

Constraints from loess on the Hf–Nd isotopic composition of the upper continental crust

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Abstract

 Knowledge of the average composition of the upper continental crust is crucial to establish not only how it formed but also when. While well constrained averages have been suggested for its major and trace element composition, no values exist for its Nd and Hf isotopic compositions even though radiogenic isotopic systems provide valuable information on its average model age.

 Here we present Nd and Hf isotopic data determined on a large number of loess deposits from several continents. We demonstrate that these deposits have very uniform Nd and Hf isotopic compositions. We obtain an average Nd isotopic composition that is similar to previous estimates for the upper continental crust (ε_{Nd} = -10.3±1.2 (1 σ)) and we establish a new Hf average value at ε_{Hf} = -13.2±2 36 (1σ). This average falls on the "Terrestrial Array", demonstrating that the two parent-daughter ratios are not decoupled during crust formation. Trace element data acquired on the same set of samples allow us to calculate an average $147Sm/$ ¹⁴⁴Nd ratio for the upper continental crust: 0.1193 ± 0.0026 , a value slightly higher than previous estimates. Based on the relationship between Sm/Nd and Nd isotopes, we estimate the average Nd extraction age of upper 42 continental crust from the depleted mantle at $T_{DM}(Nd) = 1.82\pm0.07$ Ga. This model age is entirely consistent with previous suggestions made for example by Goldstein et al. [\[1984\]](#page-18-0).

 Assuming that for each individual sample, the Hf model age cannot be younger than the Nd model age, our new Nd-Hf isotopic data provide a value for the very 47 poorly known Lu/ 177 Hf ratio of the upper crust. Our estimate is 176 Lu/ 177 Hf = 0.0125±0.0018, a value significantly lower than commonly used values [0.0150 – 0.0159, *[Griffin et al.](#page-18-1)*, 2002; *[Goodge and Vervoort](#page-18-2)*, 2006; *[Hawkesworth et al.](#page-19-0)*, 2010] but higher than Rudnick and Gao's [\[2003\]](#page-20-0) estimate of 0.0083. The impact of our 51 new Lu/ 177 Hf ratio on crustal model ages of zircon populations is not simple to evaluate but the Hf model ages calculated with this new Lu/Hf ratio could be younger by up to 500 Ma.

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1 - Introduction

 The formation of the continental crust was certainly one of the most important events that affected the overall composition of the Earth, because the isolation at the surface of a light and enriched layer left behind a depleted residue in the mantle. This observation explains why so many studies concentrate on the processes that produce continental crust and on the timing of crustal growth [e.g., *[Armstrong](#page-16-0)*, 1968; *[McCulloch and Wasserburg](#page-19-1)*, 1978; *[Armstrong](#page-16-1)*, 1981; *[Allègre and Rousseau](#page-16-2)*, 1984; *[Goldstein et al.](#page-18-0)*, 1984; *[Taylor and McLennan](#page-20-1)*, 1985; *Condie*[, 1993;](#page-17-0) *[Wedepohl](#page-21-0)*, 1995; *[Gallet et al.](#page-18-3)*, 1998; *[Vervoort et al.](#page-21-1)*, 2000; *[Griffin et](#page-18-1) al.*[, 2002;](#page-18-1) *[Rudnick and Gao](#page-20-0)*, 2003; *[Harrison et al.](#page-19-2)*, 2005; *[Hawkesworth and Kemp](#page-19-3)*, [2006;](#page-19-3) *[Blichert-toft and Albarède](#page-17-1)*, 2008; *[Belousova et al.](#page-17-2)*, 2010; *[Hawkesworth et al.](#page-19-0)*, [2010;](#page-19-0) *[Kemp et al.](#page-19-4)*, 2010; *[Dhuime et al.](#page-17-3)*, 2011; *[Garçon et al.](#page-18-4)*, 2011; *[Iizuka et al.](#page-19-5)*, [2013\]](#page-19-5). In this study, we focus on the information provided by two isotopic systems, Sm-Nd and Lu-Hf. The two systems are strongly coupled during magmatic processes leading to the definition of the "Terrestrial array", an array along which all terrestrial magmas fall [*[Vervoort et al.](#page-21-2)*, 1999; *[Vervoort et al.](#page-20-2)*, [2011\]](#page-20-2). However, even though crustal magmas fall on the "Terrestrial Array" the location of the average composition of the continental crust remains unknown.

 Knowing the position of present-day average crust on the Terrestrial array is important because it provides information not only on the processes active during the formation of the crust but also because the two isotopic systems record the past history of crustal materials and can be used to calculate an average extraction age of crust from the mantle. Finally, by using the constraints provided by the Sm-Nd isotopic system and those coming from the Lu-Hf isotopic 83 system, we can evaluate the average crustal $176Lu/177Hf$ ratio, a very poorly known but crucial parameter necessary to calculate crustal residence ages of zircon populations [e.g.,*[Griffin et al.](#page-18-1)*, 2002; *[Harrison et al.](#page-19-2)*, 2005; *[Kemp et al.](#page-19-6)*,

 2007; *[Blichert-toft and Albarède](#page-17-1)*, 2008; *[Belousova et al.](#page-17-2)*, 2010; *[Kemp et al.](#page-19-4)*, 2010; *[Condie et al.](#page-17-4)*, 2011; *[Yao et al.](#page-21-3)*, 2011; *[Iizuka et al.](#page-19-5)*, 2013].

 To address all these issues, we focus on a combined Nd and Hf isotopic study of a variety of recent loess deposits from several continents. Loess deposits result from the local accumulation of windblown dust coming from usually large continental areas. They represent natural mixtures of erosion products of continental surfaces and were therefore used by several previous workers to determine the average composition of the upper continental crust [e.g., *[Taylor](#page-20-3) [and McLennan](#page-20-3)*, 1981; *[Taylor et al.](#page-20-4)*, 1983; *[Taylor and McLennan](#page-20-1)*, 1985, [1995;](#page-20-5) *[Barth et al.](#page-16-3)*, 2000]. We show that they are excellent proxies for the average composition of large areas of upper continental crust for both the Sm-Nd and the Lu-Hf isotopic systems. We then suggest an average value for the upper continental crust for the two systems and we determine an average model age. Finally, the combined Nd and Hf isotopic data allow us to suggest a value for the Lu/ 177 Hf ratio of the upper continental crust.

