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1 The Sn isotope composition of chondrites: implications for volatile element 2 depletion in the Solar System

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9 **Abstract**

10 The origin of volatile element depletion in terrestrial planets and meteorites relative to a solar
11 composition represented by CI carbonaceous chondrites remains an unsolved problem. The
12 isotope compositions of moderately volatile elements may offer the possibility to distinguish
13 between the various processes that may have caused this depletion (e.g., partial condensation
14 or partial evaporation). We report high precision Sn isotope measurements in carbonaceous
15 chondrites and ordinary chondrites and the results are reported as $\delta^{124/116}\text{Sn}$. Four
16 carbonaceous chondrites (Orgueil CI, Murchison CM2, NWA 5240 CV3 and Allende CV3) show a
17 limited range in $\delta^{124/116}\text{Sn}$ (-0.02‰ to 0.11‰) with an average value of $0.04 \pm 0.11\%$ (2 s.d.) for
18 a wide range of Sn concentrations (0.63 ppm to 1.57 ppm). The absence of Sn isotope
19 fractionation among carbonaceous chondrites suggests that volatile depletion may have taken
20 place under thermodynamic equilibrium conditions between solid and vapor in the Solar
21 Nebula. Alternatively, the mixing of two components, a volatile-free component containing no
22 Sn and a volatile-rich component could explain this trend. This latter hypothesis is consistent

23 with the overall trace element pattern found in carbonaceous chondrites, showing a constant
24 relative abundance when normalized to CI chondrites for the most volatile elements. In
25 contrast with carbonaceous chondrites, ordinary chondrites exhibit a larger range of Sn isotope
26 compositions ($\delta^{124/116}\text{Sn}$ from -2.02‰ to 0.64‰), but neither the degree of metamorphism (3-6)
27 nor the group (H, L, LL) is correlated with Sn isotopic variations, or with the Sn contents (range
28 0.20 to 1.44 ppm). Nineteen out of twenty-one ordinary chondrites are enriched in light Sn
29 isotopes compared with carbonaceous chondrites and the bulk silicate Earth. The trace element
30 patterns of volatile elements in ordinary chondrites suggest that equilibrated ordinary
31 chondrites have been disturbed by parent body processes related to metamorphic or shock
32 overprinting but also inherited isotope fractionation found in unequilibrated ordinary
33 chondrites. Last, the isotope composition of the bulk silicate Earth (BSE) indicates that the
34 volatile element depletion observed in the Earth took place in conditions perhaps similar to
35 those of carbonaceous chondrites, as a simple model describing the effect of Earth's core
36 formation on Sn isotopes shows that the Sn isotope composition of the bulk Earth is identical to
37 that of the BSE and of carbonaceous chondrites.

38

39 **1. Introduction**

40 The elemental abundances in CI carbonaceous chondrites (excluding H, C, N, noble gases, Li, B,
41 and partly Be) are similar to the composition of the Sun. In contrast to CI chondrites, other
42 carbonaceous and ordinary chondrites, as well as terrestrial planets show depletions in
43 moderately volatile elements (e.g., Halliday and Porcelli, 2001). These elemental depletions are
44 roughly correlated with volatility as expressed by their condensation temperatures (Wood et al.

45 2019). Several processes in the early history of the Solar System have been proposed to explain
46 these observations: (i) preexisting volatile element depletion in dust relative to vapor in the
47 Interstellar Medium (Palme and Hutchison, 2001; Yin, 2005), (ii) partial condensation from a hot
48 Solar Nebula (Larimer, 1973; Wasson and Chou, 1974; Grossman and Larimer, 1974; Wai and
49 Wasson, 1977; Cassen, 1996; Cassen, 2001; Albarède, 2009), (iii) partial evaporation during
50 thermal events or due to the migration of volatile-rich material into hotter regions (Alexander,
51 2005; Ciesla, 2008), (iv) mixing of components with variable volatile element depletion resulting
52 from previous partial vaporization or condensation processes (e.g., Anders, 1964; Larimer and
53 Anders, 1967; Larimer, 1973; Braukmüller et al., 2018; Alexander et al., 2001; Alexander, 2019a),
54 (v) volatile loss during impacts or planetary magmatism (Anders et al., 1976; Larimer and
55 Anders, 1967; Wolf et al., 1980; Hin et al. 2017), (vi) volatile loss during metamorphism (Dodd,
56 1969; Wasson, 1972; Schaefer and Fegley, 2010). For planetesimals and planets, the
57 incorporation of elements into the core may complicate this picture as many volatile elements
58 are also siderophile or chalcophile. Clearly, the ‘volatility trend’ showing a decrease in
59 concentrations in the bulk silicate Earth as a function of condensation temperatures (e.g.,
60 Palme and O’Neill, 2013) is better constrained for lithophile elements that are not affected by
61 core formation.

62 It should be mentioned here that process (i) has been dismissed by Palme (2001) as it would
63 introduce heterogeneities in the $^{87}\text{Sr}/^{86}\text{Sr}$ due to differences in Rb/Sr ratios and these
64 differences have not been found (Palme, 2001). Thus, the preferred hypothesis is that the
65 moderately volatile element depletion is directly linked to early Solar System processes.

66 In principle, the isotope analysis of moderately volatile elements in meteorites and terrestrial
67 samples could help distinguish between the processes of partial condensation and partial

68 evaporation. Partial condensation should lead via kinetic effects to light isotope enrichment in
69 the solid, while partial evaporation produces a solid residue enriched in heavy isotopes
70 (Humayun and Cassen, 2000; Davis and Richter, 2013; Bourdon and Fitoussi, 2020). The
71 maximum isotope fractionation should take place in the case of free evaporation in vacuum.
72 However, the isotope fractionation is often limited when evaporation does not take place in
73 vacuum, while elemental depletion is still observed (Young, 2000; Ozawa and Nagahara, 2001;
74 Yu et al., 2003; Davis and Richter, 2013). For example, Humayun and Clayton (1995) and Wang
75 and Jacobsen (2016) had reported no K isotope fractionation in chondrites, achondrites and in
76 the Earth, although K has a clear elemental depletion in terrestrial planets, chondrites and
77 achondrites compared to CI chondrites. A more recent study by Bloom et al. (2020) found
78 variations of K isotopes among chondrites with no clear correlation between K isotope
79 variations and K concentrations. More specifically, evaporation experiments indicated that if
80 the system is in chemical equilibrium, then there is a back-reaction (i.e., condensation) that
81 reduces the kinetic isotope effect of evaporation (e.g., Wombacher et al., 2008; Davis and
82 Richter, 2013). This corresponds to conditions of relatively high partial pressure of the species
83 of interest and in this case the solid can isotopically re-equilibrate with the ambient gas, leading
84 to limited isotope fractionation. Therefore, the elemental fractionation without K isotope
85 fractionation was interpreted as the result of a nearly complete thermodynamic equilibrium
86 caused by the relatively high K partial pressure in the ambient gas (Alexander et al., 2000;
87 Alexander and Grossman, 2005; Fedkin et al., 2006). Altogether, this shows that the isotope
88 fractionation of volatile elements in meteorites and planetary materials can also be a measure
89 of whether a condensed phase was in thermodynamic equilibrium with the vapor phase (Davis
90 and Richter, 2013) .

