the median diameter and the percentages of the related size fractions of a sample with a relative error of less than 1%.

For carbonate oxygen isotope analysis, the sample powders were reacted with 100% phosphoric acid for 12 h under 50°C. The produced CO2 was collected by liquid nitrogen, and the isotopic ratios of 18O/16O were measured by a MAT 252 mass spectrometer. The isotopic data were reported in the conventional δ notation as per mil (‰) deviation relative to the Pee Dee belemnite standard with an uncertainty of ±0.02‰. To identify the mineral phase of carbonates in the sediments, 10 samples with various depths were selected for X-ray diffraction analysis. The results showed that the carbonates were mainly composed of calcite.

For dissolved salt concentrations, 1 g sample was weighted by a balance with a precise of 0.1 mg and transported into a 50 mL tube. Thirty milligrams of ultra-pure water was added into the tube. The tubes were put in the shaker and shaken 1 h with vibration of 50 per minute. Then the samples were filtered through 2 μm filters, which were previously weighted. The samples on the filter were dried and weighted. The concentrations of dissolve salt were calculated by the weight lost. The precision calculated by replicate samples is better than 14.8%.

2.2. Chronology

The chronology was established using a combination of 210Pb/137Cs dating (for the uppermost 50 cm of the core) and AMS 14C dating. The top sediments (50 cm) were dated by 210Pb/137Cs method. The activities of
$^{137}$Cs, $^{210}$Pb, and $^{226}$Ra were measured by counting-rays using a low-background, well-type, high-purity germanium detector (EGPC 100P-15R) supplied by Eurosystem. Each sample was packed in a 5 cm polyethylene tube for 3 weeks of storage in sealed containers to allow radioactive equilibration (Hamilton et al., 1994). Each sample was counted for 48 h. Total activity of $^{210}$Pb was determined by the gamma at 46.5 keV. Meanwhile, the supported $^{210}$Pb were determined by measuring the activities of the short-lived daughter nuclides of $^{226}$Ra, $^{214}$Pb (295 and 352 keV), and $^{214}$Bi (609 keV) for calculating excess $^{210}$Pb ($^{210}$Pb$_{excess}$). $^{137}$Cs was measured by the gamma at 662 keV. An activity standard, having the same geometry and density as the sample, was used. Details of energy and efficiency calibration methods, and of quality control, were given by Foster et al. (2005). The counting uncertainties are about 0.2 disintegrations per minute (dpm)/g for $^{210}$Pb and 0.03 dpm/g for $^{137}$Cs. Radiometric dates were calculated using the constant rate of supply $^{210}$Pb dating model (Appleby, 2002; Appleby et al., 1986; Goldberg, 1963) and corrected where appropriate using the 1963 depth determined from the $^{137}$Cs stratigraphic record as a reference level (Appleby, 2002). The result shows that the top 50 cm of the core was deposited during the interval from 1950 to 2000 with an uncertainty less than 3 years.

For the sediments in the down core, a total of 10 AMS $^{14}$C ages were obtained, 9 from total organic matter and 1 from a grass leaf. All samples were treated and analyzed in the Radiocarbon Lab of Perking University. It is well known that the organic carbon in lake sediments is composed of aquatic and nonaquatic carbons. The radiocarbon activities of aquatic carbon might be influenced by the effect of carbon reservoir in lakes, and the nonaquatic carbon might contain the recycled older carbon, both of which cause the measured ages older than the real ages of sediments.

To constrain the influence from lake carbon reservoir and old carbon, we measured the radiocarbon of organic matter in top samples and a leaf from a horizon with a depth of 246.5 cm, and three couple samples of organic matter and carbonate in the sediments. The radiocarbon age of the top samples is less than 50 years, indicating little affect from both lake carbon reservoir and old carbon in the studied lake (Figure S1 in the supporting information).
the organic matter (10.89 ± 0.13 ka) and leaf (10.52 ± 0.08 ka), and between organic matters and carbonates (Figure S1). The ages of sampled horizons were derived by the age-depth model of Blaauw with accuracy better than 70 years (Blaauw, 2010). Most importantly, all the boundaries where the climate shifts occur have independent radiocarbon age control (Figure 2).

