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The effects of terrestrial weathering on samarium-neodymium isotopic composition of ordinary chondrites

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Abstract:

Following their fall to Earth, meteorites experience weathering. In this systematic study, we evaluate the trace element composition of ordinary chondrites from the Antarctic cold desert, $e^{\lambda t}$, Atacama (Chile) and Lut (Iran) hot deserts, with an emphasis on rare earth elements (REE). Our data confirms that terrestrial weathering of meteorites in hot deserts changes their trace element (Sr, Ba, REE, Hf, Th, and U) concentrations. However, weathering effects in majority of Antarctic samples are limited to slight Ba, REE, Hf, and Th depletions and in some case to U enrichment. In comparison to the Antarctic meteorites, hot desert samples show greater disturbances and REE fractionation relative to the average fall values. We measured the Sm-Nd isotopic composition of the hot desert meteorites that have heavily iv, CNRS, IRD, Coll. France, INRAE, CEFEGE

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affected REE compositions. Our Sm-Nd isotopic data show a significant effect of terrestrial weathering evidenced by non-CHUR 147 Sm/ 144 Nd and 143 Nd/ 144 Nd ratios. Measurements show a higher variation and lower values of 147 Sm/ 144 Nd for the Atacama samples than those from the Lut Desert. Deviations from CHUR 147 Sm $/144$ Nd value are in positive accordance with the degree of La/Lu fractionation caused by weathering. The ɛNd values of Atacama and Lut deserts meteorites range from -2.20 to +1.61, which is wider than the -1.07 to +0.64 range for falls. We suggest that disturbance of primary Sm/Nd ratios resulting from mixing with terrestrial components originating from soil during weathering is responsible for lower 147 Sm/ 144 Nd ratio in these meteorites. The majority of the Atacama meteorites regardless of their weathering degrees have their REE compositions and ɛNd affected by terrestrial contamination. Both 147 Sm/ 144 N $\overline{1}$ ra io and ϵ Nd values show no straightforward relationship with weathering degree. However, in both cases the samples with the highest negative isotopic disturbances are H chondrites from the Atacama and Lut deserts. In addition, Ba concentration shows a negative correlation with 147 Sm/ 144 Nd ratio. Care must be taket into account while dealing with samples collected from hot deserts, even fres. -looking ones. in these meteorites. The majority of the λ acar

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trial contamination. Both $^{147}Sm^{144}N1$ ra io and

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Keywords: Sm-Nd, Samarium-Neodymium, Chondrite, Weathering, Hot desert, Contamination

1. INTRODUCTION

The systematics of rare earth elements (REE) and the Sm-Nd isotopic chronometer are essential in petrogenetic and radiometric studies of terrestrial and extraterrestrial rocks (e.g., DePaolo, 1988). As a result of their slightly different nuclear and chemical properties, REE respond to common petrological processes, such as partial melting and partial evaporation, by developing fractionated light-REE (from La to Sm) or heavy-REE (from Eu to Lu) elemental patterns (e.g., Hanson, 1980; Davis and Richter, 2014). Radioactive decay of 147 Sm ($t_{1/2}$ = 106 Gyr) to 143 Nd is an important tracer for chemical differentiation processes affecting the REE during planetary evolution. In addition, the Sm-Nd isotopic system is one of the most precise and useful dating methods in geology. These properties make REE and Sm-Nd isotopes powerful tools to study the petrogenesis and origin of different magmatic rocks.

Except for returned samples from a few objects such as the Moon and one asteroid 25143 Itokawa and comet Wild 2, excluding small objects such a interplanetary dust particles, meteorites are the only samples available from other solar system bodies. Meteorites allow studying the formation, evolution, and structure of the solar system. Ongoing developments in analytical chemistry, improved elemental and isotopic measurements, and accessibility of more meteoritic material has led us to have a comprehensive though still incomplete vision about the early stages of solar system evolution. These data enable us to see relationships between different meteorite groups, their formation ages, petrogenesis, etc.

The vast majority (~98%) of meteorites available for study are collected in hot and cold deserts. This is particularly the case for some rare nete orite types such as Martian and Lunar meteorites, angrites, etc. These meted rites collected in deserts are referred to as finds, and unlike observed falls, have had relatively long residence times on Earth. Their terrestrial ages range from the sof thousands years (kyr) for most hot deserts finds (Jull et al., 2013) to hundreds of kyr for Antarctica and Atacama finds (Drouard et al., 2019; Welt: n et al., 1997). The exposure of meteorites to terrestrial environments during this residence time alters their mineralogy, chemistry, and isotopic properties (e.g., Bland et al., 2006). Meteorite weathering is a complex process controlled by di' example factors including terrestrial residence time, climate, soil composition at the recovery site, meteorite type, size, and shape (e.g., Pourkhorsandi et al., 2017a, Honnann et al., 2018). As observed in samples collected from different regions of the λ acama desert, meteorite weathering can be variable at sub-regional scale (Munay to et al., 2013). Understanding meteorite weathering processes is critical to ϵ void any data misinterpretation while working on meteorite finds. Terrestrial weathering of extraterrestrial materials is not only limited to "old" finds, but can occur during the short time span between meteorite falls and their recovery (Bischoff et al., 2011; Walker et al., 2018), and even during laboratory storage of samples returned by space missions (Velbel, 2014). All of this evidence points to a potential "threat" from terrestrial weathering on the integrity of cosmochemical data obtained from finds and show the importance of a detailed documentation of meteorite weathering processes. ity (~98%) of meteorites available for study are
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Most of the studies of meteorite weathering have dealt with mineralogy and major element geochemistry (Bland et al., 2006; Golden et al., 1995; Gooding, 1982; King et al., 2020; Velbel et al., 1991). A few studies also have focused on particular trace