2 - Source of samples

 The samples in this study were collected from several locations around the world: Western Europe, Tajikistan, China and Argentina (see Figure 1). Most were previously studied by Gallet and co-workers [*[Gallet et al.](#page-18-5)*, 1996; *[Gallet et al.](#page-18-3)*, [1998;](#page-18-3) *[Jahn et al.](#page-19-7)*, 2001] who reported major and trace element data as well as Nd and Sr isotopic compositions for loess samples from China, Western Europe and Argentina. The Tajikistan loess deposits come from the Chashmanigar section and were previously studied by Ding et al. [\[2002\]](#page-18-6) and by Yang et al. [\[2006\]](#page-21-4) who used sedimentological and major element data to constrain climate conditions during the past 1.8 Ma. However, they did not perform any trace element or isotopic analyses. Finally, two samples of Sahara dust collected in southern France were also analyzed.

 The samples belong to two different types of loess deposits. The Western Europe Loess samples are periglacial deposits that were produced during the last glacial period by mechanical erosion of the crust followed by transport by wind over limited distances [*[Haase et al.](#page-18-7)*, 2007]. The situation is rather similar for the Argentinian loess deposits whose origin is mainly the Andes [*[Smith et al.](#page-20-6)*, 2003]. The Chinese and Tajikistan loess deposits represent much larger volumes and have a very different origin: the materials come from large desert areas. For the Chinese loess, particles come mainly from the deserts located Northwest in Mongolia [*Sun*[, 2002\]](#page-20-7); in southern Tajikistan, the particles originate from the Karakum and Kyzylkum deserts located northwest in Turkmenistan, Uzbekistan and Kazakhstan [*[Ding et al.](#page-18-6)*, 2002]. In both cases, the deserts developed on continental crust that formed over large periods of geological time [e.g., *[Lee et al.](#page-19-8)*, [2011;](#page-19-8) *[Yoshida](#page-21-5)*, 2012] and particles were transported over long distances by high- energy winds to accumulate in protected areas [*[Ding et al.](#page-17-5)*, 1999; *[Ding et al.](#page-18-6)*, [2002;](#page-18-6) *[Guo et al.](#page-18-8)*, 2002]. Consequently, they represent the average composition of much larger crustal areas than the periglacial deposits. Finally, the dust blown from the Sahara over Southern France does not constitute a real loess deposit [*[Grousset et al.](#page-18-9)*, 2003], but it provides valuable information on the average composition of the largest desert area in the world.

3 - Analytical techniques

 All samples were crushed using an agate mortar. Trace element concentrations 136 were obtained after dissolution in a $HF-HClO₄$ mixture using Parr bombs. The complete procedure is that of Chauvel et al. [\[2011\]](#page-17-6), but the dissolution time was increased to insure complete dissolution of the very resistant minerals, an issue particularly important when using the relationship between REE and Hf concentrations and when measuring Hf isotopes. Both accuracy and reproducibility of the data are estimated as better than 5% for most trace elements, as shown by the analyses of international rocks standards and duplicate analyses of samples (see Supplementary table).

 Neodymium and hafnium were isolated from an aliquot of the solution prepared for trace element analysis and using standard ion-chromatography as described by Chauvel et al. [\[2011\]](#page-17-6). Total procedural blanks were less than 100 picograms for both Nd and Hf. The Nd and Hf isotopic compositions were measured using the Nu Plasma HR MC-ICP-MS at ENS Lyon. Mass bias corrections for Nd and Hf

149 isotopic compositions were performed using $146Nd/144Nd = 0.7219$ and $179Hf/177Hf = 0.7325$ respectively. Over the course of analyses, daily averages obtained for the Nd Ames-Rennes standard ranged between 0.511958±19 and 0.511976±14, while those obtained for the Hf Ames-Grenoble standard ranged between 0.282165±9 and 0.282168±19. All isotopic ratios reported in Table 1 are normalized to the reference values of 0.511961 and 0.282160 as published by Chauvel and Blichert-Toft [\[2001\]](#page-17-7) and Chauvel et al. [\[2011\]](#page-17-6). Complete duplicate analyses of several samples show that the reproducibility of isotopic measurements is better than 100 ppm for both Nd and Hf isotopic ratios (Table 1).

4 - Results

 New trace element data for the loess are provided in the Supplementary table. Some samples had been previously analyzed for their trace element contents by Gallet et al. [\[1998\]](#page-18-3) and by Jahn et al. [\[2001\]](#page-19-7) but these were reanalyzed because some key elements (e.g., Hf) were not measured in many of these older studies. The new values reported here are generally similar to those previously published but there are notable exceptions. For example, the new determinations of REE, Th, Zr and Hf concentrations are systematically higher than those reported by Jahn et al. [\[2001\]](#page-19-7) suggesting that dissolution of samples in 2001 was incomplete, leaving some non-dissolved zircon and monazite grains (see supplementary figure).

 Figure 2 shows the trace element patterns obtained for all loess samples. As previously shown by Gallet et al. [\[1996\]](#page-18-5) and Jahn et al. [\[2001\]](#page-19-7), the Chinese loess are remarkably homogeneous with normalized trace element concentrations close to the Upper Continental Crust values of Rudnick and Gao [\[2003\]](#page-20-0) (see Figure 2b). The new Tajikistan data shown in Figure 2c display similar features. This is not the case, however, for the periglacial loess sampled in Western Europe and shown in Figure 2a. The latter samples have much more variable concentrations and they display large positive Zr-Hf anomalies, most probably due to the presence of excess zircon. Finally, the Argentinian loess samples and the two Sahara dust samples shown in Figure 2d have trace element patterns generally parallel to the average upper continental crust (UCC) value, without positive Zr-Hf anomalies, but with more variability.

