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Key Points:
- New clay mineral and Sr-Nd-Hf data for the last glacial Greenland dust sources
- Clay and Sr-Nd data suggest two equally plausible Greenland dust sources
- Hf isotopes of fine grain loess separates may be source diagnostic

Supporting Information:
- Supporting Information S1

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Two possible source regions for central Greenland last glacial dust

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Abstract Dust in Greenland ice cores is used to reconstruct the activity of dust-emitting regions and atmospheric circulation. However, the source of dust material to Greenland over the last glacial period is the subject of considerable uncertainty. Here we use new clay mineral and $<10\,\mu m$ Sr–Nd isotopic data from a range of Northern Hemisphere loess deposits in possible source regions alongside existing isotopic data to show that these methods cannot discriminate between two competing hypothetical origins for Greenland dust: an East Asian and/or central European source. In contrast, Hf isotopes ($<10\,\mu m$ fraction) of loess samples show considerable differences between the potential source regions. We attribute this to a first-order clay mineralogy dependence of Hf isotopic signatures in the finest silt/clay fractions, due to absence of zircons. As zircons would also be absent in Greenland dust, this provides a new way to discriminate between hypotheses for Greenland dust sources.

1. Introduction

Wind-borne mineral aerosol (here referred to as "dust") influences global climate directly and indirectly through diverse physical and biogeochemical processes and is therefore considered a major component of the climate system [*Harrison et al.*, 2001; *Tegen*, 2003]. Dust particles affect the radiation budget through scattering and absorption of incoming solar and outgoing infrared radiation [*Tegen and Lacis*, 1996; *Liao and Seinfeld*, 1998] and by altering cloud optical properties, amounts, and lifetimes [*Albrecht*, 1989; *Yin et al.*, 2002; *Andreae and Rosenfeld*, 2008]. Furthermore, airborne mineral dust provides micronutrients (e.g., iron and silica) to marine and terrestrial ecosystems [*Martin*, 1990; *Duce and Tindale*, 1991; *Falkowski et al.*, 1998], thereby affecting productivity and influencing the carbon cycle and eventually atmospheric greenhouse gas content [*Archer et al.*, 1998; *Mahowald et al.*, 2005].

Not only does dust affect climate but also the generation and transport of dust itself are extremely sensitive to climate and environmental change. As recorded in ice cores [*Thompson and Mosley-Thompson*, 1981; *Steffensen*, 1997; *Lambert et al.*, 2008] and marine [e.g., *Winckler et al.*, 2008] and terrestrial sediments (e.g., loess) [*Kohfeld and Harrison*, 2001; *Derbyshire*, 2003; *Stevens and Lu*, 2009; *Újvári et al.*, 2010; *Lambert et al.*, 2015], there have been large and systematic variations in dust loading during the past glacial-interglacial cycles with dust concentrations during the Last Glacial Maximum (LGM, 19–26 ka) being an order of magnitude higher than during the Holocene [*Fuhrer et al.*, 1999; *Ruth et al.*, 2003; *Fischer et al.*, 2007; *Albani et al.*, 2015]. The possible reasons for enhanced dust flux include increased wind speeds, reduced strength of the hydrological cycle, expansion of dust source areas, and the physiological effects of low atmospheric CO_2 concentration on terrestrial plant productivity [*Yung et al.*, 1996; *Harrison et al.*, 2001; *Tegen*, 2003]. Aeolian dust has long been recognized as an important tracer for large-scale atmospheric circulation. Since ice core dust is purely aeolian in origin, discrimination of its potential source region(s) would contribute to a better

understanding of the past dust activity and climatic/environmental causes. Furthermore, ice core dust source information provides critical experimental constraints for model simulations of the past atmospheric circulation patterns [*Svensson et al*., 2000; *Bory et al*., 2002].

Previous studies on the clay mineralogical and $^{87}Sr/^{86}Sr$ and $^{143}Nd/^{144}Nd$ isotopic compositions of dust in the Greenland Ice Sheet Project (GISP2) and Greenland Ice Core Project (GRIP) ice cores and potential source area (PSA) samples by *Biscaye et al*. [1997] and *Svensson et al*. [2000] suggest that the last glacial dust in central Greenland ice cores likely originated from Asia. The Sr–Nd isotopic signature of Greenland ice core dust has been explained as binary mixtures of Chinese/Asian dust and circum-Pacific volcanic material, while both North America and the Sahara were eliminated as PSAs because of clay mineralogical and Sr–Nd isotopic considerations [*Biscaye et al*., 1997; *Svensson et al*., 2000]. This hypothesis of Asian sources of mineral dust has become the prevailing current paradigm for Greenland dust source and informs climate models and reconstructions over the past dust dynamics and atmospheric conditions during the last glacial period.