91 The measurements of Rb isotopes in chondrites and terrestrial samples reveal small resolvable
92 variations with a total range of a few per mil units (Nebel et al., 2011; Pringle and Moynier,
93 2017). Notably, there is a positive correlation of $\delta^{87}\text{Rb}$ with Rb/Sr ratios in carbonaceous
94 chondrites that has been interpreted as a mixture between a volatile-rich component and a
95 volatile-depleted component (Pringle and Moynier, 2017). This interpretation still does not
96 explain the origin of ^{87}Rb -depleted signature in Rb-depleted chondrites. Similarly, the absence
97 of a $\delta^{87}\text{Rb}$ difference between Earth and chondrites has been interpreted as reflecting a mixture
98 between 90% of a putative volatile free component (Rb \sim 0 ppm) and 10% of a CI chondrite
99 component (Nebel et al., 2011; Pringle and Moynier, 2017). However, no specific mineral
100 phase or mineral assemblage has been shown to represent this volatile-free component. High
101 precision isotopic measurements of Cd (T_{50} = 652 K by Lodders, 2003, and 502 K by Wood et al.,
102 2019) show that most carbonaceous chondrites including CI, CV, CM, CO and CR have nearly
103 identical Cd isotope compositions (Baker et al., 2010; Wombacher et al., 2008, 2003). In
104 contrast with K and Cd isotopes, Zn isotopes are characterized by decreasing $\delta^{66/64}\text{Zn}$ values
105 with Zn depletion in carbonaceous chondrites, from CI (average $\delta^{66/64}\text{Zn}_{\text{CI}} = 0.46 \pm 0.08\text{‰}$, 2 s.d.)
106 to CK/CR chondrite ($\delta^{66/64}\text{Zn} = -0.69 \pm 0.03\text{‰}$, 2 s.d.) (Barrat, 2012; Albarède, 2009; Luck et al.,
107 2005; Pringle et al. 2017). This observation was initially interpreted to indicate partial
108 condensation in the early Solar System (Albarède, 2009). Pringle et al. (2017) have attributed
109 this effect to mixing of components involving a volatile-rich component (matrix) and a volatile-
110 poor component that could possibly be chondrules. In addition, the $\delta^{66/64}\text{Zn}$ value of BSE has
111 been estimated to be $0.28 \pm 0.05\text{‰}$ (2 s.d.) (Chen et al., 2013; Doucet et al., 2016) or $0.16 \pm 0.06\text{‰}$
112 (Sossi et al., 2018). These values could suggest a role for partial condensation in the solids that
113 accreted to ultimately form the Earth. Similarly, $\delta^{65}\text{Cu}$ and $\delta^{128/126}\text{Te}$ for carbonaceous
114 chondrites decreases with increasing volatile element depletion (Luck et al., 2003; Hellmann et

115 al., 2020). Other studies of volatile elements such as B, Se and Tl have revealed no resolvable
116 variations in planetary materials (Baker et al., 2010; Vollstaedt et al., 2016) and the conclusions
117 of these studies was that elemental depletion was not necessarily coupled with isotope
118 fractionation. In the case of Se, a more recent study (Labidi et al., 2018) has shown variations in
119 Se isotopes among chondrite groups (specifically the EH chondrites) and this was interpreted as
120 a result of kinetic fractionation during sulfidation. In addition, the $\delta^{82/78}\text{Se}$ of carbonaceous
121 chondrites is correlated negatively with the inverse of Se concentrations (Labidi et al., 2018),
122 similarly to what has been found for Zn and Cu. Overall, based on these studies, there is no
123 simple interpretation regarding the origin of the elemental depletions in meteorites, and while
124 several processes such as free evaporation can be ruled out, the origin of the depletion remains
125 an open question (e.g., Davis, 2006).

126 Tin is a moderately volatile element with a 50% nebular condensation temperature of 704 K
127 (Lodders, 2003) and recalculated to be 604 K by Wood et al. (2019), which means it would now
128 belong to the category of highly volatile elements. It is depleted in the bulk silicate Earth
129 relative to CI chondrites, by a factor of 33 ± 3 (Jochum et al., 1993). It has ten stable isotopes and
130 is also a potential tracer for investigating the origin of volatile element depletions in planetary
131 materials and the nature of Earth's building blocks. Since Sn is moderately siderophile, its
132 concentration could be affected by core formation, in addition to volatility-driven processes
133 such as condensation and evaporation. The distribution of Sn among different meteorites is
134 poorly known and few direct concentration measurements are available in the literature (e.g.,
135 Shima, 1964; Hamaguchi et al., 1969; De Laeter et al., 1974; Braukmüller et al., 2018; Creech
136 and Moynier, 2019). The analysis of the metallic fraction of ordinary chondrites (Shima, 1964)
137 has shown that Sn is enriched in the metal phase by a factor of up to 15, but this enrichment

138 could be disturbed by thermal metamorphism. Metal-silicate partition coefficients (D) also
139 indicate that Sn is siderophile with a strong dependence on oxygen fugacity, with a D
140 decreasing with increasing pressure (Capobianco et al., 1999; Righter and Drake, 2000; Righter
141 et al., 2010; Righter et al., 2018). Tin can also become chalcophile in the absence of metallic
142 iron. For example, in terrestrial peridotites, Sn is enriched in the minor sulphide phase (Witt-
143 Eickschen et al., 2009). Although Sn isotope compositions of meteorites were analyzed in early
144 studies (e.g., De Laeter and Jeffery, 1967, 1965; Devillers et al., 1983; Loss et al., 1990;
145 McNaughton and Rosman, 1991; Rosman et al., 1984; Rosman and Mcnaughton, 1987), isotope
146 fractionation was not detected outside of the analytical precision (2 s.d. > 0.4‰ using the
147 $\delta^{124/116}\text{Sn}$ notation). Recent studies of Sn isotopes in terrestrial rocks (Badullovich et al., 2017;
148 Wang et al., 2018) have been used to determine the Sn isotope composition of the bulk silicate
149 Earth. Creech and Moynier (2019) have analyzed a series of chondrites with both ordinary
150 chondrites and enstatite chondrites exhibiting lower $\delta^{124/116}\text{Sn}$ values than that of the bulk
151 silicate Earth determined by Badullovich et al. (2017). The observed variations were attributed
152 to Sn volatility. These authors concluded that if the Earth was built from enstatite chondrites,
153 this initial Sn isotope signature was removed from the bulk silicate Earth either by volatilization
154 or metal separation. Here, we apply a newly developed MC-ICP-MS technique (Wang et al.,
155 2017) to analyze the Sn isotope composition of carbonaceous and ordinary chondrites to
156 reexamine these questions.

157

158 2. Samples and analytical methods

159 A wide variety of chondrites were analyzed in this study as shown in Table 1. Four
160 carbonaceous chondrites, Orgueil (CI), Murchison (CM2), NWA 5240 (CV3) and Allende (CV3)
161 were analyzed for Sn concentrations and Sn isotope compositions. Twenty-one ordinary
162 chondrites with various degrees of metamorphism (Type 3 to 6) belonging to the three groups
163 (H, L, LL) were also analyzed. Among these samples, ten of them belong to the Type 3
164 chondrites and thus represent unequilibrated ordinary chondrites (UOC) that should have
165 better preserved nebula features, if any.

166 Diamond disks using for cutting samples often contain bronze that could cause severe Sn
167 contamination. Therefore, to avoid surface contamination, the samples were first crushed into
168 small pieces in an agate mortar and only the internal pieces were carefully selected under
169 optical microscope. The chosen pieces were washed with Milli-Q water in a Teflon beaker. The
170 beaker was then immersed in an ultrasonic bath for 10 min to remove any powder that could
171 originate from the surficial parts of the original sample. The selected pieces were dried under a
172 laminar flow hood. They were then ground to a fine powder in an agate mortar (for the amount
173 of powdered material, see Table S1, Supplementary Material).