3. Results

3.1. Hydrological Condition of Lake Xiarinur

The hydrological condition of lakes is the key for interpreting the climatic indicators of lake sediments, because the climatic implications of indictors are different under various hydrological conditions. For Lake Xiarinur, no evidence indicates that the lake has had feeding rivers and the banks of which have ever been broken. This conclusion gets further support by the covariation between the carbonate $\delta^{18}O$ ($\delta^{18}O_{\text{carb}}$) and $\delta^{13}C$ ($\delta^{13}C_{\text{carb}}$), the reliable indicator of lake hydrology (Leng et al., 2006; Li & Ku, 1997). For closed lakes, $\delta^{18}O_{\text{carb}}$ and $\delta^{13}C_{\text{carb}}$ would decrease with large freshwater input, while they would increase with a decrease in lake volume via evaporation and vapor exchange (Leng et al., 2006). For Lake Xiarinur, the $\delta^{18}O_{\text{carb}}$ displayed a covariation with the $\delta^{13}C_{\text{carb}}$ during the whole Holocene except for the interval of 7.0 to 4.6 ka B.P. (Figure S2). The decoupling between $\delta^{18}O_{\text{carb}}$ and $\delta^{13}C_{\text{carb}}$ might be associated with the slow change of hydrological condition and/or increased alkalinity. It is obvious that the rates of $\delta^{18}O_{\text{carb}}$ and $\delta^{13}C_{\text{carb}}$ changes during the interval from 7.0 to 4.6 ka B.P. were slower than the other isotopic shifts (Figure S2). Under the condition of steady state or with slow changes, dissolved inorganic carbon approaches isotopic equilibrium with atmospheric CO$_2$ (Leng et al., 1999; Li & Ku, 1997), causing little changes in $\delta^{13}C_{\text{carb}}$ (around +1‰ to +3‰). As a result, the covariation of $\delta^{18}O_{\text{carb}}$-$\delta^{13}C_{\text{carb}}$ becomes weak or disappears (Li & Ku, 1997). In addition, the increased alkalinity indicated by dissolved salt concentrations might also contribute to the decoupling between $\delta^{18}O_{\text{carb}}$ and $\delta^{13}C_{\text{carb}}$. High alkalinity may damp the response of $\delta^{13}C_{\text{carb}}$ to changes in freshwater input and lake productivity, thus influencing the extent of $\delta^{18}O_{\text{carb}}$-$\delta^{13}C_{\text{carb}}$ covariations (Li & Ku, 1997). Most importantly, the dominance of eolian deposits in the sediments of Lake Xiarinur indicated by the similarity in grain size distributions of the lake sediments with the eolian depositions (Holocene...
loess and modern dust) distinct from the clastic materials in rivers and sediments in open lakes (Figure 3) provides an additional strong test for the hydrological closure of Lake Xiarinur.

### 3.2. Indicator of Eolian Activity

Theologically, clastic materials are transported into close lakes mainly by wind and/or surface runoff. For Lake Xiarinur, the catchment areas are small, and the precipitation is low, limiting the development of strong surface runoff. Therefore, the clastic materials should be transported into the lake mainly by wind rather than by surface runoff.

To further constrain the indicator of eolian activity, we use a lognormal distribution function method based on aerodynamic and hydrodynamic mechanisms (Qin et al., 2005; Xiao et al., 2013; Yin et al., 2008) to decompose the grain size distributions (GSDs) of the sediments in Lake Xiarinur. A total of five fractions are identified from the sediments of Lake Xiarinur. Based on aerodynamic and hydrodynamic mechanisms, Fraction 1 (<2 μm), Fraction 3 (15–65 μm), and Fraction 5 (>150 μm) are closely associated with eolian activity, while Fraction 2 (2–15 μm) and Fraction 4 (65–150 μm) might be influenced by the hydrological dynamics of lakes (Qin et al., 2005; Xiao et al., 2013; Yin et al., 2008).

To demonstrate the reliability of this decomposition, we compare the GSD of our lake sediments with those of the eolian dusts, the sand dune sands around the lake, the Holocene loess in the margin of Qingdag desert, the clastic materials transported by the rivers, and the sediments in the open lakes that are located in same region with Lake Xiarinur. The GSD of Lake Xiarinur displays a multiple mode, wide kurtosis and positive skewness, with a dominant mode of 1 to 65 μm for all the sediments except for sand horizons that occurred around 2.0 ka B.P. and 0.6 ka B.P. This pattern is similar to the GSD of the eolian dust and the Holocene loess in surrounding areas, but distinct from those of the clastic materials transported by rivers and the sediments in the open lakes (Figure 3), indicating a dominance of eolian component in the sediments of Lake Xiarinur. Instead, the GSD of sand horizons is similar to that of the sand dune sands around the lake, which is dominated by a mode of 150 to 500 μm (Figure 3). These evidences support 65 μm and 150 μm, respectively, as the threshold values for differentiating the fractions of eolian dust-fall and saltated sand dune sands from other components as suggested by the aerodynamic and hydrodynamic model (Qin et al., 2005; Yin et al., 2008).