However, *Svensson et al*. [2000] noted that important dust source areas may exist from which no PSA samples were analyzed so far, while some other studies have suggested a different or more complicated picture for Greenland dust sources. *Burton et al*. [2007] found less radiogenic Sr isotopic signatures in LGM dust from the GRIP ice core compared to those published by *Biscaye et al*. [1997] and *Svensson et al*. [2000] and concluded that the composition of dust transported to Greenland in cold periods is influenced by remote sources like the Gobi or Sahara deserts. A model simulation by *Werner et al*. [2002] suggests that a combination of Caspian Sea-Asian sources accounted for 42% of the total central Greenland dust flux during the LGM, but they emphasize that other sources could also make a significant contribution to the Greenland dust deposition. At the same time, *Aleinikoff et al*. [2008] raise the possibility of North American dust transport to Greenland based on Pb isotopic data of detrital potassium feldspars from loess. Model simulations by *Mahowald et al*. [2006] suggest that most of the dust deposited at Greenland could come from sources in the continental U.S., Alaska, or Siberia, while in their most recent study, *Mahowald et al*. [2011] simulate both East Asian and Alaskan sources for LGM dust in the GRIP ice core. More recently, *Újvári et al*. [2012] revealed that Sr–Nd isotopic compositions of $<5\,\mu m$ grains of Hungarian LGM loess samples in east central Europe partly overlap with those of coeval dust samples from the GISP2 and GRIP ice cores. These inconsistencies mean that it is now critical to systematically test between the various possible dust sources relevant for Greenland during the last glacial period.

Here we present new clay mineralogical and $^{87}Sr/^{86}Sr$, $^{143}Nd/^{144}Nd$, and $^{176}Hf/^{177}Hf$ isotopic data from fine separates ($<10\,\mu m$) of 11 LGM loess samples collected around the Northern Hemisphere (Table S1 in the supporting information) and compare them to existing LGM Greenland ice core dust data (see Figure 1 for position of new and published samples used in the analysis). Based on these new data and the published evidence, we demonstrate that there are two equally plausible explanations for the origin of the last glacial mineral dust recovered from the GISP2/GRIP ice cores. We emphasize that currently no unique source discrimination is possible using both the published and our new data. This underscores the urgent need to reconsider a wider range of possible Greenland dust sources, to incorporate this source uncertainty in modeling, and also to identify more diagnostic dust tracers.

2. Material and Methods

Fine ($<10\,\mu m$) separates of Northern Hemisphere loess sediments were used as PSA samples in this study. All the details of sampling, pretreatments, size separations, and clay mineralogical as well as Sr–Nd–Hf isotopic measurements are given in the supporting information file associated with this paper.

3. Results and Discussion

3.1. Clay Mineralogical Constraints on Possible Dust Sources

Clay mineralogy is a potentially useful tracer of ice core dust provenance, as its distribution on the continents is a first-order function of weathering, itself largely a function of climate [*Biscaye et al*., 1997]. Several clay and other trace mineral distributions are latitude dependent. The most sensitive indicator of this latitude dependency is the kaolinite to chlorite (K/C) ratio [*Biscaye*, 1965], which is an indication of the relative intensities of chemical to physical weathering processes. In a tropical climate we would expect the K/C ratio