174 The sample dissolution procedure was detailed in our previous study (Wang et al., 2017). Briefly,
175 about 30 mg to 300 mg powdered sample (Table 1) was mixed with ^{117}Sn - ^{122}Sn double spike
176 solution. Then a mixture of 5 mL of concentrated HF and 5 mL of concentrated HNO_3 was added
177 and heated on a hot plate at 160 °C for at least 48 hours and subsequently dried down. Then, 4
178 mL of aqua regia were added to decompose fluorides, after which the solution was dried down
179 and redissolved in 2 mL of concentrated HCl, heated and evaporated twice. Finally, the sample

180 was dissolved in 1 mL of HCl 0.4 N for further processing. For carbonaceous chondrites, at the
181 end of dissolution process, the samples were taken up in 10 mL of ~1% H₂O₂ (this addition does
182 not result in any evaporative loss of Sn as the chemistry yields were ~100%, Wang et al. 2017)
183 and were exposed to UV light for 8 hours to break down organic matter. This treatment
184 efficiently destroys any remaining organic molecules and was shown to stabilize the signal
185 during MC-ICPMS measurements. After the UV treatment, they were evaporated and
186 redissolved in 0.4 M HCl for ion-exchange chromatography.

187
188 The purification of Sn was achieved with a three-step ion-chromatography using anion-
189 exchange resin. The protocol, as described in Wang et al. (2017), can separate Sn from the
190 matrix, the elements producing direct isobaric interferences of Sn (i.e., In, Cd and Te) and the
191 elements potentially forming molecular interferences (i.e., Ge, As, Se, Mo and Ag). The overall
192 yield of the chemical separation and purification of Sn was equal to 100±5% (n = 9). Any
193 possible isotope fractionation produced by ion-chromatography and mass spectrometer was
194 corrected with the ¹¹⁷Sn-¹²²Sn double-spike technique (Wang et al., 2017).

195 Isotopic measurements of Sn were performed on a multi-collector inductively coupled plasma
196 mass spectrometer (MC-ICP-MS) Neptune Plus (Thermo Scientific), equipped with an Aridus II,
197 Jet sampler cone and X skimmer cone, at the Laboratory of Geology at ENS Lyon. The NIST SRM
198 3161a standard solution which was purchased from Chemical products for Analysis (CPA) was
199 used as a primary standard. The samples were bracketed by a mixture of NIST SRM 3161a
200 standard with the double spike solution. An in-house secondary standard, which was made
201 from cassiterite (SnO₂), was measured repeatedly during a measurement session to check
202 measurement accuracy. The preparation of this secondary standard was described in Wang et

203 al. (2017). The intensities of Sn isotopes were corrected by an “on-peak zero” technique for
 204 each sample to correct molecular isobaric interferences formed from Ar₃ (e.g., ³⁶Ar⁴⁰Ar₂ and
 205 ³⁸Ar₂⁴⁰Ar on ¹¹⁶Sn, ³⁸Ar⁴⁰Ar₂ on ¹¹⁸Sn) as it was shown in Wang et al. (2017) that the level of Ar₃
 206 could increase the uncertainties by 30% on the ¹¹⁶Sn/¹²²Sn ratio and a factor of ~8 on the
 207 ¹²⁰Sn/¹¹⁶Sn ratio. Additionally, ¹¹⁶Sn was corrected for the ¹¹⁶Cd isobaric interference by
 208 monitoring ¹¹¹Cd and assuming a natural ¹¹¹Cd/¹¹⁶Cd ratio. The uncorrected δ^{124/116}Sn values of
 209 our samples are lower and mass-independent when comparing with δ^{124/118}Sn (see below and
 210 Table S2, Supplementary Material), indicating the interference of ¹¹⁶Cd on mass 116. The Sn
 211 isotope compositions were calculated using a Matlab® program for Sn double-spike inversion
 212 based on Rudge et al. (2009). The results are reported as δ^{124/116}Sn relative to the NIST SRM
 213 3161a standard:

$$\delta^{124/116}\text{Sn} = \left(\frac{{}^{124}\text{Sn}/{}^{116}\text{Sn}_{\text{sample}}}{{}^{124}\text{Sn}/{}^{116}\text{Sn}_{\text{NIST3161a}}} - 1 \right) \times 1000 \quad (1)$$

215 The details of this method, including operating conditions, spike calibration and data reduction
 216 are described in Wang et al. (2017). The internal reproducibility on the NIST SRM 3161a
 217 standard with a concentration of 10 ppb Sn during a single analytical session is better than
 218 ±0.06‰ (2 s.d.) on δ^{124/116}Sn. Repeated Sn isotope measurements of the USGS standard
 219 reference andesite AGV-1 from multiple digestions, chemical purification and mass
 220 spectrometer measurements yielded a reproducibility of ±0.11‰ on δ^{124/116}Sn (2 s.d., Wang et
 221 al., 2017) during the measurement period of samples reported here. This value is adopted as a
 222 realistic external reproducibility for meteorite samples with better apparent reproducibility (i.e.,
 223 the calculated reproducibility of Allende, based on four independent digestions, is ±0.07‰ but
 224 the uncertainty was still reported as ±0.11‰, Table S2, Supplementary Material). The Sn

225 concentrations of the samples were obtained by an isotope dilution technique using the
226 $^{117}\text{Sn}/^{116}\text{Sn}$ ratio and the precision is better than 4% (RSD).

227 **3. Results**

228 The Sn isotope compositions of twenty-five samples are reported in Table 1 along with Sn
229 contents and plotted in Figure 1. The $\delta^{124/118}\text{Sn}$ values were also calculated using
230 ^{118}Sn , ^{117}Sn , ^{122}Sn and ^{124}Sn for the inversion and are listed in Table 1. Figure 2 shows that the
231 $\delta^{124/118}\text{Sn}$ versus $\delta^{124/116}\text{Sn}$ diagram is characterized by a slope of 0.743 ± 0.040 , which is close to
232 the theoretical slope for equilibrium fractionation (0.737) or the kinetic isotope fractionation
233 (0.744). This indicates that isobaric interferences, mass-independent effects including
234 nucleosynthetic isotope anomalies for Sn are smaller than the analytical uncertainties for these
235 samples. A mass-independent fractionation due to variation in nucleus charge radius (Moynier
236 et al. 2009) would affect ^{117}Sn (and ^{119}Sn), thereby disturbing the double-spike inversion and
237 thus fail even the calibration of double-spike (details see Wang et al. 2017), while
238 nucleosynthetic anomalies would affect differently ^{116}Sn (s-only process), ^{117}Sn ^{118}Sn (s- and r-
239 process), $^{122},^{124}\text{Sn}$ (r-only process) (Beer et al., 1989).