 The Nd and Hf isotopic compositions of all loess samples analyzed in this study are listed in Table 1. The data are plotted in Figure 3 where they are compared to data published in the literature for MORB, OIB, oceanic sediments and crustal rocks. With the exception of the Argentinian samples, all loess define a restricted 187 range of ε_{Nd} and ε_{Hf} values (-10.3±1.2 and -13.8±4.2) and lie on the terrestrial array of Vervoort et al. [\[2011\]](#page-20-2) in the field of crustal materials. The Argentinian 189 loess have less negative values (ε_{Nd} = -1.7 and ε_{Hf} = -2.8), a feature attributed to the abundance of Andean volcanic rocks in the source area [*[Gallet et al.](#page-18-3)*, 1998; *[Smith et al.](#page-20-6)*, 2003].

5 - Discussion

 Constraining the average composition of the continental crust is a very active field of research [e.g., *[Armstrong](#page-16-0)*, 1968; *[McCulloch and Wasserburg](#page-19-1)*, 1978; *[Armstrong](#page-16-1)*, 1981; *[Allègre and Rousseau](#page-16-2)*, 1984; *[Goldstein et al.](#page-18-0)*, 1984; *[Taylor and](#page-20-1) [McLennan](#page-20-1)*, 1985; *Condie*[, 1993;](#page-17-0) *[Wedepohl](#page-21-0)*, 1995; *[Gallet et al.](#page-18-3)*, 1998; *[Vervoort et](#page-21-1) al.*[, 2000;](#page-21-1) *[Griffin et al.](#page-18-1)*, 2002; *[Rudnick and Gao](#page-20-0)*, 2003; *[Hawkesworth and Kemp](#page-19-3)*, [2006;](#page-19-3) *[Belousova et al.](#page-17-2)*, 2010; *[Hawkesworth et al.](#page-19-0)*, 2010; *[Dhuime et al.](#page-17-3)*, 2011; *[Garçon et al.](#page-18-4)*, 2011]. Several studies have suggested average values for the upper continental crust but these mainly focus on its major and trace element composition (see Rudnick and Gao [\[2003\]](#page-20-0) for a review). Knowing its radiogenic isotopic compositions is also important since it provides constraints on the timing of crust formation through geological time. Several Pb isotope compositions have been suggested for the upper continental crust [e.g., *[Zartman](#page-21-6) [and Doe](#page-21-6)*, 1981; *[Newsom et al.](#page-19-9)*, 1986; *[Rudnick and Goldstein](#page-20-8)*, 1990; *[Garçon et al.](#page-18-10)*, [2013b\]](#page-18-10) as well as some average Nd model ages [*[Allègre and Rousseau](#page-16-2)*, 1984; *[Goldstein et al.](#page-18-0)*, 1984; *[Goldstein and Jacobsen](#page-18-11)*, 1988; *[Jacobsen](#page-19-10)*, 1988]. These show that the continental crust formed, on average, about 1.8 to 2 Ga ago. Here, we will use loess as a proxy to more precisely constrain the Nd and Hf isotopic composition of average upper continental crust. We will then use these data to re-evaluate when the upper crust separated from the mantle and use the

 combined Nd and Hf isotopic data to constrain the Lu/Hf ratio of upper continental crust. It could be argued that the sample set might not be representative of average upper crust and we will demonstrate that it is.

5.1 - Loess: an excellent proxy for the average upper continental crust Nd isotopic ratio

 Loess has been widely used by Taylor and McLennan [\[1985\]](#page-20-1) to establish the average REE composition of the upper continental crust because these sediments represent the average composition of large surface areas. Since the REE content is representative of the upper continental crust, loess can be used to evaluate its Nd isotopic composition [*[McCulloch and Wasserburg](#page-19-1)*, 1978; *[Taylor et al.](#page-20-4)*, 1983; *[Gallet et al.](#page-18-3)*, 1998].

 The measured Nd isotopic ratios of nearly 50 loess samples are remarkably uniform and define two groups (Figures 3 & 4). The first comprises only the 227 Argentinian loess and is relatively radiogenic with an average ε_{Nd} of -1.7. Gallet et al. [\[1998\]](#page-18-3) concluded that the unusual composition of these samples was due to the overwhelming presence of recent volcanic products coming from the Andes and could not be considered as representative of large areas of the upper continental crust. We concur with their conclusion. The second group of samples consists of all other analyzed loess samples (Figure 3). While the Chinese and 233 Tajikistan loess have indistinguishable isotopic compositions $(\epsilon_{Nd} = -9.82 \pm 0.44$ 234 and ε_{Nd} = -9.46±0.36, respectively), the two Sahara dusts have lower ε_{Nd} at -12.4. 235 Finally, the Western European loess are more heterogeneous and average at ϵ_{Nd} $236 = -11.25 \pm 1.28$. We attribute this larger range of isotopic compositions to processes acting during the formation of periglacial loess, when relatively small volumes of material were produced by erosion of proximal sources during the last glacial period. Such small volumes of glacial sediments were not well mixed by long distance transport and their composition is a function of the diversity of local terranes in Western Europe. In contrast, the Tajikistan and the Chinese loess are remarkably homogeneous, a feature consistent with their origin as dust blown from large desert areas [*[Ding et al.](#page-17-5)*, 1999; *[Ding et al.](#page-18-6)*, 2002]. Nevertheless,

244 taken together, all these samples define a well-constrained average at ε_{Nd} = 245 10.3±1.2 (1 σ) (see inset of Figure 3).