elements such as Ba, Sr, and REE (Al-Kathiri et al., 2005; Crozaz et al., 2003; Pourkhorsandi et al., 2017a; Shimizu et al., 1983; Stelzner et al., 1999; Zurfluh et al., 2011). The number of such studies is smaller when it comes to isotopic investigations. However, paucity of information on this topic does not correlate with its importance. In their work on the observed CM chondrite fall Sutter's Mill, Walker et al. (2018) showed disturbances in ¹⁸⁷Re-¹⁸⁷Os and Re/Os ratios in the fragments which had experienced only one post-fall rain event. Observing these effects on a fall meteorite emphasizes the importance of this matter. Isotopic changes caused by meteorite weathering are also reported in H (Stephant et al., 2018), O (Stelzner and Heide, 1996), noble gases (Cartwright et al., 2010; Kaneoko, 1983; Schultz et al., 2005), Sr (Borg et al., 2016, 2003; Brandon et al., 2004; Ela do et al., 2014; Shih et al., 2007), Hf (Sokol et al., 2008; Tatsumoto et al., 1981), Os (Borg et al., 2003; Brandon et al., 2012), Fe (Saunier et al., 2010), U- and Th- series as well as Cs (Weber et al., 2017), and Pb (Tatsumoto et al., $1\sqrt{3}$, isotopes.

The terrestrial weathering effects on meteorites' Sm-Nd isotopic system, the focus of this work, are contradictory. For example, (B randon et al., 2004; Debaille et al., 2007; Elardo et al., 2014; Haloda et ε ¹, 2009) report effects of hot desert weathering in their measurements on Martian meteorites to be insignificant. Meanwhile, most of the studies such as (Borg et al., 2018; Edmunson et al., 2005; Shih et al., 2007) and most of the studies on desert methorites do not report any Sm-Nd isotopic disturbances in the weathered meteorites with already disturbed Rb-Sr systematics. ble gases (Cartwright et al., 2010; Kaneo...> 19

t al., 2016, 2003; Brandon et al., 2004; Ela do κ ol et al., 2008; Tatsumoto et al., 1231), Os (Bo

12), Fe (Saunier et al., 2010), U- a.·d Th- serie

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Despite their importance in cosmochemical studies, REE and especially the Sm-Nd isotopic system have been rarely studied in a systematic manner to track meteorite weathering e^{t} ects on their composition. In this study we evaluate REE composition of ordinary chondrites from Antarctica, Atacama and Lut hot deserts. We also analyzed the Sm-Nd isotopic systematics of meteorites with heavily affected REE compositions.

Ordinary chondrites are the most abundant meteorite types among falls and finds. Meteorites with different weathering degrees, as determined by their mineralogy, were analyzed in order to pinpoint the relationships between REE composition in meteorites with the region where they were found and weathering degree. The second goal of this work is to investigate the effects of weathering on Sm-Nd isotopic composition of meteorites from hot deserts.

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2. SAMPLES AND ANALYSES

2.1. Studied meteorites

Table 1 shows the list of the ordinary chondrites used in this study. Hot desert and Antarctic samples are from the CEREGE (Aix-en-Provence, France) and Royal Belgium Institute of Natural Sciences (Brussels, Belgium) collections, respectively. Ferromagnesian silicates (olivine and pyroxene), (Fe,Ni) metal, and troilite (FeS) are the main primary components of this type of meteorites (Weisberg et al., 2006). During terrestrial weathering they alter to Fe oxyhydroxides (e.g., Pourkhorsandi et al., 2019). To study the possible effects of the meteorite find region on the trace elements and Sm-Nd isotopic composition, meteorites from the Asuka dense collection area (DCA) in Antarctica (Zekollari et al., 2019), the Atacama (Chile) (Drouard et al., 2019; Hutzler et al., 2016; Gattacceca et al., 2011) and Lut (Iran) (Pourkhorsandi et al., 2019) hot deserts were selected. In order to observe possible effects of (Fe, Ni) metal abundance and mineral chemistry on REE budgets during weathering, we chose members from $b \in \mathbb{N}$ H and L groups, as well as LL for Antarctic samples. These three are the main groups of ordinary chondrites, which are distinguished by having different (F_{ϵ}, N_i) metal and total Fe contents. H chondrites have higher (Fe, Ni) metal and total F a than members of L (low total Fe) and LL groups (low metal and total Fc contents) (Wasson and Kallemeyn, 1988). To avoid analyzing possible isotopically unequilibrated exotic grains, most of the analyzed meteorites were selected $\frac{1}{2}$, $\frac{1}{2}$ thermally equilibrated petrologic types (4 to 6). Finally, meteorites with different weathering degrees were selected. Weathering scales differ between A itarctic and other meteorites. For the Antarctic meteorites, the Meteorite Working Group at NASA and NIPR use weathering categories "A," "B," and "C" to denote minor, moderate, and severe rustiness at a macroscopic scale. For hot desert weathering, the scale is based on microscopic observations: W2 denotes moderate oxidation of metal (20-60% being affected), W3 for heavy oxidation of metal and troilite (60-95% being replaced), and W4 for complete (>95%) oxidation of metal and troilite (Wlotzka, 1993). Weathering they after to Fe oxynyaroxides (e.g
dy the possible effects of the meteorite final, age-Nd isotopic composition, meteorites from the LCA) in Antarctica (Zekollari et al., 2019), the Al
019; Hutzler et al., 201

Table 1. Studied meteorites listed based on their find locations.