240 The elemental concentrations of Sn for carbonaceous chondrites in this study range from
241 0.63 ± 0.01 ppm to 1.57 ± 0.13 ppm (2 s.e.). Orgueil (CI) retains the highest concentration among
242 them (1.57 ± 0.13 ppm) and is close to the values of 1.64 ± 0.49 ppm measured by De Laeter et al.
243 (1974), 1.5 ± 0.15 ppm by Friedrich et al. (2002), 1.15 ± 0.06 ppm by Fehr et al. (2005), 1.63 ± 0.3
244 ppm by Lodders et al. (2009), and 1.55 ± 0.05 ppm by Braukmüller et al. (2018), as well as the
245 compilation value of 1.68 ± 0.17 ppm given by Ebihara et al. (1982) (Table S3, Supplementary
246 Material). Murchison (CM2) contains 0.99 ± 0.08 ppm Sn, also in agreement with literature

247 values of 1.0 ± 0.1 ppm by De Laeter et al. (1974), 0.89 ± 0.09 ppm by Friedrich et al. (2002),
248 0.72 ± 0.2 ppm by Fehr et al. (2005), 0.96 ± 0.05 ppm by Braukmüller et al. (2018), and 0.81 ± 0.27
249 ppm by Creech and Moynier (2019). The average Sn concentration for Allende (CV3) obtained
250 with four individual dissolution replicates is 0.63 ± 0.01 ppm (2 s.e.), also consistent with the
251 values of 0.69 ± 0.27 ppm given by De Laeter et al. (1974), 0.58 ± 0.12 ppm by Friedrich et al.
252 (2003), 0.55 ± 0.05 ppm by Fehr et al. (2005), 0.68 ± 0.01 ppm by Braukmüller et al. (2018), and
253 0.65 ± 0.02 ppm by Creech and Moynier (2019). Another CV3, NWA 5240 contains 1.04 ± 0.08
254 ppm Sn. Carbonaceous chondrites show a limited range of $\delta^{124/116}\text{Sn}$ values from -0.02 ± 0.11 to
255 $0.11\pm 0.11\text{‰}$ (2 s.d.). According to our analytical uncertainty of $\pm 0.11\text{‰}$, $\delta^{124/116}\text{Sn}$ of
256 carbonaceous chondrites are indistinguishable from each other, despite differences in their Sn
257 concentrations. The four meteorites yield a mean $\delta^{124/116}\text{Sn}$ value for carbonaceous chondrites
258 equal to $0.04\pm 0.11\text{‰}$.

259 As shown in Table 1, the Sn concentrations of ordinary chondrites vary from 0.20 ± 0.03 to
260 1.44 ± 0.12 ppm and are generally lower than for carbonaceous chondrites (0.63 ± 0.01 to
261 1.57 ± 0.13 ppm). Our Sn concentration range in ordinary chondrites is similar to that reported
262 for L and LL chondrites (Friedrich et al., 2003; Friedrich et al., 2004; Creech and Moynier, 2019,
263 Table S3, Supplementary Material), although there are Sn concentrations as high as 4.7 ppm
264 reported for ordinary chondrites in Friedrich et al. (2003). Ordinary chondrites show a large
265 $\delta^{124/116}\text{Sn}$ variation from -2.02 ± 0.11 to $0.64\pm 0.11 \text{‰}$ (2 s.d.). Compared with carbonaceous
266 chondrites, nineteen out of twenty-one ordinary chondrites have lighter Sn isotope
267 composition than those found for carbonaceous chondrites. The remaining two, Forest Vale (H4)
268 and Dhurmsala (LL6) have $\delta^{124/116}\text{Sn}$ values that are heavier than carbonaceous chondrites.

269 Overall, our data set is entirely comparable with the observations reported by Creech and
270 Moynier (2019) for the Sn concentrations. The Sn isotope data is more difficult to compare due
271 to the lack of a common standard. Altogether, Creech and Moynier (2019) reported similar
272 range of variations in $\delta^{124/116}\text{Sn}$ with strongly negative $\delta^{124/116}\text{Sn}$ in ordinary chondrites
273 (from -2.16‰ to 1.09‰, calculated based on the reported $\delta^{122/118}\text{Sn}$ values of the
274 Supplementary Information of Creech and Moynier, 2019) and constant $\delta^{124/116}\text{Sn}$ for
275 carbonaceous chondrites of CM, C2-ung, CO and CV (from 0.61‰ to 1.23‰ with an average of
276 0.86‰) identical within error to their value of the bulk silicate Earth (0.98‰). The offset in
277 $\delta^{124/116}\text{Sn}$ between ordinary chondrites and carbonaceous chondrites is similar to that reported
278 in this study. Creech and Moynier (2019) also measured Sn isotopes in enstatite chondrites and
279 found that their mean $\delta^{124/116}\text{Sn}$ value is 0.3‰ below that of their BSE. This led them to
280 conclude that if one were to build the Earth from enstatite chondrites, it would require an
281 isotope fractionation between metal/sulfide and silicate during core formation.

282 **4. Discussion**

283 **4.1 Tin isotope signature of carbonaceous chondrites**

284 As shown in Table 1 and Figure 1, the Sn isotope compositions of carbonaceous chondrites (CI,
285 CM and CV) are nearly identical, with an average $\delta^{124/116}\text{Sn}$ value of $0.04 \pm 0.11\text{‰}$ (2 s.d.). Thus,
286 despite the difference in Sn concentrations in the various carbonaceous chondrite groups, there
287 is no trend in the $\delta^{124/116}\text{Sn}$ with Sn concentration diagram. These results are consistent with
288 the findings of Creech and Moynier (2019) who also reported constant $\delta^{122/118}\text{Sn}$ in
289 carbonaceous chondrites. This feature is difficult to explain by the effect of terrestrial alteration,
290 especially since one sample is a find from Northwestern Africa that has been exposed to surface

291 waters. The tin concentrations in surface waters was reported below 0.005 $\mu\text{g/L}$ in a recent
292 study (McVay et al., 2019). Therefore, as there is no systematic Sn isotope differences between
293 the ‘falls’ and ‘finds’ of our study, we contend that the Sn isotope signature in chondrites does
294 not reflect the effect of terrestrial weathering. Similarly, the effect of shocks can be dismissed
295 as it would have caused dispersion in the Sn isotopes versus Sn concentration pattern found in
296 carbonaceous chondrites.

297 The effect of aqueous alteration on the parent body of carbonaceous chondrites could in
298 principle have mobilized Sn and modified its isotope composition. One cannot rule out that
299 these effects have taken place, however they do not seem to have modified the $\delta^{124/116}\text{Sn}$
300 values in the bulk meteorites.

301 The absence of Sn isotope fractionation, despite variations in Sn contents relative to the bulk
302 Solar System represented by CI chondrites indicates that the Sn depletion recorded in
303 carbonaceous chondrites can be tentatively attributed to an early process in the Solar System.
304 It could be explained from several scenarios: (i) Sn depletion was produced by volatilization
305 under equilibrium conditions or (ii) mixing of a Sn-free component with an Sn-rich matrix
306 similar to the CI chondrites (e.g., Braukmüller et al., 2018; Alexander, 2019a). For the first
307 scenario, as Sn is a siderophile element, its main budget should reside in the metal phase,
308 except in CI chondrites that have no metallic iron. During formation of chondrules at a
309 temperature above 1500 K, it should have rapidly equilibrated with the gas phase, avoiding
310 kinetic isotope fractionation due to evaporation. The magnitude of equilibrium isotope
311 fractionation between Sn metal and SnS vapor can be estimated using the data from Polyakov
312 et al. (2005) and the vibration frequency of SnS in the gas phase (Douglas et al., 1961) to show
313 that the SnS(gas)-Sn(metal) isotope fractionation at 1500 K is 0.04 ‰, suggesting that under

314 equilibrium conditions the fractionation should be smaller than the observed range in
315 carbonaceous chondrites.

