Hf-Nd isotopic variability in mineral dust from Chinese and Mongolian deserts: implications for sources and dispersal

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Mineral dust provenances are closely related to the orogenic processes which may have distinct Hf-Nd isotopic signatures. Here we report the clay-sized (<2 μm) Hf-Nd isotope data from Asian dust sources to better constrain the source and transport dynamics of dust deposition in the North Pacific. Our results show that there is a more positive radiogenic Hf isotopic composition with clay-sized fractions than the corresponding bulk sample and a decoupling of the Hf-Nd couplets in the clay formation during the weathering process. The clay-sized Hf-Nd isotopic compositions of the desert samples from the Sino-Korean-Tarim Craton (SKTC) are different from those of the Gobi and deserts from the Central Asian Orogeny Belt (CAOB) due to varying tectonic and weathering controls. The Hf-Nd isotopic compositions of dust in the North Pacific central province (NPC) match closely with those from the Taklimakan, Badain Jaran and adjacent Tengger deserts, implying that the NPC dust was mainly transported from these potential sources by the westerly jet. Our study indicates that dusts from the CAOB Gobi deserts either didn’t arrive in NPC or were quantitatively insignificant, but they were likely transported to the North Pacific margin province (NPM) by East Asian winter monsoon.

Mineral dust accounts for more than 50% of the atmospheric dust loading, with the clay fraction (<2 μm) comprising about half of mineral dust1. It plays an important role in the marine and terrestrial geochemical cycles and impacts global climate by scattering and absorbing solar radiation, changing cloud properties, affecting bio-geochemical cycles and providing important surfaces for atmospheric reactions in the earth–atmosphere–ocean system2–6. Fine Asian dust (<2.5 μm) is a consistent component of the troposphere over the eastern Pacific and western North America7. Asian dust is the second largest source of dust on Earth and has been studied intensively over the past decade, especially with 87Sr/86Sr and 143Nd/144Nd ratios of <75 μm silicate particles which are seen as a powerful tool to identify source areas8–12. However, the isotopic geochemistry of the clay-sized fraction of Asian dust has not been studied; especially the clay-sized Hf isotopic fingerprints of its provenance have not been reported.

Hf-Nd isotopes have been widely used for provenance research in the field of global geochemical cycles13–16. Hf and Nd isotopes plot with a single “mantle-crust” or terrestrial array in igneous and clastic rocks, indicating that Hf and Nd isotopes coupled during processes that operate the Earth’s crust. Asian eolian dust in the Pacific Ocean is offset towards more radiogenic Hf from the global silicate earth array17. The recent conclusion that the oceanic ferromanganese crusts and terrigenous clays, deviate significantly from the Terrestrial Array towards higher εHf values relative to their εNd values, mainly result from the incongruent behavior of Hf during continental weathering18–23. This Hf-Nd decouple is still under debate18,21,24–26. Furthermore, eolian dust input with high εHf was only discovered in recent years17,27–30. Biscaye et al.17 suggested that the probable source area of the GISP2 dusts was in East Asia by comparing the Nd isotopes of fine fraction (<5 μm) dust particles extracted from Asian Gobi/sandy deserts and Greenland ice cores. Previous studies have confirmed that coarse silt grains are only transported short distances (<3 km) through saltation and short-term suspension due to their greater density. The smaller the dust particles are, the longer they stay in suspension in atmosphere and the further they will be transported. However, clay-sized particles might be lifted to the upper troposphere (>8 km) and transported over a long distance by the westerly jet32–34 and clay
minerals were relatively enhanced in samples in remote locations. In particular, compared to the non-clay fraction (>2 μm), the clay-sized fraction (<2 μm) has unique minerals phases (dominant by clay minerals like illite, kaolinite, chlorite and smectite) and is removed from the atmosphere by wet deposition (precipitation scavenging). Thus, the clay-sized isotopic fingerprints from Asian deserts may be ideal targets not only for provenance tracing of long-distance transported mineral dust, but also provide an unparalleled window for understanding the global dust cycle, especially, eolian dust preserved in deep-sea sediments.