Based on the aerodynamic mechanism, Fractions 1 and 3 are mainly transported in suspension, while Fraction 5 (>150 μm) reflects the fraction supplied by saltation (Qin et al., 2005; Xiao et al., 2013; Yin et al., 2008). However, Fraction 1 might be transported from the long-distant areas and has a different origin from other eolian fractions (Prins et al., 2007; Prins & Vriend, 2007; Sun et al., 2008). Although Fractions 3 and 5 are transported by suspension and saltation, respectively, both should consistently reflect the intensity of eolian activity in source regions for two reasons: (1) saltation bombardment is an important mechanism for fine particles leaving the ground, and the efficiency of saltation bombardment is linearly proportional to the fraction of dust contained in the parent soils (Lu & Shao, 1999), and (2) the mobility and transportation of both fractions are closely related to the same forcing factors, such as soil moisture, vegetation coverage, and wind strength. Therefore, we use the flux of Fractions 3 and 5 as indicator of eolian activity. The reliability of our indicator is shown by the high correlation (R = 0.81) between our records and meteorological registers of gale days during the past 60 years (Figure 3).

Though Fraction 1 might be transported from the long-distant areas and might have a different origin from other eolian fractions, our result indicates that the flux of Fraction 1 should have the same driver as those of Fractions 3 and 5 for Lake Xiarinur. The flux of Fraction 1 displays a pattern of covariation with Fractions 3 and 5 in most cases (Figure 4). In addition, the high flux of Fraction 1 corresponds to the strong East Asian winter monsoon (EAWM) in the early Holocene, not to the strong westerlies (Figure 4), though previous studies assumed that this fraction might be transported from long-distant areas by the westerlies (Prins et al., 2007; Sun et al., 2004; Vriend & Prins, 2005). The component of Fraction 1 under strong EAWM (average value of 3.8% during 5.0–9.0 ka B.P.) is more than 2 times than the late Holocene (1.2%). These evidences consistently suggest that the flux of Fraction 1 should be closely related to the EAWM.

### 3.3. Indicator of Aridity

Changes in aridity were reconstructed from authigenic carbonate oxygen isotope compositions (δ18O_{carb}) and soluble salt contents. The oxygen isotope composition of authigenic carbonates is controlled by the
temperature and isotope compositions of the water from which it is precipitated (Craig, 1965; Horton et al., 2016; Leng & Marshall, 2004). The amplitude of $\delta^{18}O_{\text{carb}}$ changes in the record of Lake Xiarinur is 5.7‰, which falls into the range of lake carbonate oxygen isotope shifts in the monsoon regions (4.0–9.9‰) across China (Zhang, Chen, et al., 2011). Given temperature-dependent equilibrium isotope fractions between the calcite and water is approximately $-0.24$‰/°C (Craig, 1965), this large amplitude of Holocene $\delta^{18}O_{\text{carb}}$ changes (around 23.75°C shift inferred by 5.7‰ of $\delta^{18}O_{\text{carb}}$) cannot be interpreted by temperature change alone. Therefore, $\delta^{18}O_{\text{carb}}$ should be mainly controlled by the oxygen isotope composition of lake water, which is controlled by the isotope compositions of rainfall sources, temperature-precipitation relationship, and/or precipitation/evaporation ratios (Leng et al., 2006). The climate of Lake Xiarinur is dominated by the East Asian summer monsoon, the moisture of which is derived from tropical and equatorial Pacific Ocean. On the glacial-interglacial time scales, the combined impact of ice volume and temperature is around 0.5‰ for the oxygen isotope compositions of ocean water (Dayem et al., 2010). Obviously, the change of isotopic composition in moisture source regions is too small to explain the large amplitude shift of our $\delta^{18}O_{\text{carb}}$ record. Modern meteorological observations show that Lake Xiarinur is located in the areas where the $\delta^{18}O_{\text{carb}}$ of precipitations is positively related to the temperature (Dayem et al., 2010; Johnson & Ingram, 2004; Liu et al., 2008). Based on this positive relationship, our long-term record would suggest a warmer climate in the late rather than the early Holocene, which is contrary to both regional and global paleotemperature reconstructions (Marcott et al., 2013), and also contradicts the Holocene temperature changes inferred by the insolation changes (Berger & Loutre, 1991). Therefore, the oxygen isotope composition of the water in Lake Xiarinur, similar to most of the lakes in semiarid and arid areas across the globe (Horton et al., 2016; Leng et al., 2006; Li & Ku, 1997), is dominated by the precipitation/evaporation ratio. In addition, the salt content reflects the salinity of the lake water, which in closed lakes is directly related to the precipitation-evaporation ratio.