316 The second scenario assumes that the matrix contains all the Sn and is similar to a CI chondrite
317 composition while another component is Sn-free. This scenario can clearly explain the constant
318 $\delta^{124/116}\text{Sn}$ in our carbonaceous chondrites. This scenario can be tested simply by plotting the Sn
319 concentration normalized to Si as a function of the fraction of matrix determined in the model
320 of Alexander (2019a). The results are plotted in Figure 3 and show that there is indeed a
321 positive trend suggesting that the fraction of matrix can account for the Sn budget in these
322 meteorites. Importantly, if one calculates the mixing line between the matrix and the volatile-
323 poor component using the Si concentration in the components given in Alexander (2019a), one
324 can show that the mixing line yields a Y-intersection of about 0.02 (Figure 3a). If one simply
325 uses Sn concentrations (Figure 3b), then the trend shows that the matrix-free component could
326 include a significant fraction of Sn. This is consistent with the observations of Hellmann et al.
327 (2020) who showed that, more generally, there was a non-zero abundance in Sn and several
328 other volatile elements in the chondrule component of carbonaceous chondrites. Thus, we
329 consider based on our data that the second model would seem to agree with our observations.

330 It has been found, however, that chondrules in a carbonaceous chondrite (CR) and an L3.05
331 chondrite did contain Sn at a significant level, suggesting that chondrules are not devoid of
332 volatile elements (Mahan et al., 2018b). However, this may be due to a parent body aqueous
333 alteration. For Allende, Sn-bearing alteration phases have been found in Ca-Al rich inclusions
334 (Ma and Beckett, 2018), indicating Sn mobility towards high temperature components due to
335 aqueous alteration. Thus, it is possible that the Sn concentrations in chondrules may not
336 represent a pristine feature.

337 The nature of the volatile poor component in carbonaceous chondrites was reexamined by
338 Hellmann et al. (2020) who showed that if one uses mass fraction of matrix instead of volume
339 fraction then the moderately volatile element concentrations obtained for the matrix-free
340 endmember, then the concentration of these elements is not equal to zero but that it is
341 remarkably constant when normalized to the CI value. This puzzling information suggests that
342 the chondrule-like fraction is not entirely devoid of these trace elements. If one further
343 assumes that this chondrule-like fraction carries an isotopically fractionated signature, then it
344 can explain the trend between the isotope signature of moderately volatile elements and
345 concentrations (e.g. Luck et al. 2005; Hellmann et al. 2020). Hellmann et al. (2020) further
346 argued that these observations may be reconciled by considering that the chondrule
347 component is itself a mixture of a CI-like component (that dominates the trace element
348 signature) and an isotopically fractionated and more refractory component that would explain
349 the Zn, Cu, K, Te and Rb isotope signatures (Luck et al., 2003; Luck et al., 2005; Pringle et al.,
350 2017; Hellmann et al., 2020; Bloom et al., 2020). In this case, the elements that would be fully
351 condensed would carry no isotope difference with CI chondrites. The isotope fractionation
352 would be seen only in elements that are partly condensed and Te, Zn, Rb, Cu and Cu would fall
353 in this category while Sn, Cd and Tl would be unfractionated.

354 The difficulty in constraining this scenario is that the condensation temperature of elements is
355 only known for canonical conditions (e.g. Wood et al. 2019) but there may be differences or
356 uncertainties owing to variations in the dust/gas ratio during condensation (e.g. Ebel 2000) or
357 uncertainties in the activity coefficients of the elements in the condensates. To illustrate the
358 effect of changing conditions of condensation we have determined the condensation
359 temperatures of the elements of interest for a dust/gas ratio of 1000 (Figure 4) using the
360 activity coefficients given by Wood et al. (2019). This figure illustrates large variations in

361 condensation temperatures in these conditions compared to the conditions of the Solar Nebula.
362 This shows that it is difficult to know precisely the condensation temperatures of trace
363 elements. Only elements that would be partly condensed would potentially have fractionated
364 isotope signatures. As a toy-model, one could produce the observed isotope patterns in
365 carbonaceous chondrites by considering that the 'chondrule component' consists of one
366 component condensed at high temperature (around 1500 K), while the second component
367 would be formed by condensation around 700 K (Figure 4). In the first high temperature
368 component (dashed line a in Figure 4), one would observe fractionated isotope signatures for
369 Cu, Zn, K (no condensation of Sn, Cd) while the second component (dashed line b in Figure 4)
370 with partial condensation would exhibit fractionation in Rb and Te isotopes (and full
371 condensation of Sn, Cd). This qualitative model is obviously ad-hoc and should be considered as
372 merely illustrative. The chondrule component as defined in Hellmann et al. (2020) would then
373 consist of a CI-chondrite mixed with these higher temperature components in proportions that
374 would match the $\sim 1/10$ depletion in moderately elements.
375 Further constraining such a scenario would require more precise determinations of the
376 condensation temperature in various T , P and fO_2 conditions. One would also need to construct
377 an astrophysical scenario that would justify the presence of components formed at various
378 temperature to make it more realistic.

379

380 **4.2 Sn isotope fractionation in ordinary chondrites**

381 The pattern of volatile element abundances in ordinary chondrites has been interpreted by
382 partial condensation from the Solar Nebula (Ikramuddin et al., 1977; Wai and Wasson, 1977;

383 Morgan et al., 1985), by mixing and fractionation of various components (Alexander, 2019b), by
384 metamorphism on the parent body (e.g., Schaefer and Fegley, 2010) or by shock volatility (e.g.,
385 Friedrich et al., 2004). Similarly, there has been competing interpretations of the isotope
386 signature of volatile elements in ordinary chondrites. For example, Zn isotope variations in
387 ordinary chondrites were interpreted as due to condensation of a vapor enriched in light
388 isotopes (Luck et al., 2005), while Wombacher et al. (2008) argued that the Zn-Cd trace element
389 patterns combined with the Cd isotope indicated open-system parent body metamorphism.
390 The disturbed positive trend between Zn versus Sn concentrations (Figure 5) is consistent with
391 the observations made for Cd-Zn and suggests that Sn has also been affected by secondary
392 processes rather than being solely controlled by volatility (in which case it should show a well-
393 defined positive correlation). In contrast, Pringle et al. (2017) interpreted the light Zn isotopic
394 compositions of ordinary chondrites being due to the loss of a Zn-bearing sulphide in the
395 nebula. The stable Te and Ag isotope record of ordinary chondrites has also been interpreted
396 as the result of multiple stages of metamorphism involving both evaporation and condensation
397 (Schönbächler et al., 2008; Fehr et al., 2018). Interestingly, the complex processes that were
398 invoked for explaining the Zn isotope record of ordinary chondrites did not cause any Se
399 isotope fractionation according to the study of (Vollstaedt et al., 2016), while a more recent
400 study by Labidi et al. (2018) suggests a correlation between $\delta^{82/78}\text{Se}$ and Se concentrations in
401 carbonaceous chondrites.