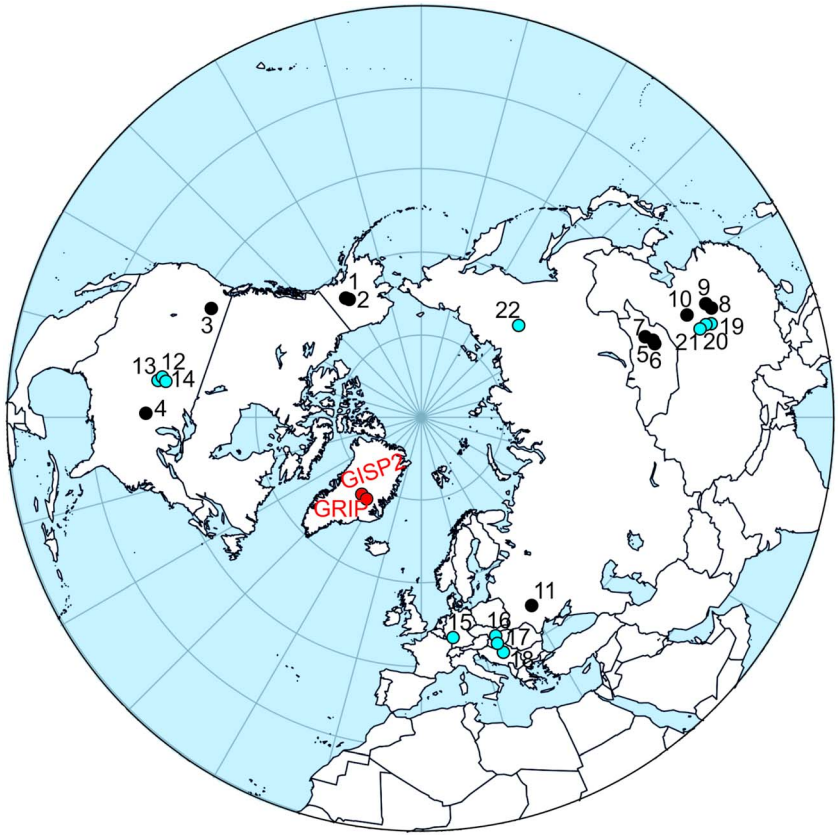

Figure 1. Location of ice cores in Greenland and potential source area (PSA) samples around the Northern Hemisphere. Black dots denote PSAs analyzed by *Biscaye et al*. [1997] and *Svensson et al*. [2000]: 1 and 2. Moose Mountains, AK, U.S.; 3. Pullman, WA, U.S.; 4. Pancake Hollow, IL, U.S.; 5–7. Gobi desert; 8. Weinan, China; 9. Luochuan, China; 10. Yulin, China; and 11. Muzichi, Kijev, Ukraine. Blue dots denote PSA samples analyzed in this study: 12. Judkins, NE, U.S.; 13. Prairie Lake, NE, U.S.; 14. Obert, NE, U.S.; 15. Nussloch, Germany; 16. Mende, Hungary; 17. Dunaszekcső, Hungary; 18. Titel, Serbia; 19. Lingtai, China; 20. Xifeng, China; 21. Beigoyuan, China; and 22. Tumara Valley, NE-Siberia, Russia.

to be >1–2 [*Scheuvens et al*., 2013], whereas in a boreal climate it is <0.5–1 [*Griffin et al*., 1968]. Beyond latitude-indicating species, any source area has a characteristic spectrum of clay minerals, partly reflecting local lithology and climate and drainage characteristics. Source area discrimination is therefore a matter of comparing relative abundances of an entire suite of minerals, some of which may be diagnostic, while others are conversely too common across possible source areas and thus noncharacteristic [*Biscaye et al*., 1997].

In our combined new and literature-based data set, the K/C ratios of European and Alaskan loess deposits and some Chinese loess samples overlap with those of the last glacial Greenland dust (0.3–0.8; Figure 2). Only North American PSAs exhibit much higher K/C ratios (>3.6; Table S2), primarily because of extremely low chlorite contents found in Nebraska loess. In terms of content, all the PSA samples analyzed so far have lower kaolinite contents than Greenland dust, except for the Alaskan loess. While the illite contents of Alaskan, central European, and Siberian loess samples are in the same range as Greenland LGM dust (40–60%), some Chinese loess samples reveal higher illite contents (Figure 2 and Table S2). Ice core dust samples are poor in smectite (2–5%) similar to Chinese, Siberian, and Alaskan PSAs (Figure 2 and Table S2) [*Svensson et al*., 2000]. Central/east central (C/EC) European loess in contrast has higher smectite contents (20–30%), while Nebraska loess was found extremely rich in smectite (except for Jud).

Based on clay mineralogy alone, only the continental U.S. PSAs with very high K/C ratios and extremely high smectite contents can be excluded as potential sources. Alaskan and Chinese loess samples seem to be the best candidates in terms of clay mineral compositions, although Chinese loess has higher illite and lower kaolinite contents and K/C ratios than Greenland dust in our data sets. Siberian and C/EC European loess