3.4. How Well Do Our Proxies Represent the Asian Dust Source Regions?

3.4.1. Regional Comparison of the Holocene Climate Changes

The records of $\delta^{18}O_{\text{carb}}$ and soluble salt from Lake Xiarinur indicate that climate was humid in the early Holocene and became significantly drier in the late Holocene (Figure 2). This temporal pattern has been demonstrated by various evidences across the Asian dust source regions. First, our record is comparable with the high-resolution stalagmite $\delta^{18}O$ records from Dongge and Sanbao caves (Wang et al., 2005; Wang et al., 2008) on both the millennial and the centennial time scales (Figure 2), which is often used as the benchmark of the East Asian summer monsoon (EASM). Both records indicate that the EASM was high in the early Holocene (11.5 to 7.0 ka B.P.), and the Holocene Thermal Maximum (HTM) occurred during the interval from 9.0 to 7.0 ka B.P. Second, the lake $\delta^{18}O$ records, which are mainly related to the Asian summer monsoon precipitations, indicate a consistent pattern across China characterized by low $\delta^{18}O$ values before 7.0 ka B.P. and an increasing trend in the late Holocene (Zhang, Chen, et al., 2011), supporting a humid in the early but a dry climate in the late Holocene (Figure S3). Third, the pollen records from Lake Dalí, Dalhai, and Bayanchagan (Jiang et al., 2006; Peng et al., 2005; Wen et al., 2010; Xiao et al., 2006), which are located in same region with Lake Xiarinur, also indicate that the climate was warm and humid in the early and then became drier in the late Holocene. Similarly, the synthesis of pollen records from 20 sites in the EASM margin across north China (Zhao & Yu, 2012) shows that the climate was moist and dry in the early and late Holocene, respectively (Figure S4). Finally, a similar trend was also indicated by various records from central and
north Asia (Figure S4). These records include (1) the pollen-derived indexes of aridity and precipitations from Lake Son Kul in Kyrgyzstan and the $\delta^{18}O$ record of ostracode shells from Hoton Nur in southwest Mongolia (Huang et al., 2014; Mathis et al., 2014; Ricketts et al., 2001; Rudaya et al., 2009), (2) the index of humidity from both Lake Hovsgol from Mongolia and Lake Baikal from Russia (Prokopenko et al., 2007), and (3) multiproxy records of Lake Ulaan from south Mongolia (Lee et al., 2013). All these evidences consistently support that the climate was humid in the early Holocene and became drier later across the Asian dust source.

In the late Holocene there were two drought events around 2.5 and 0.5 ka B.P. indicated by the two sand horizons, corresponding to the relative high values of stalagmite $\delta^{18}O$ (Figure 2). However, the $\delta^{18}O_{carb}$ of Lake Xiarinur did not show significant shifts during these intervals. The sediments are composed of desert sands, indicating presence of mobile sand dunes in Qtindag region and desiccation of Lake Xiarinur. Therefore, the isotope compositions of sediments mainly reflect the isotopic signatures of local clastic materials and eolian dusts rather than authigenic carbonates. This assumption is consistent with the climate records around this region. Similar to Lake Xiarinur, Lake Huangqihai became a playa around 2.2 ka (Zhang, Jia, et al., 2011). The climate records from Lakes Daihai (Peng et al., 2005; Xiao et al., 2006), Hulun (Xiao et al., 2009), Dali (Xiao et al., 2008), Juyanzhe (Hartmann & Wünnemann, 2009), and Anguli-nuur (Wang et al., 2010) consistently indicate that the climate was dry and lake level is low around these two periods. The record from Daihai indicates that the precipitation reached the minimum value of the Holocene, and climate was extremely dry around 2.5 ka and 0.5 ka B.P. (Peng et al., 2005; Xiao et al., 2006). In addition, the average rate of palynological change from fossil pollen records ($n = 26$) in eastern monsoonal China also reached the lowest value of the Holocene around 2.5 ka and was low around 0.5 ka B.P. (Zhao et al., 2009).