402

403 Overall, the Sn isotope signature observed in ordinary chondrites is similar to that of other
404 moderately volatile elements. First, the Sn concentration in ordinary chondrites (Figure 5)
405 shows a similar pattern to that of Ag (Schönbächler et al., 2008) and Cd (Wombacher et al.,
406 2008) which are both very scattered suggesting the importance of secondary processes. Second,

407 the observation of both enrichment and depletion in light Sn isotopes relative to the CI
408 chondrite composition indicates the interplay of at least two processes (e.g., evaporation +
409 condensation) rather than a single process (e.g., evaporation only). Last, the significant range
410 in $\delta^{124/116}\text{Sn}$ found in unequilibrated ordinary chondrites (-2.02 to -0.19‰) suggests that
411 nebular processes must have played a role in the observed variations. For volatile elements, we
412 do not necessarily expect that their stable isotope record should be homogenized prior to
413 accretion and this is what we observe, which is L and LL chondrites that do not have identical
414 $\delta^{124/116}\text{Sn}$ values in Types 3.0-3.2 samples (Table 1). Furthermore, the mean $\delta^{124/116}\text{Sn}$ value for
415 unequilibrated ordinary chondrites is $-0.71 \pm 0.27\text{‰}$ (2 s.e.), which is resolvably below the mean
416 value obtained for carbonaceous chondrites ($0.040 \pm 0.054\text{‰}$, 2 s.e.), as confirmed by a
417 statistical t-test. This means that ordinary chondrites indicate that a nebular event produced an
418 Sn isotope fractionation relative to the Solar composition. Such an offset could in principle be
419 explained by a kinetic isotope effect during partial condensation (Figure 6), rather than
420 equilibrium fractionation that should be more subdued.

421 It was hypothesized by Larimer (1973) and Wasson and Chou (1974) that the variations in
422 metallic iron abundances found among the ordinary chondrites was due to partial loss of
423 metallic Fe during an early stage of the protoplanetary disk (e.g., Pignatale et al., 2019). Since
424 the budget of siderophile elements is dominated by the metallic phase, the partial separation of
425 metal should deplete these elements in the resulting bulk L and LL chondrites. It turns out that
426 this is the case for the most refractory elements down to Ge (Larimer 1973) whose
427 condensation temperature is 883 K given by (Lodders, 2003) and 830 K by Wood et al. (2019).
428 For the more volatile elements such as Sn, there is no observed difference among the three
429 ordinary chondrite groups, suggesting that elements more volatile than Ge (including Sn)

430 condensed in the remaining metal after this fractionation event. The absence of variations in Sn
431 concentrations for the various types of ordinary chondrites (Figure 7, Figure 8) would indeed
432 suggest that the Sn budget was not controlled by the variable amount of metal present in
433 ordinary chondrites. Therefore, the separation of metal itself cannot explain the Sn isotope
434 fractionation in ordinary chondrites and this isotope fractionation by condensation would have
435 taken place once the metal was separated from silicate.

436 Although ordinary chondrites may carry a nebular signature in Sn isotopes, there is also a clear
437 signature of parent body processes. In the context of metamorphism, Sn is not the most mobile
438 element compared with other volatile elements (Schaefer and Fegley, 2010). The data
439 compilation of Schaefer and Fegley (2010) shows that Sn does not exhibit greater depletion
440 from Type 3 to Type 6 chondrites, although there is clearly a lack of data compared with other
441 elements. The calculated sequence of volatility at 1 bar calculated by Schaefer and Fegley (2010)
442 would place Sn between Se and Tl. Selenium shows similar concentrations for Type 3 to Type 6,
443 while a marked depletion is observed for Tl (Schaefer and Fegley, 2010). A limitation of this
444 calculation is that the 'formation temperature' for volatile elements at 1 bar are 2300, 2302 and
445 2225 K for Sn, Se and Tl, respectively, in ideal solid solutions, which are well above the
446 temperatures experienced by ordinary chondrites during metamorphism. These
447 thermodynamic calculations are, therefore, more useful to evaluate relative volatility than to
448 estimate the absolute temperatures at which these elements were mobilized. Note that the
449 chosen pressure is at the low end of pressures inside a 100 km planetesimal, given that the
450 permeabilities of chondrites determined in Bland et al. (2009) is in the 10^{-17} m² range indicating
451 slow gas flow at the scale of a planetesimal (Sugiura et al. 1984). This indicates that gas
452 pressure should be controlled by the lithostatic pressure.

453 In investigating the mobility of Sn in ordinary chondrites, an important question is to identify
454 the primary mineral hosts of Sn. In principle, most of the Sn should be contained in the metal
455 fraction and this is confirmed by a few analyses that reported Sn concentrations for metal and
456 silicate separates (Hamaguchi et al. 1969; Mason and Graham 1970). The analysis of Shima
457 (1964) showed a greater Sn concentration in the metal fraction by at least a factor of 10. For
458 the oxygen fugacities calculated in ordinary chondrites (Grossman et al. 2008), there should
459 indeed be a predominance of Sn as Sn(0) in the metal fraction with a small amount as Sn(II) in
460 the silicates. There should be by comparison less Sn stored in sulfides (e.g., van Acken et al.,
461 2012). Upon metamorphism, it has been suggested that Type 3 chondrites could have first been
462 oxidized due to the release of water and then reduced due to the presence of graphite and
463 organic matter (Huss et al., 2006), while Type 4 to 6 would have been oxidized (McSween and
464 Labotka, 1993), as water is released from phyllosilicates, which explains the positive correlation
465 between $\Delta^{17}\text{O}$ and fayalite content in olivine in ordinary chondrites (Rubin, 2005). The
466 associated oxidation in types 4-6 could have caused a redistribution of Sn into silicate phases by
467 reducing the metal/silicate partition coefficient.

468

469 In order to reevaluate whether Sn could be mobilized during parent body processes, we have
470 modelled the thermodynamic conditions during metamorphism between 200 K and 1200 K to
471 evaluate whether a significant amount of Sn could be mobilized in a gas phase containing water
472 that could oxidize Fe and release siderophile elements such as Sn or aqueous phase using the
473 FACTSAGETM software (Bale et al., 2009; Bale et al., 2016). We have not attempted to model
474 precisely the distribution of Sn in the silicate due to a lack of partitioning data. The FACTSAGE
475 software includes a large data base of solid solutions data for iron-dominated metals (FT-Stel)
476 that is particularly relevant for studying Sn that partitions into the iron metal at the relevant

477 oxygen fugacities. In order to model this system, we have used the chemical compositions of H
478 and L chondrites reported in Wasson and Kallemeyn (1988) for the following elements (H, C, O,
479 Na, Mg, Al, Si, S, Cl, Ca, Fe, Ni, Sn). Overall, there can be a sizeable amount of Sn in the gas
480 phase for temperature above 600-650 K. The main species in the vapor phase is SnCl₂ for
481 temperatures ranging between 750 K and 1200 K (Figure 9) at a pressure of 1 bar. This means
482 that there should be little Sn loss for the Type 3.1-3.3, while Sn loss could have taken place for
483 Types greater than 3.4 (see compilation of metamorphic temperatures by Huss et al. 2006).
484 Thus, it becomes clear with these simulations that the abundance of chlorine in ordinary
485 chondrites (11-400 ppm in type 3-4; Sharp et al. 2013; Clay et al. 2017) is sufficient to allow
486 partitioning of Sn in the gas phase at relatively low temperatures. It has been suggested by
487 McSween and Labotka (1993) and Rubin (2005) that metamorphism was accompanied by
488 oxidation and this was due to hydrous fluids. Thus, we also investigated the mobility of Sn in a
489 putative water phase. However, our thermodynamic calculations show that there was little Sn
490 in the aqueous phase. One may note also that the L chondrite parent body was presumably
491 affected by a cataclysmic event but since Sn isotopes in L chondrites seem rather similar to
492 those of H and LL chondrites and the $\delta^{124/116}\text{Sn}$ is not correlated with shock level in L chondrites,
493 it is hard to call upon a specific effect on Sn isotopes in the L chondrite group.