 Given the large number of Chinese and Tajikistan samples in our data set, it could 247 be argued that our average ε_{Nd} value is only representative of the upper 248 continental crust present in Russia and Central Asia. However, similar ε_{Nd} values have been reported for all sorts of other areas in the world: The Carpathian loess 250 deposits in Eastern Europe have an average ε_{Nd} value at -10.21 \pm 0.34 [Újvári et *al.*[, 2012\]](#page-20-9) and a placer deposit that averages the area drained by the Rhone River 252 in Western Europe has an ε_{Nd} value of -9.3 [*[Garçon et al.](#page-18-4)*, 2011]. This suggests 253 that most of the European continent has an ε_{Nd} value at about -10. The only two US loess samples analyzed for Nd isotopes have lower values at -13 and -15 [*[Taylor et al.](#page-20-4)*, 1983] which might indicate that the American upper crust could 256 have a different average value. However, the ε_{Nd} values reported by Goldstein et al. [\[1984\]](#page-18-0) and Goldstein and Jacobsen [\[1988\]](#page-18-11) for particulates collected on major rivers from the American continent do not confirm these low values but provide a range between -20.8 and -4.4. Worldwide aerosols can also be used to estimate the composition of exposed continental crust. Grousset and Biscaye [\[2005\]](#page-18-12) compiled the existing data and showed that aerosols have much more variable compositions than loess or river particulates, but they could estimate that the 263 world mean ε_{Nd} value was around -11 [*[Grousset and Biscaye](#page-18-12)*, 2005]. Finally, Goldstein et al. [\[1984\]](#page-18-0) analyzed a large number of river sediments and aeolian 265 dusts from various continents and provided an average ε_{Nd} value at -11.4 \pm 4 for all these deposits. Given the similarity of all the values listed above, we suggest 267 that our estimate (ε_{Nd} =-10.3) is probably accurate and representative of continental crust exposed today at the surface of the Earth. Estimating the 269 uncertainty is more difficult. Here we use the 1σ error based on our data set (±1.2) but we acknowledge that with a larger set of samples, the value could change.

5.2 - Loess: also a good proxy for the average upper continental crust Hf isotopic ratio

 Several authors have argued that the Zr and Hf contents of loess should not be used as analogues for their source [*[Taylor et al.](#page-20-4)*, 1983; *[Barth et al.](#page-16-3)*, 2000; *[Rudnick](#page-20-0) [and Gao](#page-20-0)*, 2003]. These arguments are based on the observation that the loess that they had studied had a very large excess of Zr and Hf relative to the REE, a feature attributed to the presence of excess zircons. All the samples that they studied, however, are periglacial loess sediments. Our Western European loess samples have the same large positive anomaly in Zr and Hf (see Figure 2) and we argue that many or all periglacial loess deposits contain this feature. In contrast, the Tajikistan, Chinese and Argentinian loess deposits as well as the Sahara dusts either have no anomaly or a slight negative anomaly (Figure 2). Since zircons are rich in Hf and have usually very unradiogenic Hf isotopes, their presence has strong impact on the whole rock Hf isotopic composition. As a result, the measured Hf isotopic ratios reported in the Table 1 for the Western European loess cannot be directly used to establish the average composition of the upper continental crust. It is, however, possible to correct for the "zircon effect" by 290 using the relationship between Nd/Hf and $\Delta \varepsilon$ _{Hf} (the vertical deviation from the terrestrial array as defined by Carpentier et al. [\[2009\]](#page-17-8)) as shown in Figure 4. 292 Nd/Hf is well constrained in all estimates of upper continental crust, while $\Delta \epsilon_H$ is a good measure of mineralogical sorting during sediment formation [*[Carpentier](#page-17-8) et al.*[, 2009;](#page-17-8) *[Vervoort et al.](#page-20-2)*, 2011; *[Garçon et al.](#page-18-13)*, 2013a]. In Figure 4, loess deposits define a positive correlation. The Western Europe loess have low Nd/Hf (i.e., positive Hf anomalies), lie below the terrestrial array of Vervoort et al. [\[2011\]](#page-20-2) 297 and have negative $\Delta \varepsilon_{Hf}$ values, as expected by the presence of excess old zircons. In contrast, the Tajikistan loess samples have relatively high Nd/Hf (i.e., slight 299 negative Hf anomalies) and slightly positive $\Delta \varepsilon$ _{Hf} values, as expected for sediments deficient in zircon relative to their crustal source [*[Carpentier et al.](#page-17-8)*, [2009;](#page-17-8) *[Vervoort et al.](#page-20-2)*, 2011; *[Garçon et al.](#page-18-13)*, 2013a]. The Chinese and Argentinian loess have compositions that plot in between these two end members. Using the values recommended for the Nd/Hf ratio of upper continental crust, we can 304 therefore estimate the $\Delta \varepsilon_{\text{H}f}$ value for average upper continental crust. The most 305 recent estimation of $(Nd/Hf)_{UCC}$ provided by Rudnick and Gao [\[2003\]](#page-20-0) constrains 306 the $\Delta \varepsilon$ _{Hf} value for upper continental crust at +1.6 with an error of less than ± 2 , 307 based on the range of estimated $(Nd/Hf)_{UCC}$ (see Figure 4).

308 This $\Delta \varepsilon_{\text{Hf}}$ value translates into an ε_{Hf} value of -13.2 ±2 (1 σ) using the average ε_{Nd} value of -10.3 determined above and the terrestrial array equation of Vervoort et 310 al. [\[2011\]](#page-20-2) $\epsilon_{\text{Hf}} = 1.55.\epsilon_{\text{Nd}} + 1.21$. Such value is not identical to the average of all 311 loess measurements (-13.8 ± 4.2) but it is similar and the error on the average is 312 smaller. These ε values correspond to a ¹⁴³Nd/¹⁴⁴Nd of 0.512102 \pm 67 and a 313 176Hf/ 177Hf of 0.282412±56 (see Table 2).