3.4.2. Regional Comparison of the Eolian Activity Records

We compared our record with various eolian records from central to eastern Asia, including (i) the lake records from central and eastern Asia, (ii) Holocene changes of mobile sand dunes in dust source regions, and (iii) the corresponding records in the near downwind deposition region (Chinese Loess Plateau). All these records are consistent with our record, suggesting a higher eolian activity in the late rather than the early Holocene.

The lake records from central Asian dust source regions (Lake Tuolekule and Genggaihai) and downwind areas suggest a similar pattern of eolian activity changes during the Holocene (Figure S5). The eolian record from Lake Tuolekule shows that the eolian activity was relatively low in the early Holocene, reached the lowest during HTM, and then increased after 6.0 ka B.P. (An et al., 2011). The high fraction of eolian sand in the sediments of Lake Genggaihai also supports the strengthening eolian activity in the late Holocene (Qiang et al., 2014). In addition, the record from Qinghai Lake basin also shows a higher eolian activity in the late rather than the early Holocene (Lu et al., 2015). In the downwind eastern Asia, the records from Marr lake of Sihailongwan in the northeast China and Cheju island in South Korea (Lim et al., 2005; Zhu et al., 2013) clearly show that the eolian activity was low in the early Holocene and reached the lowest in the middle Holocene and increased in the late Holocene (Figure S5). Furthermore, the high-resolution record of Lake Sihailongwan is similar in patterns with the record of Lake Xiarinur on both the centurial and the millennial time scales (Figure S5).

Eolian sand represents dune-field expansion and/or dune buildup with increased eolian activity, whereas paleosols indicate stabilization of dunes resulting from low eolian activities and ameliorated vegetation cover. We compiled the available chronological data of the sand horizons and the paleosols in the
sandy fields and deserts (710 chronological data from more than 150 profiles), which cover the entire Asian dust source regions (Figure 1), including the deserts and sandy fields of Mu Us (He et al., 2010; Liu & Lai, 2012; Lu et al., 2013; Sun et al., 2006), Qinqia (Li et al., 2002; Lu et al., 2013), Horqin (Lu et al., 2013; Sun et al., 2006; Yang et al., 2012; Zhao et al., 2007), and Hulun Buir (Li et al., 2002; Li & Sun, 2006; Sun et al., 2006) in northern and northeastern China, the sand fields of Gobi in the north- eastern Qinghai-Tibetan Plateau (Lu et al., 2013), the deserts of Badain Jaran, Tengger in central northern China (Lu et al., 2013), and the deserts of Taklimakan (Lu et al., 2013), Gurbantünggüt and Kumtag (Li & Fan, 2011; Lu et al., 2013), Tengger and Qaidam Basin (Qiang et al., 2010; Yu & Lai, 2012, 2014) in northwestern China, and Gobi in southern Mongolia (Hülle et al., 2010). These chronological data consistently indicate that the paleosols were mainly developed in the early Holocene with sporadic sand dune sand depositions, while the sand dune sand depositions were widespread in the late Holocene (Figure S6), supporting stronger eolian activity and a larger dust source area in the late rather than the early Holocene. This pattern is consistent with the changes of the boundaries of all sandy fields and deserts, which shifted toward the south from the early to the late Holocene (Lu et al., 2013). The biome simulations also show smaller areas of Asian dust source regions in the early rather than late Holocene (Dallmeyer et al., 2013; Sun et al., 2006; Yang et al., 2012; Zhao et al., 2007), and Hulun Buir (Li et al., 2002; Li & Sun, 2006; Sun et al., 2006) in northern and northeastern China, the sand fields of Gobi in the north-easterly Qinghai-Tibetan Plateau (Lu et al., 2013), the deserts of Badain Jaran, Tengger in central northern China (Lu et al., 2013), and the deserts of Taklimakan (Lu et al., 2013), Gurbantünggüt and Kumtag (Li & Fan, 2011; Lu et al., 2013), Tengger and Qaidam Basin (Qiang et al., 2010; Yu & Lai, 2012, 2014) in northwestern China, and Gobi in southern Mongolia (Hülle et al., 2010). These chronological data consistently indicate that the paleosols were mainly developed in the early Holocene with sporadic sand dune sand depositions, while the sand dune sand depositions were widespread in the late Holocene (Figure S6), supporting stronger eolian activity and a larger dust source area in the late rather than the early Holocene.