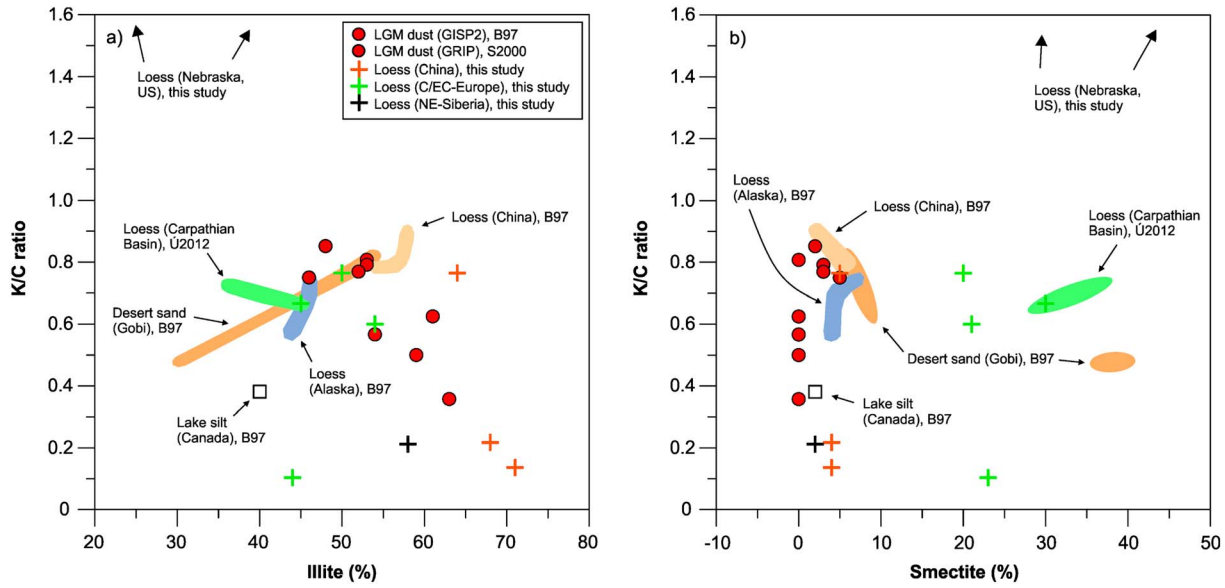

Figure 2. Kaolinite/chlorite (K/C) versus (a) illite and (b) smectite contents of PSA and Greenland LGM dust samples. Data sources: B97—*Biscaye et al.* [1997], S2000—*Svensson et al.* [2000], and *Ú2012—Újvári et al.* [2012]. Error bars are not presented, but the overall uncertainty is ~±10%.

deposits are still considered potential sources, although the former has slightly lower kaolinite, while the latter has higher smectite contents. A crucial problem in discriminating the provenance of Greenland dust is that relatively little is known about the impact of long-range transport on mineralogical compositions. While there is some evidence that the illite/kaolinite ratio remains stable during emission and long-range transport of mineral aerosol [*Glaccum and Prospero*, 1980; *Caquineau et al.*, 1998], smectite tends to agglutinate during transport due to its high hygroscopic capacity [*Singer et al.*, 2004]. The very fine grain size of smectite at emission and its tendency to form aggregates during atmospheric transport result in a fractionation effect [*Tomadin et al.*, 1989; *Scheuvens et al.*, 2013]. While smectite occurrence exhibits a latitudinal dependence in atmospheric dust samples collected along the western coast of Africa [*Stuut et al.*, 2005], *Chester et al.* [1972] found high variability in smectite contents (2–43 wt %) and attributed this as likely resulting from physical fractionation processes during emission, transport, and deposition, as well as heterogeneous smectite contents in the source sediments [*Schütz and Sebert*, 1987]. Thus, without a deeper understanding of this fractionation and source heterogeneity, the usefulness of smectite as a source marker remains uncertain [*Scheuvens et al.*, 2013], particularly in source discriminations based on loess samples exhibiting relatively little (10–20%) interregional differences in smectite contents. At the same time, large and significant differences in smectite contents can still to some extent be considered source diagnostic, in particular if other clay mineralogical parameters (e.g., K/C ratio) are also divergent. *Aleinikoff et al.* [2008] argued that higher smectite contents in continental U.S. dust deposits do not necessarily rule these out as sources for central Greenland last glacial dust, as smectite in Nebraska loess source rocks forms hard-to-remove coatings on larger grains that would drop out near the source. Nevertheless, it seems still unlikely that fractionation processes during transport would lead to 60–70% smectite depletion for the Nebraska loess, which would be required for it to match the smectite contents of LGM Greenland dust. In any case, while the interpretation of smectite content is somewhat ambiguous, neither the very low kaolinite and chlorite contents nor the high K/C ratios favor continental U.S. sources.

3.2. Sr–Nd Isotopic Compositions and Mixing Models: Two Plausible Explanations

$^{87}Sr/^{86}Sr$ isotopic ratios cover a range from 0.715178 to 0.722874, with the Nebraska loess exhibiting the least radiogenic and Chinese loess the most radiogenic Sr isotopic compositions (Table S2). $^{143}Nd/^{144}Nd$ isotopic ratios were found in a relatively narrow range from 0.512044 to 0.512160 ($\varepsilon_{Nd}(0)$: −11.4 to −9.2; Table S3; for the definition of $\varepsilon_{Nd}(0)$ see the supporting information). Less radiogenic Nd isotopic compositions are characteristic for one of the Nebraska and Siberian loess samples. In Sr–Nd isotopic space, the C/EC European loess deposits most overlap with the last glacial Greenland dust samples (Figures 3a and 3b), and while the