To better understand how the Asian dust cycle influences marine sediments and sea water in North Pacific Ocean, we conducted a detailed investigation on the clay-sized Hf-Nd isotopic compositions from the Gobi/sandy deserts in North China and neighboring Mongolia. Our objectives are to address the following questions. 1) What controls the Hf-Nd isotopic composition of clay-sized fractions within desert sands? 2) What are the general characteristics of Asian dust and how do those characteristics differ from other dust sources? 3) What are implications for the source and transport pathway of eolian sands in the North Pacific Ocean?

Results

Sampling sites of desert sands are situated on the Sino-Korean-Tarim Craton (SKTC, including North China Craton and the Central Asian Orogeny Belt (CAOB)) and the Central Asian Orogeny Belt (CAOB). The study areas include the Chinese deserts, Mongolian Gobi and northwest Pacific Ocean, as shown in Figure 1 and Figure 3. The Hf and Nd isotopic data of the clay-sized fractions of the Chinese deserts and the Mongolian Gobi are presented in Figure 2A (see Table S1 in Supplementary information). The Chinese deserts have εNd values ranging from -17.30 to 0.98 (mean = -8.40) and εHf from -5.94 to 4.63 (mean = -0.97). The Mongolian Gobi is more radiogenic in Nd and Hf isotope compositions which ranges from -5.99 to -2.67 (mean = -4.43; n = 9) and from -2.56 to 3.68 (mean = 0.81; n = 9), respectively. It is clear that the clay-sized fractions have higher radiogenic Hf isotopic composition than silt-to-sand silicate fractions (>2 μm) (see Table S1 and Table S2 in Supplementary information). The εNd (εNd = -5.4) of <2 μm fraction in sample BT-46 are similar to those of <75 μm fractions (εNd = -5.6), while the εHf of >2 μm fractions of BT-46 have εHf values ranging from -21.01 to -6.72 (see Figure S2 and Table S3 in Supplementary information). The Sm/Nd values both >2 μm and <2 μm fraction are in good agreement with nearly constant Sm/Nd ratio (Sm/Nd = 0.18) (see Table S3 in Supplementary information). However, Lu/Hf (Lu/Hf of <2 μm fraction = 0.1) of the <2 μm fraction is much higher than that (Lu/Hf of >2 μm fraction = 0.05) of the >2 μm fraction (Supplementary information).

Discussion

Tectonic controls on the clay-sized Hf-Nd isotopes. The Hf-Nd isotopic signatures of the Chinese and Mongolia Gobi Deserts are consistent with previous Sr-Nd isotopic observations, suggesting that the isotopic composition of deserts is closely related to the tectonic setting of the surrounding mountains. Hf isotopic systematics can distinguish between orogenic processes dominated by the generation and reworking of continental crust and those dominated by additions of juvenile crust. The geological setting of Paleozoic exposures clearly shows that there are two first-order geological and tectonic units in the research area (i.e., the CAOB and the SKTC) (Fig. 2A). The Gobi Desert, Gubanunggut Desert and Hulun Buir Sandy land are on the CAOB, whereas the Taklimakan, Qaidam, Badaim Jaran, Tengger and Mu Us Deserts are on the SKTC. The Tarim, Qaidam and Alxa blocks belonged to the Sino-Korean tectonic domain during the Archean-Mesoproterozoic. The relatively high clay-sized εHf - εNd value within CAOB indicates that the clay-sized fraction is generated from CAOB juvenile continental crust, which was formed by the collision between the Siberian Plate and the southern blocks during the early stages of the orogeny about 1.0 Ga and continued to about 250 Ma. The clay-sized εHf - εNd values controlled by CAOB are higher than SKTC terrane, whereas...
the variability of both $\varepsilon_{\text{Hf}}$ and $\varepsilon_{\text{Nd}}$ in the CAOB are smaller than the corresponding SKTC $\varepsilon_{\text{Hf}}$ and $\varepsilon_{\text{Nd}}$. There are two obvious end-members easily discerned from present clay-sized $\varepsilon_{\text{Hf}} - \varepsilon_{\text{Nd}}$ compositions shown in Figure 2A. The clay-sized fractions derived from the old continental shield produce the lowest $\varepsilon_{\text{Hf}}$ and $\varepsilon_{\text{Nd}}$ values, especially the samples from the Mu Us and Hobq Deserts. The isotopic regions are consistent relative to the clay-sized $\varepsilon_{\text{Hf}} - \varepsilon_{\text{Nd}}$ values from deserts of the same geologic setting, suggesting that not all the isotopic differences are caused entirely by the heterogeneity of material at their source. Geologically, blocks and/or cratons formed the Chinese continent through multiple collisions and aggregation41. The clay-sized Hf-Nd isotopes of the Qaidam Desert were similar to the SKTC terrane, we thus conclude that the Qaidam basin was attributed to SKTC, even if the Qaidam basin was influenced by the proximity to Altunshan Fault, Tarim craton and Central China Orogen42,45.