4. Discussion

The eolian activity displays a close association with climatic drying (Figure 2). The low eolian activity coincides to the humid climate in the early Holocene. The lowest value of the eolian activity occurred during the interval from 8.5 to 6.5 ka B.P., coinciding to the low δ18Ocarb. However, the eolian activity was still relative low from 7.2 to 5.5 ka B.P., though the climate began to dry indicated by the increased δ18Ocarb. We suggest the climate was still humid enough to sustain vegetation coverage, and the delayed response of vegetation coverage to climate changes was the main cause of the low eolian activity during this interval. This conclusion is consistent with the pollen records from Lake Bayanchagan (Jiang et al., 2006), which is located in same region of Lake Xiarinur. The pollen record indicates that the coverage of tree and shrub reached the highest proportion during this interval and then decreased abruptly, while the wettest climate occurred between 10.5 and 6.5 ka B.P. (Jiang et al., 2006). What is more, the eolian activity also displayed an increasing trend with the δ18Ocarb increase during this interval (Figure 2), consistent with the close association between eolian activity and climate drying. After 5.0 ka B.P., the eolian activity increased significantly with climate drying indicated by substantial increase of δ18Ocarb and salt contents, and the two sand horizons correspond to two drought events (Figure 2).

In order to explore the relationship between eolian activity and the long-distant Asian dust export, we compared our record with the nonsea salt Ca2+ concentrations (nssCa2+) of the GISP2 Greenland ice core (Fischer et al., 2007; Mayewski et al., 1997), the dusts of which primarily, if not exclusively, originated from Asia during the interval from the Last Glacial Maximum to the present (Biscaye et al., 1997; Bory et al., 2002; Uno et al., 2009). The most obvious feature is the decoupling of the intensity of eolian activity and climate drying in northern China from dust accumulations in the long-distant downwind areas (Figure 4). The value of nssCa2+ decreased by more than 20% from the early (8.4 ± 1.4 ppb) to late (6.6 ± 0.8 ppb) Holocene, while the record from Lake Xiarinur shows that the average flux of the eolian fraction increased from 5.7 to 31.0 (10−2 g/yr/cm²). The high dust accumulation in GISP2 (Mayewski et al., 1997) corresponded to the humid climate and low eolian activity of the dust source region in the early Holocene, and the low dust accumulation coincided to the climatic drying and high eolian activity in the late Holocene (Figure 4). In addition, the records from North Pacific (eight sites) and Subarctic North Pacific (Rea & Leinen, 1988; Serno et al., 2015; Wang et al., 1998), which form a broad longitudinal transect from subtropical to Subarctic North Pacific, and across the prevailing westerlies in North Pacific, also indicate that the flux of Asian dust depositions was higher in the early rather than the late Holocene (Figure S7), though the temporal resolution of these records is low. All these evidences clearly indicate a limited role of climate drying in long-distant Asian dust export during the Holocene, challenging the concept of aridity as the major forcing of Asian dust export across the globe.
Dust export requires both sufficient source strength to generate dust particles and atmospheric circulation sufficiently energetic to transport dust to the point of deposition (Guo et al., 2002; Seki et al., 2015). The source strength includes the changes of dust source areas, soil moisture, and vegetation coverage. As discussed above, the environment of Asian dust source regions was relatively humid, and source areas were smaller with greater vegetation in the early Holocene (Dallmeyer et al., 2015; Lu et al., 2013), indicating a low source strength.

Changes of dust source and dust deposition efficiency (wet to dry deposition) have a potential influence on long-distant dust export and thus the dust accumulations in the Northwest Pacific and Greenland. However, the evidence available currently indicates a little role of these factors. Radiogenic isotope and mineralogical compositions of modern dust and the dust in Greenland ice cores indicate a dominant role of the eastern Asia from the Last Glacial Maximum to present, and no apparent contribution from other sources including African and North American deserts (Biscaye et al., 1997; Bory et al., 2002; Bory et al., 2003; Svensson et al., 2000; Uno et al., 2009). The isotopic, mineral, and chemical compositions of deep-sea sediments in North Pacific Ocean also suggest a dominant component of Asian dust (Nakai et al., 1993; Olivarez et al., 1991).