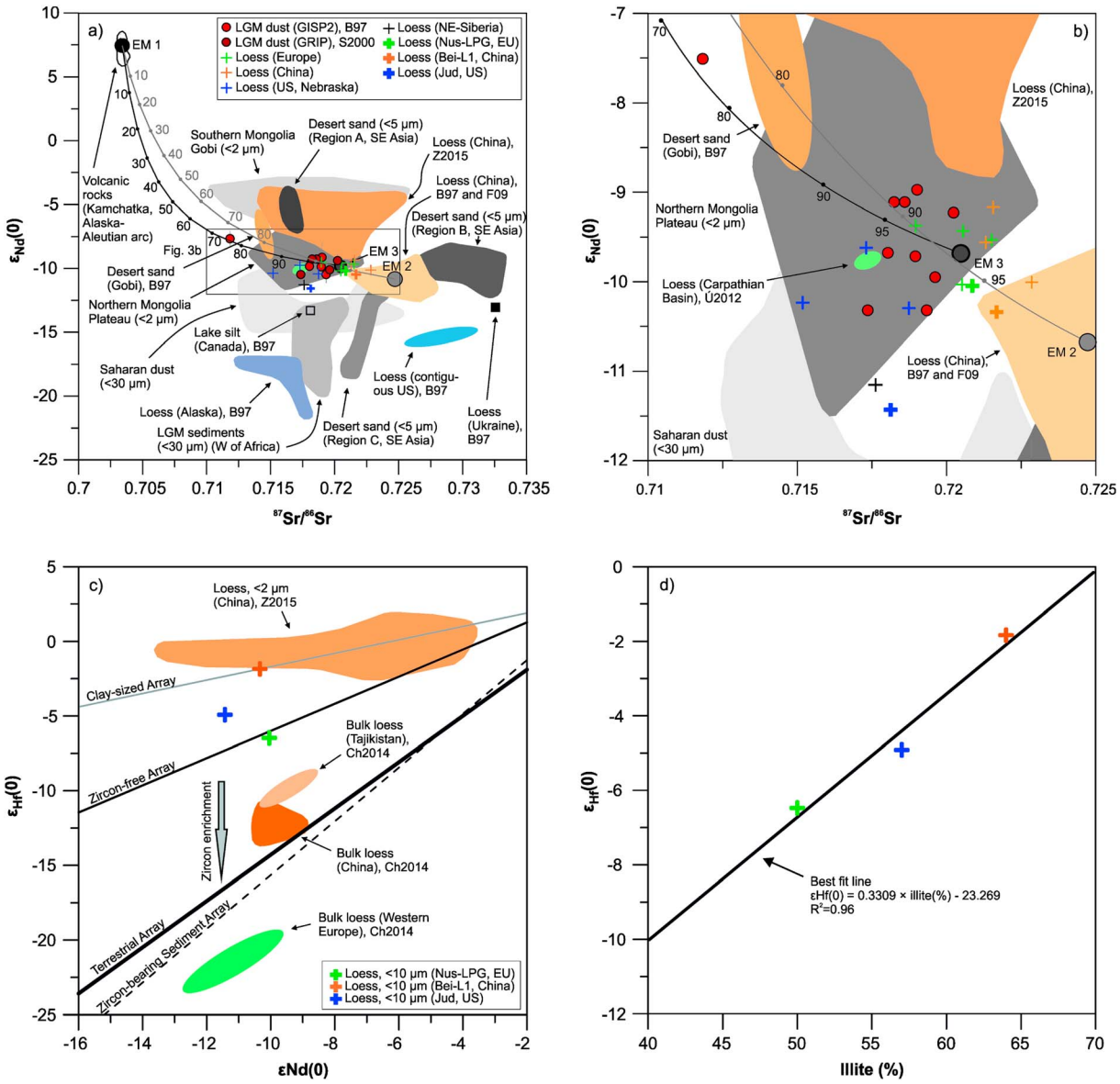

Figure 3. (a) Sr–Nd isotopic compositions of the last glacial ice core dust and PSA samples. (b) Boxed area around Greenland dust and likely potential sources enlarged. (c) Nd–Hf isotopic compositions of PSA samples analyzed in this study and comparison with those of bulk loess and clay separates from the literature. (d) PSA sample illite content versus $\varepsilon_{Hf}(0)$ diagram. Note that error bars are not shown for the sake of figure clarity, but they are mostly smaller than symbols of PSAs. Crosses denote isotopic data measured by thermal ionisation mass spectrometry (TIMS) at the UV, Vienna, while bold crosses mark those obtained by TIMS/multicollector-inductively coupled plasma–mass spectrometry (MC-ICP-MS) at RHUL, London, and/or Leeds. All displayed data are from acetic acid-treated loess size separates (<2 μm/<10 μm). Data sources for loess in Figures 3a and 3b: B97—*Biscaye et al.* [1997], S2000—*Svensson et al.* [2000], F09—*Feng et al.* [2009], Ú2012—*Újvári et al.* [2012], and Z2015—*Zhao et al.* [2015]. Isotopic data of desert sand regions A, B, and C are from *Chen et al.* [2007]. LGM sediments (west of Africa) and Saharan dust data originate from *Grousset et al.* [1988, 1992, 1998]. Circum-Pacific volcanic rocks isotopic data are from *Kepezhinskas et al.* [1997] and *George et al.* [2003]. Black and gray hyperbolas are mixing lines between end-members EM_1, EM_2, and EM_3. For end-member compositions and mixing calculations see Text S1 (supporting information). Data sources for Figure 3c: bulk loess—*Chauvel et al.* [2014] and clay fractions of loess in China—*Zhao et al.* [2015]. The terrestrial, the zircon-free/zircon-bearing sediment, and the clay-sized arrays are from *Vervoort et al.* [2011], *Bayon et al.* [2009], and *Zhao et al.* [2014].