494
495 The exact effect of releasing SnCl₂ during metamorphism of ordinary chondrites (500-850 K) on
496 the Sn isotope signature is more difficult to assess. In principle, if the diffusion of Sn in the rock
497 to the porous space and the migration of SnCl₂ in the gas phase is associated with kinetic
498 isotope fractionation, then the SnCl₂ species should be enriched in light Sn isotopes, while the
499 solid residue should be enriched in heavy Sn isotopes. If this gas phase is later trapped in the
500 solid, then the solid should be enriched in light isotopes while getting enriched in Sn with an

501 effect depending on the initial Sn budget of the solid. If one assumes that the initial $\delta^{124/116}\text{Sn}$
502 and Sn concentrations are equal to that of Type 3 ordinary chondrites, this process would lead
503 to a negative trend in the $\delta^{124/116}\text{Sn}$ versus Sn concentration diagram, which does not
504 correspond to the observed pattern (Figure 8). On the other hand, it is possible that the release
505 of SnCl_2 took place with equilibrium isotope fractionation rather than kinetic isotope
506 fractionation. In this case, one predicts that at 700 K, the SnCl_2 species should be enriched in
507 heavy isotopes rather than light isotopes (Wang et al., 2019). Thus, the condensation of Sn-rich
508 vapor would result in an enrichment in Sn and in heavy Sn isotopes, a process that may explain
509 the enrichment in heavy Sn isotopes in our samples of Dhurmsala (LL6) and Forest Vale (H4).
510 While this scenario is difficult to verify with the existing data, it has the merit of being more
511 compatible with the observations.

512 Overall, our Sn isotope observations for ordinary chondrites are consistent with previous
513 volatile element data and suggests that superposition of nebular processes and parent body
514 metamorphism could explain the $\delta^{124/116}\text{Sn}$ variations. It is clear that a more detailed
515 description of the proposed scenario would require an investigation of Sn isotope systematics
516 at the mineral scale, which is still beyond current analytical capabilities due to low Sn
517 concentrations.

518

519 **4.3 A Sn isotope signature of volatile element depletion in the Earth**

520 The depletion in moderately volatile elements in the Earth is not fully understood. Similarly to
521 chondrites, its Sn isotope composition may be an indicator of the process of volatile element
522 depletion. Early work had suggested, based on the moderately volatile element pattern as a
523 function of condensation temperatures that the Earth was built from two components, a

524 volatile-free component and a volatile-rich component similar to CI chondrites (Anders et al.,
525 1977). This approach was reemphasized by later work defining the model of 'heterogeneous
526 accretion' for the Earth (Wanke and Dreibus, 1988; O'Neill, 1991). More recently, Schönbacher
527 et al. (2010) have shown that a heterogeneous accretion could account for the Pd-Ag
528 systematics of the Earth and chondrites. Based on modeling nucleosynthetic anomalies and O
529 isotopes, Fitoussi et al. (2016) showed that the Earth could be built by a mixture of angrites, H,
530 CI, and CV chondrites that are heterogeneous in their volatile element contents with an overall
531 CI + CV chondrite contribution of 18%, while Dauphas (2017) argued that the Earth was mainly
532 built from an enstatite-like reservoir. More recently, Budde et al. (2019) showed that the last
533 10-20% of accretion required a carbonaceous chondrite component to explain the Mo isotopes
534 in the bulk silicate Earth.

535 The Sn isotope composition of the bulk silicate Earth is identical to that of carbonaceous
536 chondrites. However, as Sn is a siderophile element, most of the Sn budget should reside in the
537 core rather than the silicate Earth. In order to assess the Sn isotope composition of the bulk
538 Earth, one must first determine the effect of core formation on the Sn isotope composition. In
539 principle, Sn is considered as a moderately siderophile element with an estimated bulk
540 metal/silicate partition coefficient close to ~ 10 for the conditions of terrestrial core formation
541 (Righter et al., 2018). This suggests that the Sn isotope composition of BSE could potentially be
542 affected by core formation and this effect must first be assessed before focusing on the Sn
543 depletion in the bulk Earth. We established a simple model for Sn isotopic fractionation during
544 Earth's core formation as a function of various parameters (P , T , the oxygen fugacity fO_2 , etc.)
545 using the metal-silicate partition coefficient defined in the Supplementary Material. Then,
546 following Righter et al. (2017), we have used the liquidus for mantle melting of Andraut et al.

547 (2011) which is a function of pressure and two different cases: a fixed fO_2 at IW-2 or a variable
 548 fO_2 ranging between IW-4 and IW-1.6 over the considered temperature range (2000-4000 K).
 549 The oxygen fugacity increases incrementally with accretion as described in Righter et al. (2017).
 550 Figure 10 shows the calculated partition coefficient of Sn between metal and silicate (D_{Sn}) as a
 551 function of temperature corresponding to the two evolution models. It demonstrates that with
 552 fixed fO_2 , D_{Sn} is almost constant, while with variable fO_2 between IW-4 and IW-1.6, D_{Sn}
 553 decreases with increasing temperature and pressure.

554 Altogether, the difference of the Sn isotope compositions between the BSE and the bulk Earth
 555 expressed as $\Delta^{124}Sn_{BSE-Bulk Earth}$, can be calculated by the following equation (see
 556 Supplementary Material for derivation):

$$557 \quad \Delta^{124/116}Sn_{BSE-Bulk Earth} = \left[\frac{D_{Sn}X_{core}}{D_{Sn}X_{core}+1-X_{core}} \right] \times A \times T^{-2} \quad (2)$$

558 where X_{core} is the mass fraction of core in the bulk planet. A is a constant equal to 0.32×10^6
 559 for SnO-Sn system and 0.776×10^6 for SnO₂-Sn system, respectively. Since D_{Sn} can be
 560 expressed as a function of temperature (see Supplementary Material), it is possible to
 561 express $\Delta^{124/116}Sn_{BSE-Bulk Earth}$ as a function of temperature. The results of model
 562 calculations predicting $\Delta^{124/116}Sn_{BSE-Bulk Earth}$ as a function of temperature are plotted in
 563 Figure 11 for various evolutions of fO_2 during core formation. The deeper P - T conditions
 564 where metal and silicate equilibrated during core formation in the Earth are estimated to be
 565 higher than 3000 K and up to 3800 K (Righter 2011; Siebert et al., 2011; Fischer et al., 2015),
 566 associated with a maximum $\Delta^{124}Sn_{BSE-Bulk Earth}$ value of 0.005 ‰ (for Sn(II)-Sn(0)
 567 fractionation) and 0.05 ‰ for Sn(IV)-Sn(0) fractionation (Figure 11), which is much smaller
 568 than the currently achieved precision of $\delta^{124/116}Sn$ measurements (± 0.11 ‰, 2 s.d.).