For the efficiency of dust deposition, previous studies showed that atmospheric dust concentrations over Greenland changed by similar factors and at similar speeds to concentrations seen in ice (Fuhrer et al., 1999), because the dust deposition in Greenland is dominated by wet deposition during both the Last Glacial Maximum and Holocene (Alley et al., 1995; Fuhrer et al., 1999). The dust fluxes and concentration show a consistent variation over last 50,000 years, which cannot be explained by either changes in the deposition efficiency or local deposition changes (Fischer et al., 2007). Most importantly, the aerosol records with decadal or even centennial resolution integrate over many precipitation events and efficiently average this variability out (Fuhrer et al., 1999). Accordingly, the aerosol data with decadal or centennial resolution record change in the average transport of aerosol to the deposition sites (Fischer et al., 2007). This conclusion is supported by the consistence of dust accumulations in the downwind sites in Greenland and North Pacific during the Holocene, which cross a large region of Asian dust transport, indicating that the changes in deposition en route should not play a substantial role.

Therefore, the long-distance Asian dust export in the Holocene should be dominated by the of atmospheric circulation perturbations. This conclusion is consistent with the results of model studies, which indicate a dominant role of surface wind strength rather than climate-drying forced desertification and land cover in Asian dust emissions (Chow et al., 2015; Zhang et al., 2003).

The westerlies and Siberian High (SH) are the major circulation systems dominating Asian dust emission and transport. To identify the extent to which circulations link to the long-distance Asian dust transport, we compare the dust accumulations in GISP2 (Mayewski et al., 1997) with the available best reconstruction of the westerlies (Zhu et al., 2015) and the index of the SH-forced East Asian winter monsoon (EAWM) (Wang et al., 2012). The result shows that the high and low dust accumulations in Greenland and the Northwest Pacific Ocean corresponded to the strong and weak EAWM in the early and late Holocene, respectively (Figure 4). The warming winter in the Siberian Arctic from the middle to the late Holocene also suggests a weak Siberian High and thus a weak EAWM in the late Holocene by decreasing the pressure difference between the Siberian High and tropical oceans (Meyer et al., 2015). In contrast, the westerlies were weaker in the early rather than the late Holocene indicated by both the pollen discrimination index from windward western China (Zhu et al., 2015) and the ESR of silt-sized quartz particles from downwind Japan Sea (Nagashima et al., 2013) (Figure 4). The pollen-discriminated index of the westerlies from the windward Tibet Plateau indicates that the proportion of the pollen transported by the westerlies displayed an increasing trend from early to late Holocene (Zhu et al., 2015). Similarly, in the downwind areas, the proportion of the dust from Taklimakan, which is transported mainly by the westerlies into Japan Sea, was lower in the early rather than the late Holocene (Figure 4).

The shifts in longitudinal position and wind speed of the westerlies might influence Asian dust transportations and thus the dust depositions in the downwind areas during the Holocene. However, the following evidences suggest little impact of these two factors: (1) the records from deep-sea sediments of North Pacific, which form a longitudinal transect across the prevailing westerlies near 160°E, indicate that the location of the greatest eolian dust flux changes little and has always been in the vicinity of 38–40°N during past 30,000 years (Rea & Leinen, 1988); (2) the indexes of the westerlies used in this study consistently show
weak westerlies in the early Holocene (Figure 4), though they are located in different latitudinal locations; and (3) the record from central Asia, which is dominated by the westerlies, also indicates weaker westerlies in the early Holocene (Jiang et al., 2013).