1 (Ruzicka et al., 2014)**; ²** (Ruzicka et al., 2015a) **; ³** (Ruzicka et al., 2015b) **; ⁴** (Ruzicka et al., 2017) **;** ⁵ (Bouvier et al., 2017a) **; ⁶** (Bouvier et al., 2017b) **; ⁷** (Garvie, 2012) **;**

2.2. Analytical work

Using an optical petrographic microscope, meteorite fragments without visible attached or imbedded terrestrial grains, cracks, and desert varnish were chosen. Between 500 to 1000 mg of the fragments were crushed using an agate pestle and mortar, specifically used for chondritic samples.

All the elemental and isotopic analyses were carried out at the Laboratoire G-Time of the Université Libre de Bruxelles, Belgium. The analytical methodology used in this study is similar to that used by (Armytage et al., 2018; Wainwright et al., 2019).

For trace element analyses approximately 200 mg of powdered samples were dissolved using concentrated acid $(HF+HNO₃$ followed by HCl) in sealed Savillex beakers on a hot plate at 120 °C for four days with evaporation and re-dissolution between the two acid steps. Elementals analyses were conducted using an Agilent 7700 Quadrupole-Inductively Coupled Plasma-Mass Spectrometer (Q-ICP-MS) using a He collision cell. Samples were introduced with 5% $HNO₃$, and In was used as an internal standard. Standards BHVO-2 and BCR-2 were used as international reference material. Table 2 shows a comparison of the elemental concentrations of these standards obtained during this work with those \mathbf{r} from the literature.

Table 2: Whole-rock concentrations of trace e'ement (in µg/g) in the analyzed standards compared to the literature values

 1 (Wilson, 1997a); 2 (Jochum et al., 2016); 3 (Wilson, 1997b)

Based on our trace element data and generally higher interaction of the hot desert meteorites with their environment compared to meteorites from Antarctica (Koeberl and Cassidy, 1991), only hot desert samples were chosen for Sm-Nd isotopic measurements. About 500 mg of each sample were weighed and digested using a mixture of concentrated $HNO₃$ -HF followed by HCI (similar to the method described above). Neodymium and Sm were purified from the matrix using a two-column ionexchange technique as detailed in (Armytage et al., 2018). After digestion a 5% aliquot was taken for isotope dilution analysis of Nd, and Sm with a ¹⁵⁰Sm-¹⁴⁸Nd mixed spike. For both spiked and unspiked samples, first, REE were extracted from the sample using ∼2 ml of AG50W-X8 cation resin (200–100 mesh). Then HDEHP (di-2ethylhexyl-orthophosphoric acid)-coated Teflon purifying (di-2ethylhexyl-orthophosphoric acid)-coated Teflon purifying Nd from Sm and Ce, both of which cause isobaric interferences. The unspiked Nd cuts were measured on the Nu-Plasma II Multiple Collector – Inductively Coupled Plasma -Mass Spectrometer (MC-ICP-MS) of ULB, in dry plasma with an Aridus II desolvator. For unspiked Nd each analysis consists of 3 blocks of 20 runs each. The repeated measurements of Rennes international standard gave an internal reproducibility of 13 ppm (2σ, n=20). Total external reproducibility based on full duplicates is better than 33 ppm. Net dymium isotopic compositions were corrected by internal normalization to the value of $146Nd^{144}Nd=0.7219$, and by sample-standard bracketing using the recommended value of the Rennes standard (¹⁴³Nd/¹⁴⁴Nd=0.511961; Chauvel and Blichert-Toft, 2001). Total blank was negligible (8 pg) and reproducibility was better than 99% as measured over seven replicate analyses. The spiked aliquots were measured on the Nu Plasma I MC-ICP-MS at ULB. For analysis of spiked Sm and Nd the mass fractionation factors for each element were derived by solving iteratively non-linear equations combining the exponential fractionation law and the spike natural mixing equations as in (Debaille et al., 2007). For isotope dilution analysis of Nd, and Sm with
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3. RESULTS

3.1. Trace elements

Whole-rock trace element composition of the analyzed meteorites are reported in Table 3. The CI-normalized spider diagrams of Antarctica and hot desert samples

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(Atacama and Lut) along with those of average composition of different groups of ordinary chondrites (falls) are shown in Fig. 1. For EM 091, EM 120, and Kerman 026 average values of two cuts are presented.

Table 3: Whole-rock concentrations of trace element (in μ g/g) in the studied chondrites from the Antarctica, and Atacama and Lut hot deserts. Average CI and ordinary chondrites abundances are from (Barrat et al., 2012) and (Wasson and Kallemeyn, 1988), respectively.