It is noteworthy that samples D17 and Nmy-8 are SKTC end members whereas the other Horqin samples belong to CAOB end members. These are the few exceptions to the geographic distribution of the Hf-Nd isotopic composition, although both D17 and Nmy-8 are from the southern-most edge of Horqin sandy land which is located in the SKTC (Figure 2A). One possible reason for these exceptions is that the boundary between different tectonic domains (Figure 1, Block Suture) may run through the southern part of Horqin sandy land from west to east41, and in term of source materials, both D17 and Nmy-8 may actually belong to SKTC. Geographically, both D17 and Nmy-8 belong to CAOB, but their source may be the mountainous area of North China Craton as assessed by comparing clay-sized $\varepsilon_{\text{Hf}} - \varepsilon_{\text{Nd}}$ values. In fact, the sand sediments in the southern Horqin sandy land appears to be transported directly from the northern mountainous margin of North China Craton by rivers46. However, the other two exception samples (T46 and Surfer25), which are located on the northeast of Taklimakan Desert and the northern edge of Hubq Desert between CAOB and SKTC tectonic domain, respectively, do not fall within the SKTC end-member but close to CAOB end member edge, implying that they were controlled by CAOB and SKTC tectonic domains. Instead they reflect the influence of surface transport causing the isotopes to be skewed toward the CAOB by near-surface northwesterly wind47.

Clay array and continental weathering. Regression of all the clay-sized data yields a clay array:

$$\varepsilon_{\text{Hf}} = (0.45 \pm 0.04) \times \varepsilon_{\text{Nd}} + (2.81 \pm 0.35) \quad (R = 0.80).$$

The new Terrestrial Array ($\varepsilon_{\text{Hf}} = 1.55 \varepsilon_{\text{Nd}} + 1.21$), Zircon-free Array ($\varepsilon_{\text{Hf}} = 0.91 \varepsilon_{\text{Nd}} + 3.1$) and Seawater Array ($\varepsilon_{\text{Hf}} = 0.62 \varepsilon_{\text{Nd}} + 5.27$) are from Vervoort (2011)21, Bayon(2009)23 and David (2001)48, respectively. The $\varepsilon_{\text{Hf}}$-$\varepsilon_{\text{Nd}}$ diagram compares the desert clays with the Asian eolian dust extracted from the North Pacific Ocean17.
composition from the terrestrial array toward the seawater array can be generated by incongruent weathering of continental rocks, which is known as “zircon effect”\textsuperscript{24,25,49}. The zircons, with low $e_{\text{Hf}}$ have relatively high Hf concentrations and indistinguishability, and contain large amounts of unradiogenic Hf, causing relatively radiogenic Hf to enter weathering products and/or fine-grained sediments. Thus, clay minerals, the weathering products of continental rocks, are expected to be more radiogenic that primary rocks or bulk sediments. One would explain the elevated radiogenic Hf composition of the clay fraction by the zircon-free effect (mineralogical sorting or grain size effect), because the clay fractions are too fine ($<2 \mu m$) to contain any zircon. However, the clay array is above and underscored by the zircon-free sediment array\textsuperscript{25}, suggesting that the zircon-free effect alone is insufficient to generate the clay $e_{\text{Hf}} - e_{\text{Nd}}$ relationships because the clay-sized fractions contain relatively more radiogenic Hf than fine-grained sediments (zircon-free sediment). During the weathering process, clay minerals incorporate and/or absorb the incongruent released radiogenic Hf to form the decoupling of the clay-sized Hf and Nd isotopic compositions, which are determined by both the weathering regime and source provenance.