Nebraska loess is slightly less radiogenic in Sr–Nd isotopes and Chinese loess reveals a slightly more radiogenic Sr isotopic signature (Figures 3a and 3b), both are also close to Greenland dust composition. However, neither of the Chinese-Mongolian desert sources overlap in Sr–Nd isotopic compositions with those of Greenland LGM dust [*Chen et al.*, 2007; *Zhao et al.*, 2015], except for the Northern Mongolia Plateau clay (<2 μm) separates. As such, it is also plausible that this latter region may have provided mineral dust to Greenland during the last glacial period. However, as explained below, we propose that two potential dominant dust source regions can

best account for the new data here and the published work: China and C/EC Europe. Furthermore, we argue that neither of these PSAs can be distinguished as the dominant LGM Greenland dust source based on these and all published data.

A complicating factor in the analysis is that although the sampled GISP2 and GRIP ice core sections were free of volcanic markers, a low amount of volcanogenic component cannot be entirely excluded because of the weak electric conductivity signal in alkaline ice [*Hammer et al*., 1997; *Svensson et al*., 2000]. Therefore, both *Biscaye et al*. [1997] and *Svensson et al*. [2000] considered a weak (up to ~10%) contribution from circum-Pacific volcanic sources situated upwind of Greenland. Considering that, as found by *Svensson et al*. [2000], an admixture of 90–10% from Chinese loess and circum-Pacific volcanic material would account for the Sr–Nd isotopic ratios of central Greenland LGM dust (Figures 3a and 3b; gray mixing hyperbola). However, at the same time *Svensson et al*. [2000] argue that it seems unlikely that more than half of their samples contain almost the same fraction of volcanic material, and the low intersample Sr isotopic variability implies an insignificant volcanic component. Considering this negligible volcanic input, our new Sr–Nd isotopic data set from PSAs offers a potential alternative model involving C/EC Europe that is capable of explaining the ice core dust Sr–Nd isotopic compositions. A mixture of a 0 to 5% volcanic component and 95–100% C/EC European loess would yield the Sr–Nd isotopic ratios of LGM dust in GISP2 and GRIP (Figures 3a and 3b; black mixing hyperbola). Furthermore, in this model the isotopic composition of C/EC European loess itself explains the $^{87}Sr/^{86}Sr$ and $^{143}Nd/^{144}Nd$ isotopic ratios of ice core dust without invoking a volcanic source.

This latter C/EC European dust source hypothesis would, however, require a direct dust transport route from Europe to Greenland. Such a route, as modeled for the LGM by *Andersen et al*. [1998], is through strong northward advection over the central part of Eurasia, across the Arctic Ocean, and around a persistent low pressure system in the Baffin Bay. This meridional path over the Arctic Ocean was found in an air mass trajectory analysis to Summit, Greenland for the present day [*Kahl et al*., 1997], albeit with a rare occurrence. In that study though, North America was found to be an important source for all seasons, while for wintertime 67% of all 10 day trajectories reached back to Asia/Europe. It follows from this study that Asia is also among the most important sources today and this might be true for the LGM too. Indeed, some LGM atmospheric simulations favor Asian sources for the last glacial dust over central Greenland [*Reader et al*., 1999; *Werner et al*., 2002; *Mahowald et al*., 2011], while others suggest dust transport from glaciogenic/nonglaciogenic sources in the continental U.S., Alaska, or Siberia [*Mahowald et al*., 2006].