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¹EM: El Médano; ²A: Asuka; ΣREE is sum of RE \overline{F} in m La to Lu in ppm. Normalized ratios are showing the ratio elements normalized to CI-chondrites.

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Fig. 1: CI-normalized trace element composition of ordinary chondrites collected from a) Asuka dense collection area in Antarctic and b) Atacama and Lut hot deserts. Average CI and ordinary chondrites abundances are from (Barrat et al., 2012) and (Wasson and Kallemeyn, 1988), respectively.

Analyzed samples from Antarctica show relatively unfractionated REE patterns (average La_N/Lu_N ratio is 1.15 \pm 0.18) (Fig. 1a), almost mimicking those of the average fall compositions. However, most Antarctic samples show slightly lower REE concentrations than average falls. Average Σ REE for ordinary chondrites falls is 3.54 (Wasson and Kallemeyn, 1988), while our Antarctic samples' average ƩREE is 2.71 (n=11). Strontium, Ba, Hf, and in some cases Th show very similar behavior to the REE in the Antarctic meteorites. These variations do not show a pronounced relative fractionation and as such mostly can be explained by differences in the equilibrium states of different chondrites, and the wide range in ordinary chondrite chemical compositions. For example, Asuka 09436 is a H3 chondrite and shows the lowest Σ REE content (Fig 1a). The highly mobile U contents are similar to the falls values, except Asuka 09618 with a high weathering degree (\therefore) that shows an elevated U concentration. Barium shows a slight depletion in Antarctic samples with a generally lower Ba/La ratio of these meteorites relative to \mathfrak{u} , \sim e of mean falls.

In comparison to the Antarctic meteorites in it desert samples show larger disturbance and REE fractionation relative to the average fall values. Σ REE contents in the hot desert meteorites are lowe. tr_A n the average fall values, ranging from 1.09 to 2.42 in H chondrites from Atacama. For L chondrites, Atacama samples range from 0.65 to 0.87 and the average for three Lut samples is 0.56, both lower than the Atacama H chondrites. Maximum and average $(La/Lu)_N$ ratio in the Atacama H chondrites is 5.15 and 4.27, respectively. Average ratios for Atacama L and Lut (regardless of their types) chondrites are 2.13 and 1.21 (respectively) and higher than the aforementioned Artarctic meteorites. r example, Asuka 09436 is a H3 chondrit, and
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All the hot desert samples (except Kerman 009 and Kerman 013 from Lut, and EM 091 from Atacama) are depleted in Sr, while in contrast, Ba is showing a remarkable uniform positive anomaly of greater than 1. This anomaly reaches up to \sim \times 39 higher than average CI chondrites in El Médano (EM) 091. Hafnium and Th contents in all hot desert samples are depleted compared to the average falls. However, the key difference is higher ratios of Th/Hf and $(Lu/Hf)_N$ in comparison to the falls. As shown in the case of EM 091, $(Lu/Hf)_N$ can vary from 0.70 (close to mean fall values) to 1.71 in two different cuts from the same powder of a meteorite. Similar to Antarctic meteorites, hot desert samples display a positive U anomaly, though with a somewhat greater magnitude. Except for two Atacama L chondrites (EM 89, 90), U concentrations in the analyzed hot desert samples are higher than the values

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reported for the average falls. The maximum measured content is for Kerman 009 which has \sim \times 8 times higher U than average CI chondrites.

3.2.Samarium-Nd systematics

The results of the Sm-Nd isotopic measurements for the analyzed hot desert chondrites are reported in Table 4. As shown in Fig. 2, 147 Sm/ 144 Nd ratios in the hot desert meteorites exhibit significant deviations from the chondritic isochron and the values reported for ordinary chondrite falls. The 147 Sm/ 144 Nd ratios measured in hot desert chondrites range from 0.0777 to 0.1973, most often lower than the average of 0.1958 ± 0.0012 (n=22) reported for falls (Bouvier et al., 2003; Patchett et al., 2004). The average 147 Sm/ 144 Nd for the Atacama and Lut samples is 0.1433 ± 0.0287 (n=10) and 0.1876 \pm 0.0080 (n=7), respectively. This indicates \sim higher variation and lower values of 147 Sm/ 144 Nd for the Atacama samples than those from the Lut Desert. The average 147 Sm/ 144 Nd ratios of the H and L chondrites from the Atacama are 0.1345 ± 0.0287 (n=7) and 0.1638 \pm 0.0147 (n=3). For \angle 0: Desert H and L chondrites the ratios are 0.1790 ± 0.0037 (n=3) and 0.1940 ± 0.0022 (n=4), respectively. In both cases, H chondrites show higher deviations from the falls than their L counterparts. range from 0.0777 to 0.1973, most often lower

(n=22) reported for falls (Bouvier et al., 2002; P

m/¹⁴⁴Nd for the Atacama and Lut samples is 0.

980 (n=7), respectively. This indicates in higher

4Nd for the Atacama sa

Table 4: Sm-Nd isotopic compositions of chondrite whole rocks. Elemental concentrations unit is µg/g

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¹Caleta el Cobre; ²El Médano

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In addition to variation between different meteorites, differences in ¹⁴⁷Sm/¹⁴⁴Nd also occur within a given meteorite as evidenced by different values measured for duplicates which have been taken from the same powder. For instance, the standard deviation between two ¹⁴⁷Sm/¹⁴⁴Nd measurements of Kerman 026 is 0.0013, while for EM 091 and EM 120 it is 0.0254 (n=2) and 0.0046 (n=2), respectively. It should be noted that Kerman 026 was found in the Lut Desert, while the two latter are from the Atacama Desert. Note that EM 091, the sample with the highest standard deviation, is visually more weathered than the other two samples.