This Hf-Nd isotopic decoupling is attributed to the different Goldschmidt behavior of Hf and Nd during weathering. Hf is both near the Stokes’ Law and then were recovered by centrifuging\textsuperscript{56}. The samples were subsequently using excess 1M acetic acid for 10 hours in order to eliminate the influence of secondary carbonate on Hf isotopic composition. The samples were subsequently rinsed at least three times with MilliQ water to completely remove major ions and soluble salts. Different fractions were extracted by sieving the ultrasonically dispersed samples in mesh with MilliQ water, and the $<2 \mu m$ particles were separated based on the Stokes’ Law and then were recovered by centrifuging\textsuperscript{44}. The samples were subsequently rinsed at least three times with MilliQ water to completely remove major ions and soluble salts.

The Hf-Nd isotopic ratios of the extracted clay-sized fractions were measured with a Thermo Fisher Scientific Neptune MC-ICP-MS at the State Key Laboratory for

The majority of Hf-Nd isotopic data\textsuperscript{17} plot within the SKTM and CAOB areas in the Hf-Nd isotope space, while a few $>25$ Ma samples from downcore LL44-GPC3, A-2H-5 (2.4 Ma) and A-3H-1 (3.2 Ma) samples from 885/886 plot above the seawater array (Figure 2B). The Hf-Nd isotopic correlation line for modern dust ($e_{\text{Hf}} = 0.78e_{\text{Nd}} + 5.66$) is very close to our clay-sized array (Figure 2B). This consistency may indicate that the clay-sized dust deposited in the North Pacific Ocean was predominantly derived from the Northwest China and Mongolia deserts. This could explain the flat Hf-Nd isotopic correlation and the variable and radiogenic $e_{\text{Hf}}$ values of the North Pacific modern dusts\textsuperscript{7} that is characteristic of the clay-sized fractions of the Asian deserts.

The Hf-Nd isotopic values of the Asian dust end-member were reported\textsuperscript{24} as $-9.0 > e_{\text{Nd}} > -10.8$ and 2.5 $> e_{\text{Hf}} > -4$. These isotopic values are from the SKTC Hf-Nd isotope space (Figure 2B) and match closely with Hf-Nd isotope data from the Taklimakan, Badain Jaran and adjacent Tengger deserts. Because this value is determined by dusts chemically isolated from the North Pacific central province (NPC) sediments, this suggests that modern dusts deposited in the NPC were mainly from these deserts and that dusts from the Mongolian deserts were volumetrically inconsequential. Satellite observations of certain dust storm trajectories might support the above scenarios. For example, dust originating from the Taklimakan desert was observed lofted to the upper troposphere, around 8–10 km, and is deposited largely over the North Pacific\textsuperscript{2}. In contrast, remotely sensed dust observations suggest that dust from the Mongolian Gobi desert was carried in a northeastward trajectory as it leaves the Asian continent, then travels eastward and is deflected to the south near the Australians before it enters the western American coast\textsuperscript{44}. These different dust transport pathways may indicate that the clay-sized Hf-Nd isotopic signal entrained by different prevailing winds, such as winter monsoon and westerly.

The Hf-Nd isotopic data of eolian dusts isolated from pelagic sediments in NPM plot in or near the CAOB area in the Hf-Nd isotopic correlation diagram (Figure 2B). This may imply that modern eolian dusts deposited in the NPM may have a dominant CAOB origin besides the commonly accepted origin of the binary mixture of dust contributed from island arc volcanic material and Asian dust with an SKTC origin as discussed above. Based on the comparison with the $e_{\text{Hf}} - e_{\text{Nd}}$ from the time series of Ocean Drilling Program (DSDP) 885/886 and LL44-GPC3, the source of Neogene dust in NPC may come from SKTC. Our clay-sized Hf-Nd isotopic signals from major Asian Gobi/sandy deserts indicate that sources and dispersal patterns of dust deposits in the NPC and NPM are spatially different (Figure 3).