 The combined Nd and Hf isotopic values for average upper continental crust 315 define the point shown as a red star in Figure 3 at ε_{Nd} = -10.3±1.2 (1 σ) and ε_{Hf} = -316 13.2 \pm 2 (1 σ), an average that plots, within error, on the terrestrial array thereby supporting the long-standing observation that the parent-daughter ratios Sm/Nd and Lu/Hf are coupled during magmatic processes both in the mantle and the crust. It also provides a value for the average upper continental crust, a value that is very useful in modeling how the continents formed.

5.3 - Nd model age of upper continental crust

 To model the average formation age of the upper continental crust, we need well-constrained values not only for Nd and Hf isotopic compositions but also for Sm/Nd and Lu/Hf ratios. Data listed in Table 1 show that both the loess Nd 326 isotopic compositions and their $147Sm/144Nd$ ratios (0.1193 \pm 0.0026, recalculated using the Sm and Nd concentrations) are extremely constant. This feature arises from the uniform Sm/Nd ratio of most crustal minerals [*[Taylor and McLennan](#page-20-3)*, [1981;](#page-20-3) *[Goldstein et al.](#page-18-0)*, 1984; *[Taylor and McLennan](#page-20-1)*, 1985; *[Garçon et al.](#page-18-4)*, 2011]. 330 When compared to the $147\text{Sm}/144\text{Nd}$ ratio recalculated from previously published UCC estimates [*[Taylor and McLennan](#page-20-1)*, 1985; *Condie*[, 1993;](#page-17-0) *[Gao et al.](#page-18-14)*, 1998; *[Rudnick and Gao](#page-20-0)*, 2003], our value is slightly higher (0.1193 vs. ≈0.1050) a feature that may be related to the poor precision of the Sm and Nd concentration estimates used in these publications (typically 10%, see Table 2). Our value is similar, however, to values proposed by Jahn and Condie [*[Jahn and Condie](#page-19-11)*, 1995] 336 and in studies when the $147\text{Sm}/144\text{Nd}$ was determined with better precision [*[McCulloch and Wasserburg](#page-19-1)*, 1978; *[Allègre and Rousseau](#page-16-2)*, 1984; *[Goldstein et al.](#page-18-0)*, [1984\]](#page-18-0). Using this value for $147Sm/144Nd$ (0.1193±0.0026), our UCC estimate for

 $143N d/144N d$, the depleted mantle values listed in Table 2 and the following equation,

$$
T_{\rm DM} = Log \left(\frac{(\frac{143Nd}{144Nd})_{\rm UCC} - (\frac{143Nd}{144Nd})_{\rm DM}}{(\frac{147Sm}{144Nd})_{\rm UCC} - (\frac{147Sm}{144Nd})_{\rm DM}} - 1 \right) / \lambda
$$

341 (with λ =6.54*10⁻¹² year⁻¹) we can calculate a model age for the extraction of the crust from the depleted mantle.

343 We obtain a Nd model age of 1.82 ± 0.07 Ga (1σ) for the separation of crust from the mantle. Such value falls within the range of previous estimates based on Nd isotopes [e.g., *[Allègre and Rousseau](#page-16-2)*, 1984; *[Goldstein et al.](#page-18-0)*, 1984; *[Hawkesworth](#page-19-3) [and Kemp](#page-19-3)*, 2006], but has a much smaller error than previous estimates.

 The calculated model age depends heavily on the chosen parameters for the depleted mantle source. Here, we consider that depletion started at the beginning of Earth history and we chose a very depleted mantle (see Table 2). Other authors prefer more subdued values. For example, Gale et al. [\[2013\]](#page-18-15) recently provided an estimate based on the average composition of MORB while Dhuime et al. [\[2011\]](#page-17-3) suggested that the source of newly formed continental crust was less depleted than the MORB mantle (see Table 2). When these references are used, we calculate that the Nd model age of upper continental crust would be 200 Ma younger. An extra complication has been introduced recently by Jackson and Carlson's [\[2012\]](#page-19-12) suggestion that the Earth has a superchondritic composition. Gale et al. [\[2013\]](#page-18-15) already discussed the issue and showed that a serious problem occurs because the present-day isotopic composition of a superchondritic Earth is identical to their estimate of present-day depleted mantle. Indeed, this leaves no space for the development of a mantle that had become depleted as a consequence of the extraction of enriched continental crust. If we accept that the Earth is superchondritic and that the depleted mantle is more depleted than suggested by Gale et al. [\[2013\]](#page-18-15), with, for example, a Nd isotopic composition similar to our chosen estimate (see Table 2), then the entire mantle would have to be depleted to compensate for the extraction of the continental crust.

5.4 - ¹⁷⁶Lu/177Hf ratio of upper continental crust and Hf model age

 To establish the average Hf model age of the upper continental crust is not as 370 straightforward as for Nd because the $176 \text{Lu}/177$ Hf ratio of continental crust is poorly constrained, yet has a major impact on the calculations. In Table 2, we list 372 estimates of $176 \text{Lu}/177 \text{Hf}_{\text{UCC}}$ that have been suggested in the literature. The 373 ¹⁷⁶Lu/¹⁷⁷Hf values calculated using upper crustal Lu and Hf contents range mostly from 0.0057 to 0.0106 [*[Shaw et al.](#page-20-10)*, 1967; *[Shaw et al.](#page-20-11)*, 1976; *[Taylor and](#page-20-1) [McLennan](#page-20-1)*, 1985; *Condie*[, 1993;](#page-17-0) *[Wedepohl](#page-21-0)*, 1995; *[Gao et al.](#page-18-14)*, 1998; *[Rudnick and](#page-20-0) Gao*[, 2003\]](#page-20-0) but higher values have also been suggested: Goodge and Vervoort [\[2006\]](#page-18-2) suggest a Lu/ 177 Hf ratio of 0.0150 for new Proterozoic crust, while Hawkesworth et al. [\[2010\]](#page-19-0) suggested a value of 0.0159 on the basis of over 7000 measurements of granitoids. Finally, Condie and co-workers [*[Condie et al.](#page-17-4)*, 2011] suggest a value as high as 0.020. The range of suggested values is therefore very large.