The measured 143 Nd/ 144 Nd ratios are expressed as ε Nd values, where ε Nd is calculated using the equation shown below:

 ϵ Nd = $[(143Nd)^{144}Nd)$ sample/ $(143Nd)^{144}Nd)$ CHUR]-1 \times 10⁴

where CHUR is the chondritic uniform reservoir with a composition of 143 Nd/ 144 Nd = 0.51263 (Bouvier et al., 2008). Figure 3 depicts the ˃4 ɛNd-unit variation measured for desert meteorites The ɛNd values range from -2.20 to 1.61, which is greater than the -1.07 to 0.64 range for falls (Bouvier et al., 2008). The average ɛNd value for samples from the Atacama and Lut are -1.05 and -0.58, respectively.

Fig. 3: ¹⁴⁷Sm^{/144}Nd versus ϵN of the analyzed hot deserts compared to the fall ordinary chondrites. El Médano (EM) and Caleta el Cobre (CeC) are from the Atacama Desert. Kerman and Shahdad are from the Lut Desert (underlined). Data for fall ordinary chondrites are from (Bouvier et al., 2008).

4. DISCUSSION

Our data confirms that terrestrial weathering of meteorites changes their trace element (Sr, Ba, REE, Hf, Th, and U) concentrations. In addition, Sm-Nd isotopic measurements of ordinary chondrites from the Atacama and Lut hot deserts show significant effects of terrestrial weathering as manifested by their non-CHUR 147 Sm/ 144 Nd and 143 Nd/ 144 Nd ratios.

4.1. A comparison of the trace element compositions of chondrites from hot and cold deserts

Thousands of years of exposure to a terrestrial environment leads to chemical contamination of meteorites (Al-Kathiri et al., 2005; Hezel et al., 2011; Pourkhorsandi et al., 2017a). Interaction with liquid water containing different elements originating from the soil below the meteorite and the atmosphere, and chemical leaching by these liquids, plus physical and biological weathering are common in hot desert environments. All these weathering processes lead to the modified chemical patterns (Fig. 1b). Also evident in our data, are anomalies in Sr, Ba, REE, Hf, Th and U contents, which are well-known indicators of hot desert weathering (Al-Kathiri et al., 2005; Barrat et al., 2003; Folco et al., 2007; Hezel et al., 2011; Hofmann et al., 2018; Moore and Brown, 1963; Pourkhorsandi et al., 2017a; Saunier et al., 2010; Stelzner et al., 1999; Zeng et al., 2018; Zurfluh et al., 2012). In comparison, Antarctic meteorites show less chemical modification than those from the hot deserts (Fig. 1).

Starting with Sr and Ba, our data shows the chemical differences between different hot deserts in addition to differences to the Antarctic samples. Meteorites from different find locations have different trends $r \beta$ and Sr contents (Fig. 4). Relative to the average fall OCs, those from Atacama are significantly enriched in Ba and depleted in Sr, while two Lut samples $\alpha \in \mathbb{R}$ highly enriched in Sr. In comparison, Antarctic samples show values slightly lower than the average fall values. These differences can be explained by the compositional differences of the meteorite residence sites and different solid compositions. For instance, in the Lut Desert, terrestrial fluids containing Sr that originated from carbonate-rich soils infiltrate the meteorites through cracks, which ultimately leads to an increase in the Sr budget of the rock. This scenario is consistent with higher carbonate contents in Lut Desert meteorites than those from other hot deserts as reported in (Pourkhorsandi et al., 2019). Similar to Lut meteorites, high Sr contents in samples recovered from carbonate-rich soils of deserts in Oman are also in accordance with such a scenario (Al-Khatiri et al., 2005). Gibson and Bogard (1978) reported the occurrence of such uptake of Sr within as few as 19 years after meteorite fall. Enriched Ba patterns in the Atacama meteorites can be explained by higher values of Ba in these soils (360-954 ppm) reported by (Pourkhorsandi et al., 2017a) compared to the soil samples collected from the Lut Desert (332-562 ppm) reported by (Pourkhorsandi et al., 2019). In some cases, possible small-scale soil compositional heterogeneities might result in weathered meteorites with relatively different chemical compositions than the 1, 1963; Pourkhorsandi et al., 2017a; Sau, ier e