Most importantly, under strong EAWM condition in the early Holocene, the intensity of eolian activity in north China was correlated with dust accumulation in the GISP2 ice core on both millennial and centurial time scales ($R = 0.57$ with $p < 0.01$) within the dating uncertainty (Figure S8). In addition, the high component of Fraction 1 (2-μm) in Lake Xiarinur during the early Holocene, the main component of dust with a potential of long-distant transport that reflects the background of atmospheric loading (Prins & Vriend, 2007; Vriend & Prins, 2005), also supports a high dust loading in the atmosphere under strong EAWM conditions in the early Holocene (Figure 4). These evidences consistently suggest a critical role of the Siberian High in long-distant Asian dust export, contradicting with the previous assumption for the overwhelming role of the westerlies (Pye & Zhou, 1989; Sun et al., 2001; Zhang et al., 2014). This assumption is supported by the positive correlation between the dust deposition in a Greenland ice core (GISP2) and spring SLP over Siberia observed in instrumental data of the past century (Meeker & Mayewski, 2002).

The central role of the Siberian High in long-distant Asian dust export is also consistent with the most recent modern observations and the results of model studies, particularly those based on Cloud-Aerosol Lidar and Infrared Pathfinder Satellite observations (Huang et al., 2008; Liang et al., 2004; Wuebbles et al., 2007; Zhao et al., 2006). These observations provide a continuous global measurement of aerosol and cloud vertical distributions with high spatial resolution and show that the dust originated from eastern Asia where the surface wind is mainly forced by the Siberian High could be transported to long-distant downwind regions in both the lower and upper troposphere, contradicting with the previous assumption that the EAWM-forced dust from eastern Asia is mainly transported near the surface and short distances (Pye & Zhou, 1989; Sun et al., 2001; Zhang et al., 2014). A strong Siberian High would enhance the flux of dust transported to high levels of the atmosphere by increasing the frequency of midlatitude cyclone activities (Wuebbles et al., 2007) and/or transport more dust to the deep convection regions where it is lift to the upper stratosphere (Cooper et al., 2004; Wuebbles et al., 2007; Zhao et al., 2006), and then transported long distances by the westerlies. In addition, the teleconnection of the Siberian High with other atmospheric circulation patterns such as the strengthening of the Azores High, the weakening of the Arctic High, and the deepening of the Icelandic and Aleutian Low could enhance Asian dust transport to Greenland via both meridional and zonal routes (Kang et al., 2003; Ruth et al., 2007).

The characteristics of Asian dust export during the Holocene also have some implications for the climatic and environmental effects of anthropogenic activity, although our data do not allow us to distinguish anthropogenic dust from natural background. The statistical analysis on prehistoric culture sites in north China indicates that the human activity was weak before 8.0 ka B.P., and then increased from 8.0 to 4.0 ka B.P. before it fell during the interval from 4.0 to 3.0 ka, and increased significantly after 2.0 ka (Zhuo et al., 2013). The coincidence of high eolian activity with anthropogenic activity during the intervals of 6.0–4.0 ka B.P., and particularly after 2.0 ka B.P., points to a likelihood of the significant relationship between them. However, the dust accumulations in the GISP2 did not show significant changes during these intervals (Figure 4), suggesting the limited impact of anthropogenic activity on atmospheric aerosol loadings on a global scale. This characteristic is consistent with the central role of atmospheric circulation in Asian dust export. Therefore, under conditions of a low Siberian High, the anthropogenic aerosols from China, including sulfates from pollution and carbonaceous aerosols from biomass burning, are expected to be relatively poorly dispersed from the source regions; otherwise, they would have a global impact.

5. Conclusion

This study presents a multiproxy lake sediment record of Holocene eolian activity and climatic changes from a major Asian dust source region. Our data, together with the Holocene changes of both climate and eolian activities in Asian dust source regions, records of dust accumulation in the Northwest Pacific Ocean and Greenland, provide new insights into the dynamics of Asian dust export. Our results indicate that the eolian activities in Asian dust source region were closely correlated with aridity in the Holocene. The intensity of eolian activity increased with climate drying from middle to late Holocene, and the low eolian activity coincided with the humid climate of the early Holocene. However, the long-distance export of Asian dust...
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decoupled from the climate drying in the Holocene. The climate was humid, and the eolian activity was weak in the source region during the early Holocene, but the dust accumulation in the Northwest Pacific Ocean and Greenland was relatively high. Similarly, the climatic drying and the strong eolian activity in north China corresponded to a low dust accumulation in the downwind areas during the late Holocene. The high flux of long-distance export of Asian dust displays a close association with the Siberian High, not the westerlies, as widely believed before, indicating a dominant role of the Siberian High on the magnitude of Asian dust export across the globe. However, the underlying mechanism of long-distance Asian export, particularly the specific role played by the Siberian High and the westerlies, needs further studies.

References


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