382 Unfortunately, the measured $176 \text{Lu}/177$ Hf ratios of the loess samples cannot be 383 used to estimate the $176 \text{Lu} / 177 \text{H}$ of the upper continental crust because loess deposits are affected by sedimentary processes that sort minerals eroded from 385 crustal rocks. In contrast to their generally uniform $147Sm/144Nd$ ratios, all 386 minerals do not have similar $176Lu/177Hf$ ratios, and their relative proportions 387 strongly influence the whole-rock Lu/ 177 Hf ratio, which might be quite different from that of the crustal precursor. One mineral, zircon, is particularly 389 important because it dominates the whole-rock Lu/ 177 Hf ratio with its very 390 high Hf content and its extremely low Lu/ 177 Hf ratio. In addition, it is very resistant to alteration processes and survives even extreme weathering. If a high proportion of old zircon is retained in a loess deposit, then the whole rock has 393 low $176 \text{Lu}/177$ Hf and unradiogenic 176 Hf/ 177 Hf, as is the case with the Western European loess deposits (see Table 1 and Figure 4). In contrast, if the proportion of zircon is lower, as is the case for the Tajikistan loess samples, then the 396 Lu/ 177 Hf is higher and 176 Hf/ 177 Hf is more radiogenic. Thus, the large range of Lu/ 177 Hf ratios reported in loess deposits is likely controlled by sedimentary sorting processes and these values cannot be considered as representative of the Lu/ 177 Hf ratio of the crustal precursors. We therefore use a different

 approach that combines the constraints provided by the Sm-Nd and Lu-Hf isotopic systems to determine the best estimate for the upper continental crust Lu/Hf ratio.

 In Figure 5, we plot Hf model ages of individual loess samples as a function of 404 their Nd model age. For each sample, we use the measured $147\text{Sm}/144\text{Nd}$ and ¹⁴³Nd/¹⁴⁴Nd ratios as well as the depleted mantle values shown with an asterisk in Table 2. To calculate the Hf crustal model ages, we used the measured ¹⁷⁶Hf/¹⁷⁷Hf ratios together with a range of ¹⁷⁶Lu/¹⁷⁷Hf ratios as published for upper continental crust, and the depleted mantle values shown with an asterisk 409 in Table 2. Because the range of possible Lu/ 177 Hf ratios in average upper crust (between 0.0057 and 0.0200, see Table 2) is very large, it introduces a very 411 large range of possible Hf model ages $T_{DM}(Hf)$ (shown by vertical lines in Figure 412 5). If the upper crust has a high $176 \text{Lu}/177 \text{H}$ ratio of 0.0200, as suggested by 413 Condie et al. [\[2011\]](#page-17-4), then the T_{DM} (Hf) of all loess (excluding the Argentinian loess) averages 2.47±0.13 Ga, a value that is 600 Ma older than the average Nd 415 model age (1.82 \pm 0.07 Ga); if the crustal 176 Lu/ 177 Hf ratio is 0.0159, as suggested 416 by Hawkesworth et al. [\[2010\]](#page-19-0), then the $T_{DM}(Hf)$ average at 2.05 \pm 0.11 Ga, still over 200 Ma older than the average Nd model age. In contrast, using a low 418 Lu/ 177 Hf ratio at 0.0057 provides much younger Hf model ages (1.45 \pm 0.08 419 Ga). Finally, a $176 \text{Lu}/177$ Hf ratio of 0.0083, as suggested by Rudnick and Gao 420 [\[2003\]](#page-20-0), gives an average Hf model age of 1.57 ± 0.09 Ga.

 Considering the overall geochemical behavior of REE and Zr-Hf during magmatic and sedimentary processes [*[Vervoort and Patchett](#page-20-12)*, 1996; *[Vervoort and Blichert-](#page-20-13) Toft*[, 1999;](#page-20-13) *[Vervoort et al.](#page-21-2)*, 1999; *[Chauvel et al.](#page-17-9)*, 2008; *[Carpentier et al.](#page-17-8)*, 2009; *[Vervoort et al.](#page-20-2)*, 2011], it seems unlikely that the Nd model age of upper continental crust should be older than its Hf model age. The opposite is, in fact, much more likely because most Hf present in crustal materials is sequestrated in zircon, a mineral phase that is extremely robust during alteration and reworking of continental crust.

429 To constrain the $176 \text{Lu}/177$ Hf ratio of the upper continental crust, we assume here 430 that $T_{DM}(Hf)$ and $T_{DM}(Nd)$ should be identical for each individual loess and dust 431 sample and we calculate the necessary $176 \text{Lu} / 177 \text{H}$ ratio for each individual sample. Results of the calculation are shown in Figure 6 where they are also compared to the range of suggested values from literature. On average, we 434 estimate that the Lu/ 177 Hf ratio of upper continental crust is 0.0125 ± 0.0018 (1σ) .

 In summary, the loess data suggest that the average model age of upper 437 continental crust is 1.82 ± 0.07 Ga (1σ) based on Nd isotopes. Assuming that Hf model ages cannot be any younger than Nd model ages, the loess data also 439 provide constraints on the $176 \text{Lu} / 177 \text{H}$ ratio of upper continental crust, which 440 cannot be smaller than 0.00125 ± 0.0018 , the value that corresponds to 441 concordant $T_{DM}(Hf)$ and $T_{DM}(Nd)$ ages. Higher values, such as those suggested previously by other authors [*[Goodge and Vervoort](#page-18-2)*, 2006; *[Hawkesworth et al.](#page-19-0)*, 443 [2010\]](#page-19-0) are also plausible, but would result in an average upper crust $T_{DM}(Hf)$ age 444 over 200 Ma older than the $T_{DM}(Nd)$ age.