et al., 2018; Zurfluh et al., 2012). In comparisc

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majority of recovered meteorites from that region. For instance, this might be the case for Kerman 026, which regardless of being from the Lut Desert, has lower Sr values than mean falls. Contrary to the hot desert meteorites, the meteorite finds from Asuka DCA have not been in contact with soil. Our analyzed samples from the Asuka were residing on the ice at the time of their recovery. Relative to hot desert meteorites, terrestrial weathering has not been sufficiently intense to alter their chemical compositions significantly. The active weathering processes in such an environment would be abrasion by the wind and leaching of the meteorite during its limited interaction with liquid water below and on the ice surface (Harvey, 2003; Koeberl and Cassidy, 1991). For this reason, meteorite weathering in Antarctica for the majority of the elements would cause depletions rather than enrichments (for instance in Ba, Hf, Th, and some cases in REE as seen in Fig. 1a). Oxidation of chondritic components such as troilite by liquid water forms acidic environment which itself facilitates alteration (leaching) process of o_n or minerals such as Ca-phosphate which is the main host for these elements in a meteorite (Ebihara and Honda, 1983; Jones et al., 2014; Martin et al., 2013). However, there are some exceptions such as U which can be enriched compared to falls. Uranium enrichment in both falls, and Antarctic finds has been also reported from meteorites collected from other regions of the continent, with the effects of \log sources, such as water sprays from the Southern Ocean, or local rock exposures proposed as being responsible (e.g., Koeberl and Cassidy, 1991). For this reason, meteorite wether
the majority of the elements would cause depletions ratt er t nan
instance in Ba, Hf, Th, and some cases in REE as seen in Fig. 1
chondritic components such as

Terrestrial weathering has decreased the REE contents in hot desert meteorites and probably to some use the Antarctic meteorites. However, it has not significantly affected the ratios of \mathbf{h} ase elements in Antarctic samples, suggesting possibly insignificant re-adjustment of Sm-Nd isotopic system. This makes the majority of samples (or at least ordinary chondrites) collected from Asuka reliable for cosmochemical studies. Relatively significant weathering effects occur in highly altered meteorites such as in Asuka 09436, which has a weathering degree C and shows the lowest REE content. However, in the other samples the chemical composition does not show any relationship with weathering degree. Roughly 61 % of Antarctic basaltic achondrites (especially eucrites) exhibit a cerium anomaly (Kagi and Takahashi, 1998; Mittlefehldt and Lindstrom, 1991), and it has been suggested to be a fingerprint of chemical removal of REE from these meteorites. However, we

do not observe it in our samples, which suggests it is either limited to achondrites or it points to the presence of multiple weathering regimes in the Antarctica.

Unlike in the Antarctic finds, hot desert weathering appears to modify both REE abundances and ratios, likely due to concomitant action of two main processes. One is contamination by terrestrial fluids coming from soil resulting in increased light-REE/heavy-REE ratios (represented by higher La_N/Lu_N ratios), and second leaching of chondritic components such as Ca-phosphates causing a decrease in bulk REE budget (Table 3) (Al-Kathiri et al., 2005; Pourkhorsandi et al., 2017a). As suggested by (Pourkhorsandi et al., 2017a), the longer average terrestrial residence times of the Atacama meteorites are responsible for their more fractionated REE patterns in comparison to those from the Lut Desert. However, their higher abundances of ΣREE can be explained by the lower weathering rate and more limited leaching than those from the Lut Desert (Pourkhorsandi et al., 2019). (\overline{B} and et al., 2006) suggest a faster weathering rate for H than L and LL OCs, due to their higher (Fe,Ni) metal contents leading to a higher abundance of Fe oxy-hydroxides which can absorb REE from the fluids and soil and hence show high $R^r\bar{L}$ contamination at the microscale (Crozaz et al., 2003; Crozaz and Wadhwa, 2001). 'Higher REE fractionation in these meteorites can be explained by this process. However, comparison of the weathering degrees with REE composition shows a cor iplex pattern. The meteorite EM 120 with a weathering degree W2 shows the second highest fractionation (La*N*/Lu*N* = 5.07) after EM 091 (H5 W3, La*N*/Lu*N* = 5.15), while the strongly weathered (W4) EM 111 is the least fractionated H chongrite from the Atacama. If et al., 2017a), the longer average terrestrial in
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Similar to REE, Ht and Th have been affected by terrestrial weathering. This is the case for some Asuka samples and for all hot desert meteorites. As mentioned earlier, these two elements partly are fixed in Ca-phosphate structure and can get released by alteration of this mineral during weathering. Thorium is not only found in Caphosphates as Kuznetsov (2011) reported 5% of thorium in water-soluble components and 9.1% in troilite, both of which potentially can be easily affected by weathering. Uranium enrichment is the other weathering proxy that has been observed in hot desert meteorites which is suggested to form as a result of contamination by soil fragments and interaction with fluids originating from the soil (e.g., Hezel et al., 2011). As a consequence of these processes, Lu/Hf and Th/U ratios in our analyzed meteorites, in particular in hot desert samples have been

modified. Main results of such modification would be disturbances in Lu-Hf and U-Th-Pb isotopic systems which also have been reported by Bast et al. (2017) and Weber et al. (2017).