A further line of evidence is the origin of volcanic material in Greenland ice. Tephra studies on Greenland ice cores indicate that, beyond a clear dominance of proximal Jan Mayen and Icelandic sources, volcanic ash from the Pacific Arc was repeatedly transported and deposited in Greenland [*Abbott and Davies*, 2012; *Bourne et al*., 2015a, 2015b]. Together with this Pacific material, volcanic dust from only some Southern European eruptions (Vesuvius, 79 A.D. and perhaps Santorini, ~1645 B.C.) can be traced in Greenland ice cores [*Abbott and Davies*, 2012], and neither the Campanian Ignimbrite (~40 ka) nor the Laacher See (~12.9 ka) tephras could be identified in cores of the North Greenland Ice Core Project (NGRIP) and North Greenland Eemian Ice Drilling (NEEM) project [*Svensson*, 2012; *Bourne et al*., 2013]. Out of the numerous North American eruptions, only the Alaskan White River Ash (~860 A.D.) and the Mount Mazama Ash (~7.6 ka) appear in NEEM [*Jensen et al*., 2014] and GISP2 [*Zdanowicz et al*., 1999]. This data therefore point to a dominant dust transport pathway from East Asia to Greenland during the last glacial period. As shown above, such a link is supported by the Sr–Nd isotopic signatures of Chinese loess deposits and in general by their clay mineralogy. However, since the illite contents of Chinese loess are by 10–25% higher than those of ice core dust, some loss during transport may have happened. Although less consistent with tephra records and atmospheric circulation simulations, the C/EC European loess Sr–Nd signatures strongly point to a LGM Greenland dust source in this region. The clay mineral compositions are entirely overlapping, except for smectite, which is present in slightly higher amounts in C/EC European loess than in ice core dust. Nevertheless, these differences are minor (10–20%, mostly within X-ray diffraction (XRD) measurement uncertainties), and some amounts of smectite may have been lost during transport.

While the published and new data here cannot differentiate between Asian and C/EC European sources, we argue that the possibility of a dominant North American and Siberian dust source to Greenland can be excluded. With regard to published work, sourcing of Greenland material from Alaskan dust was rejected by *Biscaye et al*. [1997] using Sr–Nd isotopes. Indeed, a genetic link between Alaskan and Greenland dusts

is improbable considering the previously published Sr–Nd isotopic signatures of Yukon loess fine grain separates ($^{87}Sr/^{86}Sr$: 0.707–0.71, $\varepsilon_{Nd}(0)$: −4 to 2) [*Zdanowicz et al.*, 2006]. Likewise, the sourcing of Greenland dust from the continental U.S. seems also unlikely based on the clay mineralogy presented here, especially given the very low kaolinite and chlorite contents in Nebraska loess, which could clearly not be enriched en route to Greenland during transport (see above; Figure 2). Further evidence against the continental U.S. origin comes from the less radiogenic Sr–Nd isotopic ratios of Nebraska loess compared to those of central Greenland LGM dust in the data presented here (Figure 3). Although few of the samples overlap, the more radiogenic Sr–Nd isotopic ratios of ice core dust could only be accounted for by mixing Nebraska loess with dust from a source characterized by much more radiogenic Sr–Nd isotopic ratios than the ice core dust. At the same time, any mixture of Nebraska loess with volcanic products would result in much lower $^{87}Sr/^{86}Sr$ isotopic ratios than those of ice core dust. With regard to a possible NE-Siberian origin, this region cannot be excluded as a source based on clay mineralogy. However, the less radiogenic Nd isotopic signature of the Sib200 sample does not support a link to Greenland from this region (Figures 3a and 3b and Table S3). Nevertheless, further loess samples should be analyzed to gain insight into the mineralogical and isotopic heterogeneity of these sources.

3.3. Hf Isotopes as Promising Tracers

The ambiguities in source discrimination presented above imply that new approaches are needed to answer the still open question of the last glacial ice core dust origin in central Greenland. Hf isotopic analysis on the low amounts of material (0.7–8 mg dust/kg ice) that are usually found in ice cores from the LGM to the Holocene [*Ruth et al.*, 2003] is challenging, but feasible [*Aciego et al.*, 2009]. Here we present some initial Hf isotopic analyses on the $<10\,\mu m$ fraction of LGM loess samples. These data coupled with Nd–Hf isotopic ratios from the recent literature (Figure 3c) suggest that such efforts are valuable as it is expected that the Hf isotopes will provide more source-diagnostic data.

First, we tested the effect of acid treatment on Hf isotopes, as all of the PSAs were leached using weak acetic acid to remove carbonates (see the supporting information). Since carbonates are not the carriers of Lu and/or Hf, it was expected that the Hf isotopic signatures are not influenced by the acetic acid treatment. Indeed, similar to Nd isotopes, Hf isotopic ratios do not seem to be affected by weak acetic acid leaching (Table S3).