**Methods**

Samples of surface sand were collected from all the potential sources of Asian dust by first removing the top 5 cm and then sampling to a depth of 10–20 cm. The sampled sands and sandy lands include the Hulun Buir and Horqin sandy lands in northeastern China, Gurbantunggut and Taklimakan Deserts in northwestern China, the Qaidam Basin in the northern Tibetan Plateau, the Badain Jaran and Tengger Desert on the Alxa Plateau, the Hobq Desert and Mu Us Desert on the Ordos Plateau, and the Gobi Desert on the Mongolian plateau. The exception to our sampling routine was the Mongolian Gobi desert sample which were collected by scratching off 1–2 cm thick clay mud crust\textsuperscript{44}. According to the geological setting of Paleozoic exposures, the Chinese and Mongolian deserts are on the Sino-Korean-Tarim Craton\textsuperscript{39} (SKTC, including North China Craton\textsuperscript{69} and the Central Asian Orogyn Belt (CAOB))\textsuperscript{40}. In order to isolate just the clay-sized silicate mineral fraction for Hf analysis, and organic matter and carbonate were removed: organic matter was removed with excess hydrogen peroxide (30%) overnight and then a decarbonation step was carried out using excess 1M acetic acid for 10 hours in order to eliminate the influence of secondary carbonate on Hf isotopic composition. The samples were subsequently rinsed at least three times with MilliQ water to completely remove major ions and soluble salts. Different fractions were extracted by sieving the ultrasonically dispersed samples in mesh with MilliQ water, and the $<2 \mu m$ particles were separated based on the Stokes’ Law and then were recovered by centrifuging\textsuperscript{44}. The samples were subsequently rinsed at least three times with MilliQ water to completely remove major ions and soluble salts.

The Hf-Nd isotopic ratios of the extracted clay-sized fractions were measured with a Thermo Fisher Scientific Neptune MC-ICP-MS at the State Key Laboratory for
Mineral Deposits Research, Nanjing University. This samples were prepared as follows: First, sample digestion. 100 mg of the dry silicate residue was totally dissolved with HF–HClO4 mixture in a steel jacketed autoclave at 180 – 200°C for 72 hours16, while 100 mg of clay-sized fractions were digested with a mixture of HF–HClO4 at 110 – 140°C for 72 hours. Second, the purification for Hf and Nd with ion chromatography. The Hf analysis used a modified version of the method of Yang et al (2010)1 for purification and dissolving the samples in an Hf–HClO4 mixture and separating them by chromatographic extraction through an cation exchange resin (Bio-Rad 50 X 8 resin) Eichrom® Ln-Spec resin. Hafnium was separated from matrix by ion exchange procedures using Eichrom® Ln-Spec resin. These detailed analytical procedure for the Hf isotope measurement can be seen elsewhere15. Nd was then separated and purified by ion exchange procedures followed the detailed method from Pu et al17. All chemical digestion and purification were carried out in Class 100 ultra-clean laboratory. The total procedure blank for Lu, Hf, Sm and Nd were less than 10 pg, 50 pg, 50 pg and 60 pg, respectively, and thus negligible.

The mass spectrometric analyses were performed in Class 1000 clean laboratories. The 176Hf/177Hf standard9 (6.1629 ± 0.0005, 200 ppb) was analyzed to provide a calibration value for the standard: 176Hf/177Hf = 0.282161 ± 0.000004 (n = 20, 2σ). The JMC-475 and JMC-1 standard9 (6.1629 ± 0.0005, 200 ppb) was recalculated with the same value of 143Nd/144Nd = 0.512118 ± 0.000005 (20 ppb), which is similar to the reported referenced value of 143Nd/144Nd = 0.512115 ± 0.000007 (n = 15, 2σ). Instrumental mass bias was corrected for using 146Nd/144Nd ratio of 0.7219 and 143Nd/144Nd ratios was estimated from repeated measurements of JMC 475 and 176Hf/177Hf. Replicates for both 143Nd/144Nd and 176Hf/177Hf were processed and yielded an external reproducibility of better than ± 0.1 (2σ) for 143Nd/144Nd and ± 0.1 (2σ) for 176Hf/177Hf.

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**Author contributions**
J.J. designed the study. W.Z. performed Hf-Nd isotopic measurements. W.Z. and J.J. analyzed the data and wrote the manuscript preparation. W.B., Y.S., L.L., J.C. and H.L. polished the manuscript and contributed to the interpretation of the data.

**Additional information**
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