In conclusion, we see that the Antarctic meteorites are chemically less affected by terrestrial weathering than those from the hot deserts. Soil composition controls Sr and Ba contents of hot desert meteorites. Rare Earth element ratios in Antarctic samples are close to the average falls. By contrast, in the hot desert samples REE are strongly affected by weathering, as evidenced by both lower absolute values and higher relative fractionations. The maximum fractionation is observed in the H chondrites from the Atacama. Meteorite weathering degrees do not show straightforward relationships with their REE compositions. Higher contamination of Atacama meteorites could be also related to their relatively higher terrestrial ages than those from other regions (Drouard et al., 2019 , 1 ogether, this evidence makes hot desert meteorites, and particularly those from the Atacama, suspect samples for cosmochemical studies, in contrast to the greater reliability of Antarctic meteorites for such studies. cuonations. The maximum iractionation is obset
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4.2. Modification of Sm-Nd isotopic systematics during terrestrial weathering

The previous section showed that the transstrial weathering can fractionate elemental parent/daughter ratios, and therencie suggests that special care must be taken into account when working with Rb -Sr, U-Th-Pb, and even Lu-Hf isotopic systems particularly in hot desert meteorites. Such fractionation will create deviations from "true" isochrons. Studies utilizing Rb-Sr, U-Pb, or Lu-Hf chronometers such as (Bast et al., 2017; Borg et al., 2016, 2003; Debaille et al., 2007; Dreibus et al., 2001; Elardo et al., 2014; Shih et al., 2007; Sokol et al., 2008; Tatsumoto et al., 1981) have all reported such cases. The disruptive effects of terrestrial weathering on Rb-Sr, Lu-Hf, and U-Th-Pb isotopic system are to be expected from the Sr and U positive anomalies and modified Lu/Hf and Th/U ratios (Fig. 1). However, in the case of Sm-Nd isotopic system, it is complicated as little is known about their respective behaviors during terrestrial weathering.

In addition to our data regarding modification of elemental abundances of REE, isotopic measurements can offer supplementary evidence of modification of Sm-Nd isotopes during terrestrial weathering. This is a crucial issue as hot desert meteorites,

in particular Saharan achondrites, are amongst the main objects used for cosmochemical studies.

In their Sm-Nd study on the Golpara ureilite, Torigoye-Kita et al. (1995) suggested that terrestrial contamination, mainly concentrated in the acid leachates, was responsible for its anomalous composition. Jagoutz et al., (2001) suggested a large shift in the Sm/Nd ratio caused by contamination to be responsible for a hot desert martian meteorite's "anomalous" isotopic composition. In addition, several other works such as Debaille et al. (2007), Dreibus et al. (2001), and Elardo et al. (2014) have yielded data that partly correlated with these findings. Outside of these studies, the majority of works on hot desert meteorites using Sm-Nd isotopic system suggest very low probability of disturbance due to terrestrial weathering (Debaille et al., 2007).

Most of the data available on modifications of Sn. Na isotopic system during weathering deal with achondrites. However, as weathering mainly involves interaction of fluids with primary components thorough cracks, this would apply to ordinary chondrites as well. In fact, higher abundance \sim (Fe,Ni) metal and troilite in ordinary chondrites would offer a higher alteration capacity and higher chemical modification for ordinary chondrites.

As shown in the previous section in Table 4 and Fig. 5, Sm/Nd ratios in our analyzed hot desert samples are lower than the average falls. Disturbance of this ratio is responsible for lower 147 Sm/ 144 Nd ratio which is more evident in the Atacama samples as they have more fractionated REE patterns. Lower Sm/Nd ratios show the effects of mixing with a teri estrial component (e.g., Torigoye-Kita et al., 1995). The majority of Atacama meteorites regardless of their weathering degrees (at least $W\geq 2$) mave yielded data that partity correlated with these informings. Outs
the majority of works on hot desert meteorites using Sm-N-4 ichters
very low probability of disturbance due to terrestrial weath.org (
Most of the data are indeed contaminated.

Fig. 5: CI-normalized abundances of Sm and Nd in whole-rock Antarctic, Atacama Desert, and Lut Desert ordinary chondrites. Average ordinary chondrite and CI chondrite values from (Vasson and Kallemeyn, 1988) and (Barrat et al., 2012).

Comparison of the 147 Sm/ 144 Nd ratio with meteorite types shows higher deviations from the average value fall for H chondrites (Fig. 5). As in the case of elemental abundances, the same can be applied for isotopic compositions. The ¹⁴⁷Sm/¹⁴⁴Nd ratio does not show a straightforward relationship with weathering degree. It gets even more complicated as we see highly heterogeneous duplicates for two single meteorites (El Médano 091 and 120), while we do not observe this in two duplicates of Kerman 026 collected from the Lut Desert.

As mentioned earlier, La_N/Lu_N ratio is shown to increase during terrestrial weathering of meteorites. Plotting this ratio together with ¹⁴⁷Sm/¹⁴⁴Nd of relatively highly modified H chondrites (Fig. 6) shows a negative correlation with ¹⁴⁷Sm/¹⁴⁴Nd ratio. This correlation shows that both of these ratios are being controlled by similar processes, mainly mixing with terrestrial components. As REE elemental fractionation in meteorites proceeds towards a soil like composition during desert weathering, the ¹⁴⁷Sm/¹⁴⁴Nd ratios follow similar route towards terrestrial compositions.

Fig. 6: ¹⁴⁷Sm^{$/144$}Nd versus La, $^{\prime}$ Lu_M of the analyzed hot deserts compared to the fall ordinary chondrites. Samples from Lut are underlined. Dashed curve shows linear trend pattern of H finds. Such Jard deviation for 147 Sm^{$/144$}Nd of H falls in 0.0012 $(n=10)$. Average elemental and isotopic ordinary chondrite values are from (Wasson and Kallemeyn, 1988) ϵ nd (Bouvier et al., 2008), respectively.