Second, we analyzed the range of Nd–Hf isotopic data in the samples. As shown in Figure 3c and Table S3, Nd isotopic compositions ($\varepsilon_{Nd}(0)$) of the three acid-treated grain size separates analyzed in this study range from −11.4 to −10, close to the average upper crustal value of −10.3 [*Chauvel et al.*, 2014]. Bulk loess samples from China, Tajikistan, and western Europe have almost the same $\varepsilon_{Nd}(0)$ values as the grain size separates, although with a slightly larger range (Figure 3c). In contrast, bulk loess $\varepsilon_{Hf}(0)$ values show large variations and are always much more negative than those from the loess fine grain separates. These much less radiogenic Hf isotopic compositions of the bulk loess are attributed to higher zircon abundances in these samples [*Patchett et al.*, 1984; *Pettke et al.*, 2002; *van de Flierdt et al.*, 2007; *Lupker et al.*, 2010; *Rickli et al.*, 2010; *Garcon et al.*, 2013; *Chauvel et al.*, 2014], meaning these data are clustered around the "zircon-bearing sediment array" [*Bayon et al.*, 2009]. Zircon is a refractory heavy mineral with low Lu/Hf ratios [*Hoskin and Schaltegger*, 2003] and thus very unradiogenic $^{176}Hf/^{177}Hf$ isotopic compositions [*Kinny and Maas*, 2003; *Újvári and Klötzli*, 2015], which then also consequently dominate the bulk loess Hf isotopic signal. In contrast, fine grain separates of loess appear between the "zircon-free" and "clay-sized arrays" (Figure 3c), demonstrating that they contain little to no zircon, and their more radiogenic Hf isotopic compositions are controlled by clay minerals that incorporate/adsorb radiogenic Hf released from higher Lu/Hf phases during incongruent weathering [*van de Flierdt et al.*, 2007; *Bayon et al.*, 2009; *Zhao et al.*, 2014]. In our limited data set, the Hf isotopic compositions of fine grain separates are found to be strongly correlated with illite contents (Figure 3d), with the differences in $\varepsilon_{Hf}(0)$ values (−6.5 to −1.8) between samples reaching several ε_{Hf} units. We therefore suggest that in the absence of zircons, the Hf isotopic compositions likely reflect the different proportions of clay minerals characteristic for each loess region on the Northern Hemisphere, in particular illite. In our data set these proportions appear to be sufficiently different in the fine grain PSA data to enable discrimination. Further, the Hf isotopic composition of very fine mineral dust in Greenland is also expected to be clay mineralogy dependent because of zircon depletion and fallout during long atmospheric transport [*Aarons et al.*, 2013]. As such, it is believed that through Hf isotopes a better understanding can be gained concerning Greenland dust sources, although for this to be achieved, further clay mineralogy and Hf isotopic data are needed to better understand the first-order clay mineralogical controls on Hf isotopic signatures of fine dust.

4. Conclusions

Clay mineralogy and Sr–Nd isotopic ratios of grain size separates (<10 μm) of Northern Hemisphere loess samples demonstrate that two plausible scenarios exist for the origin of the last glacial mineral dust found in the GISP2 and GRIP ice cores. While the Northern Mongolia and Chinese Loess Plateau deposits still appear to be the most likely dust sources, the Sr–Nd isotopic compositions of LGM ice core dust can readily be explained by major contributions from C/EC European loess deposits too. Clay mineralogical compositions do not seem to contradict this hypothesis. At the same time, the clay mineralogy coupled with the less radiogenic Sr–Nd isotopic signatures of Nebraska loess does not support a sourcing of Greenland dust from major continental North American glacial dust-emitting regions. Likewise, a NE-Siberian origin also seems less likely based on the Nd isotopic ratio of a single sample, although further data would be needed from dust samples of both regions. Based on initial Hf isotope analyses of fine separates of three loess samples, an apparent dependence of Hf isotopic signatures on the relative proportions of radiogenic clay minerals (primarily illite) can be seen, as these fine dust fractions seem to be zircon free. The observed difference between major potential source regions in $^{176}Hf/^{177}Hf$ that reach several ε_{Hf} units and the first-order clay mineralogy dependence of Hf isotopic signatures means that there is strong potential for distinguishing between the two hypothesized Greenland dust sources using Hf isotopes. For this, however, Hf isotopic measurements on Greenland ice core dust samples are required.

Author Contributions

G.Ú. designed the study and performed the field work and sampling in Hungary. M.R.S., M.G., G.L.B.W., T.S., S.B.M., and M.Z. did field work and sampling in the U.S., Germany, China, Serbia, and Siberia and provided samples. G.Ú. and J.K. performed laboratory preparations (wet sedimentation and acid treatment) and laser diffraction measurements. N.T. did the XRD analyses, while U.S.K. and C.M. performed the Sr–Nd–Hf isotopic analyses in Vienna, London, and Leeds. G.Ú. wrote the paper with the active participation of T.S. and A.S. All authors contributed to the discussion/interpretation of results.

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