6 - Conclusions

 Our Nd-Hf isotopic study of a large number of loess samples provides new constraints on the average isotopic composition of the upper continental crust. We estimate that present-day upper continental crust has the following 450 characteristics: 143 Nd/ 144 Nd= 0.512101±60 and 176 Hf/ 177 Hf=0.282412±60. Using the bulk silicate Earth values determined by Bouvier et al. [\[2008\]](#page-17-10), these isotopic 452 ratios translate into $\varepsilon_{Nd} = -10.3 \pm 1.2$ ($\pm 1\sigma$) and $\varepsilon_{Hf} = -13.2 \pm 2$ ($\pm 1\sigma$) and fall within error on the terrestrial array of Vervoort et al. [\[2011\]](#page-20-2).

454 The uniform $147\text{Sm}/144\text{Nd}$ ratio (0.1193 \pm 0.0026) of loess deposits can also be considered as representative of the upper continental crust value. Combining the parent/daughter ratio with the average Nd isotopic composition, we calculate 457 that the average Nd model age of upper continental crust is 1.82 ± 0.07 Ga.

458 Finally, we constrain the $176 \text{Lu}/177$ Hf ratio of the upper continental crust using the relationship between Hf and Nd isotopes. Our preferred estimate corresponds to 0.0125±0.0018, a value that is significantly higher than most previous estimates but lower than the ratio suggested by Goodge and Vervoort [\[2006\]](#page-18-2) and Hawkesworth et al. [\[2010\]](#page-19-0). We emphasize that 0.0125 represents the

463 lower limit for $176 \text{Lu}/177$ Hf because any lower estimate makes the Hf model ages younger than the Nd model ages, a very unlikely situation given the geochemical 465 behavior of Hf and REE. The impact of this new $176 \text{Lu}/177\text{H}$ ratio on crustal differentiation ages of zircon populations such as those calculated by numerous authors [e.g., *[Griffin et al.](#page-18-1)*, 2002; *[Harrison et al.](#page-19-2)*, 2005; *[Kemp et al.](#page-19-6)*, 2007; *[Blichert-](#page-17-1) [toft and Albarède](#page-17-1)*, 2008; *[Wang et al.](#page-21-7)*, 2009; *[Belousova et al.](#page-17-2)*, 2010; *[Kemp et al.](#page-19-4)*, [2010;](#page-19-4) *[Yao et al.](#page-21-3)*, 2011; *[Iizuka et al.](#page-19-5)*, 2013] is quite variable, but the ages can be younger by up to 500 Ma in the most extreme cases.

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Table captions:

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- **Table 1**: Nd and Hf isotopic results for all loess samples.
- 728 Footnotes: All ε_{Nd} and ε_{Hf} values were calculated using the BSE value suggested

by Bouvier et al. [\[2008\]](#page-17-10). Dup stands for complete duplicate analyses. The

730 $147\text{Sm}/144\text{Nd}$ and $176\text{Lu}/177\text{Hf}$ are calculated using the Sm, Nd, Lu and Hf

- concentration data listed in the Supplementary table.
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 Table 2: Compilation of the parent-daughter ratios and Nd and Hf isotopic compositions of various reservoirs.

 Footnotes: Numbers in italics correspond to ratios recalculated using published 736 concentrations. $*$: values used to calculate T_{DM} . Data sources: Allègre and Rousseau [\[1984\]](#page-16-2), Bouvier et al. [\[2008\]](#page-17-10), Chauvel et al. [\[2008\]](#page-17-9), Condie [\[1993\]](#page-17-0), Condie et al. [\[2011\]](#page-17-4), Dhuime et al. [\[2011\]](#page-17-3), Gale et al. [\[2013\]](#page-18-15), Gao et al. [\[1998\]](#page-18-14), Goldstein et al. [\[1984\]](#page-18-0), Goodge and Vervoort [\[2006\]](#page-18-2), Griffin et al. [\[2002\]](#page-18-1), Hawkesworth et al. [\[2010\]](#page-19-0), Jahn and Condie [\[1995\]](#page-19-11), McCulloch and Wasserburg [\[1978\]](#page-19-1), Rudnick and Gao [\[2003\]](#page-20-0), Salters and Stracke [\[2004\]](#page-20-14), Shaw et al. [\[1967;](#page-20-10) [1976\]](#page-20-11), Taylor and McLennan [\[1985\]](#page-20-1) and Wedepohl [\[1995\]](#page-21-0).

 Supplementary table: Trace element concentrations measured on all loess samples as well as values published by Gallet et al. [\[1996\]](#page-18-5) for the Luochuan loess.

 Supplementary figure: Comparison between the new trace element data and the data published previously by Jahn et al. [\[2001\]](#page-19-7).

Figure captions:

 Figure 1: Map of the world showing the location of loess deposits. The precise locations of Western European, Chinese, Argentinian and Tajikistan loess samples were provided by Gallet et al. [\[1996;](#page-18-5) [1998\]](#page-18-3), Jahn et al. [\[2001\]](#page-19-7) and Ding et al. [\[2002\]](#page-18-6). The Sahara samples blown over Southern Europe were collected close to Grenoble and at 2900 m altitude in the Alps. Latitudes and longitudes are given in the Supplementary Table.

 Figure 2: Trace element patterns of the loess samples. All concentrations are normalized to the upper continental crust average of Rudnick and Gao [\[2003\]](#page-20-0). a) Western European loess, b) all Chinese loess including data published by Gallet et al. [\[1996\]](#page-18-5), c) Tajikistan loess and d) Argentinian loess and Sahara dust.