A comparison between Ba concentrations, shown to get enriched during weathering, with ¹⁴⁷Sm/¹⁴⁴Nd ratios, reveals a negative correlation. As shown in Fig. 7, H chondrites with higher Ba/Nb values (higher chemical alteration), have lower 147 Sm $/144$ Nd and show higher deviations from the CHUR value. As mentioned in the previous sections, Ba contents in these meteorites are higher in the samples collected from Ba-rich soils (Atacama) than those from the Lut Desert.

Fig. 7: ¹⁴⁷Sm^{/144}Nd versus Ba/Nb of the analyzed hot deserts compared to the fall ordinary chondrites. Samples from Lut are underlined. Dashed curve shows linear trend pattern of H finds. Standard deviation for 147 Sm^{$/144$}Nd of H falls in 0.0012 $(n=10)$. Average elemental and isotoric ordinary chondrite values are from (Wasson and Kallemeyn, 1988) and (Bouvier et al., 2008), respectively. $\frac{1}{147}$ Sm/¹⁴⁴N.

Id versus Ba/Nb of the analyzed hc. deserts co

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In addition to the Sm/Nd $\varepsilon \overline{v}$ $\frac{147}{2}$ Sm/¹⁴⁴Nd ratios, $\frac{143}{2}$ Nd/¹⁴⁴Nd ratio is also highly disturbed (Fig.3). Except Kerman 026, which as discussed earlier also shows different trace element in periavior, all the other disturbed samples are showing negative ε Nd values. Similar to the 147 Sm/ 144 Nd ratio, ε Nd values do not show a straightforward relationship with the weathering degrees. However, as in the case of the previous discussions, the samples with the highest negative ɛNd values are H chondrites from the Atacama and Lut deserts.

More studies such as laser ablation ICP-MS in-situ measurements of trace elements will be needed to pinpoint the main factors controlling contamination of trace element compositions and Sm-Nd isotopic composition of hot desert meteorites. Such studies should target different primary components, weathering cracks and chemical modifications around them, and possible terrestrial components inside weathered meteorites. Our data clearly shows that care must be taken into account while dealing with samples collected from hot deserts, even the fresh-looking

24 **Nd Desert _ Pourkhorsandi et al.** Nd Desert _ Pourkhorsandi et al.

ones, and including the Atacama Desert which is shown to be an extraordinary region hosting a high population of meteorites (Gattacceca et al., 2011), and in particular unique samples (e.g., Pourkhorsandi et al., 2017b).

5. CONCLUSIONS

25 Nd Desert Pourkhorsandi et al. Terrestrial weathering modifies trace element composition of ordinary chondrites, in particular REE. The degree of chemical modification in the Antarctic meteorites collected from the Asuka DCA is lower than that for hot desert meteorites. Antarctic meteorites show REE patterns close or only slightly below \mathbf{u} at of average falls, but without any noticeable fractionation. This also applies mostly for Sr, Hf, and Th. Uranium contents in Antarctic samples are close to f_{ξ} ^{'I}s value with some enrichments possibly caused by weathering. Barium concentration in Antarctic samples is lower than falls. Meteorite leaching (especially Ca-phos ν^L ates) can explain this depletion. Hot desert meteorites show Ba, LREE, and *U* positive anomalies which are representative of terrestrial weathering. Cuncentrations of REE in the hot desert meteorites is lower than average falls $2r$, d also they are highly fractionated. This fractionation is higher in the Atacana samples, especially H chondrites, than those from the Lut Desert. This is in ag $e^{i\theta}$ ent with their average residence on Earth: 700 kyr for Atacama meteorites versus 12 kyr for other hot desert meteorites (Drouard et al., 2019). We suggest this modification is the result of concomitant action of two main processes. One is contamination by terrestrial fluids coming from soil resulting in increased light-REE, heavy-REE ratios (represented by higher $\text{La}_{\text{N}}/\text{Lu}_{\text{N}}$ ratios), and second leaching of chondritic components such as Ca-phosphates causing a decrease in bulk REE budget. Terrestrial weathering lowers Sm/Nd ratios which in consequence disturbs 147 Sm/ 144 Nd composition. This ratio has a negative correlation with La_N/Lu_N ratio. Barium content of the weathered desert meteorites show a negative correlation with 147 Sm/ 144 Nd contents. This makes La_N/Lu_N ratio and Ba concentration proxies to check possible modification of 147 Sm $/144$ Nd ratio. Hot desert weathering changes ¹⁴³Nd/¹⁴⁴Nd composition of the meteorites as well, which is more evident in the H chondrites. This systematic work shows disturbance of Sm-Nd isotopic composition of meteorites during terrestrial weathering. Disturbance of Lu/Hf and Th/U ratio also point to possible changes in Lu-Hf and U-Th-Pb isotopic systems of these meteorites. Great caution should be paid when dealing with hot desert REE patterns close or only slightly below μ at cable fractionation. This also applies mostly for in Antarctic samples are close to f_{ϵ} l's value way weathering. Barium concentration, in Antarctite leaching (especial

meteorites, even when they look fresh. More studies, especially in-situ measurements, should be conducted to pinpoint the main factors controlling REE and Sm-Nd isotopic compositions during terrestrial weathering of meteorites.

Declaration of competing interest

The authors declare than they have no known competing interests or personal relationships than could have appeared to influence the work reported in this paper.

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Declaration of interests

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The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

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