Figure 3: Nd and Hf isotopic compositions of loess samples. ε_{Nd} and ε_{Hf} were calculated using the BSE value of Bouvier et al. [\[2008\]](#page-17-10). The terrestrial array 764 corresponds to ε_{Hf} = 1.55. ε_{Nd} +1.21 as recently compiled by Vervoort et al. [\[2011\]](#page-20-2). Data for MORB and OIB were compiled using the PetDB and Georoc databases and also include some unpublished values of Chauvel. Oceanic sediments plotted here were published in the literature [*[White and Dupré](#page-21-8)*, 1986; *[BenOthman et al.](#page-17-11)*, [1989;](#page-17-11) *[McLennan et al.](#page-19-13)*, 1990; *[Godfrey et al.](#page-18-16)*, 1997; *[Albarède et al.](#page-16-4)*, 1998; *[Pearce et](#page-19-14) al.*[, 1999;](#page-19-14) *[Vervoort et al.](#page-21-2)*, 1999; *[David et al.](#page-17-12)*, 2001; *[Woodhead et al.](#page-21-9)*, 2001; *[Vlastelic](#page-21-10) et al.*[, 2005;](#page-21-10) *[Prytulak et al.](#page-20-15)*, 2006; *[van de Flierdt et al.](#page-20-16)*, 2007; *[Carpentier et al.](#page-17-13)*, [2008;](#page-17-13) *[Bayon et al.](#page-16-5)*, 2009; *[Carpentier et al.](#page-17-8)*, 2009; *[Chauvel et al.](#page-17-14)*, 2009; *[Vervoort et](#page-20-2) al.*[, 2011\]](#page-20-2). Data source for continental crust were published by Vervoort and Patchett [\[1996\]](#page-20-12) and by Vervoort et al. [\[1999;](#page-21-2) [2000\]](#page-21-1). The inset shows how our estimated upper continental crust composition (red star) relates to the range of known isotopic values in the Earth.

777 **Figure 4**: Nd/Hf versus $\Delta \varepsilon_{\text{Hf}}$ for all the analyzed loess samples. The inset shows 778 schematically how the $\Delta \varepsilon$ _{Hf} values are calculated for each sample. The terrestrial 779 array used to calculate $\Delta \epsilon_H f$ values corresponds to $\epsilon_H f = 1.55 \epsilon_{Nd} + 1.21$ as recently reevaluated by Vervoort et al. [\[2011\]](#page-20-2). Shown by arrows along the horizontal axis are various estimates for Upper Continental Crust: Taylor and McLennan [\[1985\]](#page-20-1), Shaw et al. [\[1967;](#page-20-10) [1976\]](#page-20-11), Rudnick and Gao [\[2003\]](#page-20-0), Condie [\[1993\]](#page-17-0) and Gao et al. [\[1998\]](#page-18-14).

786 **Figure 5**: T_{DM} (Nd) versus T_{DM} (Hf) for all loess samples.

787 The T_{DM} (Nd) are calculated using the $147Sm/144Nd$ ratio measured for each sample. They are well constrained with an error smaller than the horizontal bars. 789 The T_{DM} (Hf) ages are calculated using a single stage Lu/Hf evolution for the samples coming from China, Tajikistan, Argentina and Sahara, and using a two stages evolution for the samples from Western Europe. For the latter, we assumed that the average crystallization age of the excess zircons was 0.9 Ga, an

 age consistent with the Archean to Proterozoic materials that have been repeatedly reworked during the Cadomian and Hercynian orogenies [*[Peucat et](#page-19-15) al.*[, 1988\]](#page-19-15). For all samples, the vertical bar corresponds to the range of ages 796 obtained with a $176 \text{Lu}/177$ Hf ratio going from 0.0057 (the youngest ages) to 0.0200 (the oldest ages). The inset shows how the two-stages calculation operates. A "zircon stage" is calculated between present-day and a crystallization 799 age T_{cryst} of 900 Ma, and using a $176 \text{Lu}/177 \text{H}$ ratio of 0.0001. Prior to T_{cryst} , the 800 evolution follows the same range of UCC $176 \text{Lu}/177 \text{H}$ ratios as for all other samples. The inset also highlights how single stage crustal model ages become significantly too old if the zircons did not crystallize recently.

804 **Figure 6**: Measured ¹⁴⁷Sm/¹⁴⁴Nd versus calculated ¹⁷⁶Lu/¹⁷⁷Hf for all loess samples.

 As in Figure 5, a single stage history is assumed for the Argentinian, Chinese, Tajikistan and Sahara samples and a two-stages history for the Western European samples. The grey field corresponds to the range of published values for the Lu/Hf ratio of upper continental crust [*[Shaw et al.](#page-20-10)*, 1967; *[Taylor and](#page-20-1) [McLennan](#page-20-1)*, 1985; *Condie*[, 1993;](#page-17-0) *[Gao et al.](#page-18-14)*, 1998; *[Griffin et al.](#page-18-1)*, 2002; *[Rudnick and](#page-20-0) Gao*[, 2003;](#page-20-0) *[Goodge and Vervoort](#page-18-2)*, 2006; *[Hawkesworth et al.](#page-19-0)*, 2010; *[Condie et al.](#page-17-4)*, [2011\]](#page-17-4). The two red lines show our estimated averages and the orange fields 813 correspond to the calculated error at 1σ .

Highlights

- \triangleright First estimate of the average Nd-Hf isotopic composition of upper continental crust
- The average (ε_{Nd} = -10.3±1.2 and ε_{Hf} = -13.2±2) falls on the terrestrial array
- The average Nd model age of upper continental crust is 1.82±0.07 Ga
- Fine upper continental crust 176 Lu/ 177 Hf ratio is 0.0125±0.0018

Figure 6

Table 1: Nd and Hf isotopic compositions of the loess samples

Table2: Compilation of the parent-daughter ratios and Nd and Hf isotopic compositions of various reservoirs

Supplementary table [Click here to download Supplementary material for on-line publication only: supplementary file revised.xlsx](http://ees.elsevier.com/epsl/download.aspx?id=592901&guid=a409ceb2-e927-4d2b-9aac-bc5e1f9a2c91&scheme=1) **Supplementary figure [Click here to download Supplementary material for on-line publication only: supplementary figure.pdf](http://ees.elsevier.com/epsl/download.aspx?id=592902&guid=be07e6c2-3d80-4b4d-a270-42118941a849&scheme=1)**