569 Therefore, the effect of core formation on Sn isotopes can be neglected in the case of the
570 Earth. The Sn isotope composition of BSE is predicted to be identical, within error, to that of
571 the bulk Earth, with a $\delta^{124/116}\text{Sn}$ value of $-0.08 \pm 0.11\%$ (2 s.d.) as determined in Wang et al.
572 (2018). This difference is comparable with the results obtained by Badullovich et al. (2017)
573 and Creech and Moynier (2019), although the absence of a common standard in the two sets
574 of studies makes a strict comparison difficult. As a result, the $\delta^{124/116}\text{Sn}$ value of the bulk
575 Earth is identical to that of carbonaceous chondrites. However, Cr nucleosynthetic anomalies
576 found in the Earth are not identical to those of carbonaceous chondrites (Trinquier et al.,
577 2007), indicating that the Earth is not predominantly built from carbonaceous chondrites.
578 Our modeling also disagrees with the interpretations of Creech and Moynier (2019) who
579 argued that an enstatite chondrite-like Earth could explain the observed BSE composition
580 due to the effect of core formation. The nature of the Earth's building blocks is a topic of
581 discussion (Dauphas 2017; Fitoussi et al., 2016) and the stable isotope composition of
582 building blocks could have been affected by early processes such as those described for
583 chondrites or by large impacts or magma ocean volatilization (Hin et al., 2017; Pringle et al.,
584 2014). However, in the context of a model where volatile elements were accreted to the
585 Earth by carbonaceous chondrite like material (e.g., Fitoussi et al. 2016), it is possible that an
586 early magma ocean had operated and produced a proto-Earth with an isotopically
587 fractionated Sn signature. However, the budget of Sn was subsequently overwhelmed by Sn-
588 rich material identical to that of carbonaceous chondrites. If one were to argue that the Earth
589 had acquired its volatile budget from ordinary chondrites, there should be a process able to
590 raise the $\delta^{124}\text{Sn}$ of these chondrites (mean $\delta^{124}\text{Sn}$ value = $-0.707 \pm 0.27\%$ 2 s.e.) to the BSE
591 value. One would have to call upon a large-scale evaporation such as a magma ocean that
592 would have fortuitously raised the $\delta^{124}\text{Sn}$ to a value identical to that of carbonaceous

593 chondrites and of the BSE. In addition, there is no signature of partial condensation
594 (Albarède, 2009) or removal of a sulfide component (Pringle et al., 2017) as has been argued
595 for Zn isotopes. If the Earth's budget in moderately volatile elements is due to the addition of
596 a volatile rich component, then this component must have had an Sn isotope signature
597 similar to carbonaceous chondrites (as seen with other isotope systems, e.g., Wombacher et
598 al., 2008).

599

600 **5. Conclusions**

601 The Sn isotope compositions of four carbonaceous chondrites including Orgueil CI, Murchison
602 CM, NWA 5240 CV and Allende CV have been analyzed and exhibit $\delta^{124/116}\text{Sn}$ values within a
603 limited range (average $0.04 \pm 0.11\%$, 2 s.d.) which is comparable to that of BSE ($-0.08 \pm 0.11\%$, 2
604 s.d., Wang et al., 2018). This result indicates that the Sn depletion in carbonaceous chondrites
605 cannot be used to discriminate partial evaporation from partial condensation in the early Solar
606 Nebula. Instead, it suggests that Sn isotope fractionation was limited and this could be due to
607 thermodynamic equilibrium conditions between condensed phase and the ambient vapor in
608 the early Solar Nebula. However, in light of other observations, the Sn isotope data is more
609 consistent with a model involving the mixing of a refractory component (depleted in Sn) with a
610 CI-like component volatile rich component. This model would agree with the general volatile
611 element pattern in carbonaceous chondrites.

612 The Sn isotope record in carbonaceous chondrites is consistent with other volatile elements
613 that have constant relative abundances and are linked to the abundance of a matrix-like
614 component. In contrast, other less volatile elements such as K, Rb and Cu (and possibly also Zn)

615 show an isotope fractionation pattern indicating that part of their budget is contained in the
616 refractory component of carbonaceous chondrites. A better knowledge of the condensation
617 temperature of moderately volatile element would be desirable to fully understand their
618 isotope signatures.

619 By comparison, ordinary chondrites show greater variations in Sn isotope compositions and are
620 most often enriched in light Sn isotopes compared with carbonaceous chondrites. The $\delta^{124/116}\text{Sn}$
621 values of ordinary chondrites show no correlation with degrees of metamorphism (Type 3 to 6)
622 or groups (H, L, LL) but the overall dispersion in $\delta^{124/116}\text{Sn}$ could be explained by volatilization of
623 SnCl_2 during metamorphism. The enrichment in light Sn isotopes found in unequilibrated
624 ordinary chondrites could be the consequence of partial condensation of Sn due to a kinetic
625 isotope effect but the details of such a process would require mineral scale analysis of Sn
626 isotopes. Last, our modeling shows that the Sn isotope composition of the bulk Earth should be
627 identical to that of the bulk silicate Earth and to that of carbonaceous chondrites, because of
628 the limited fractionation effect expected from core formation.

629

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636

637 **Figure captions**

638 Figure 1 $\delta^{124/116}\text{Sn}$ analyzed by MC-ICPMS using double-spike technique. $\delta^{124/116}\text{Sn}$ of BSE is
639 from Wang et al. (2018). All the uncertainties better than AGV-1 (0.11‰ for 2 s.d.) are reported
640 as 0.11‰.

641
642 Figure 2 $\delta^{124/116}\text{Sn}$ versus $\delta^{124/118}\text{Sn}$ as listed in Table 1, with a slope of 0.743 ± 0.04 . All the
643 uncertainties better than AGV-1 are given as 0.11‰ (2 s.d.) on $\delta^{124/116}\text{Sn}$ and 0.10‰ (2 s.d.) on
644 $\delta^{124/118}\text{Sn}$.

645
646 Figure 3 (a) Sn/Si normalized to CI chondrites versus mass fraction of matrix in % for
647 carbonaceous chondrites and Tagish Lake meteorite (TL). Sn and Si concentrations are from
648 Alexander (2019a) except for Sn concentrations for CI and CV chondrites which are from this
649 study. (b) Sn concentrations versus mass fraction of matrix in % for carbonaceous chondrites
650 and Tagish Lake meteorite (TL). The Y-intersect is not equal to zero, indicating the presence of
651 Sn in the chondrule component.

652
653 Figure 4 Condensation curves for a selection of moderately volatile elements using the
654 FACTSAGE™ software with a solar composition from Lodders (2003) with a dust enrichment
655 factor of 1000 and $P = 10^{-3}$ atm. Activity coefficients for trace elements were taken from Wood
656 et al. (2019). Note the significant shift in the condensation temperature of Zn and Te relative to
657 Sn that is consistent with the difference in their isotopic signatures. The dashed line (a) show
658 the condensation temperature necessary to obtain isotope fractionation in Zn, Cu and K, while
659 line (b) corresponds to the condensation temperature for isotope fractionation of Te and Rb. Cd
660 and Sn should show no fractionation in both cases.

661
662 Figure 5 Compilation of Sn concentrations versus Zn concentrations in chondrites. Data source
663 are: Wasson and Kallemeyn (1988) for the carbonaceous chondrites, Friedrich et al. (2003),
664 Friedrich et al. (2004), Friedrich et al. (2014) for the L and LL chondrites and Creech and
665 Moynier (2019).

666

667

668 Figure 6 Diagram illustrating the effect of (a) partial condensation and (b) partial evaporation
669 using a Rayleigh distillation in a $\delta^{66/64}\text{Zn}$ versus Zn normalized to the Zn concentration in CI. The
670 $\delta^{66/64}\text{Zn}$ values are given relative to the value for CI chondrite from Luck et al. (2005). Zn isotope
671 data are from Luck et al. (2005) and Mahan et al. (2018a). The various curves are calculated as
672 a function of $P_{i\text{ sat}}/P_i$ (ratio of saturation pressure to the partial pressure of the isotope of
673 interest i) for diagram (a) and $P_i/P_{i\text{ sat}}$ for diagram (b) with the model of Dauphas et al. (2015) for
674 the global isotope fractionation factor. Three conditions were calculated using $P_{i\text{ sat}}/P_i = 0.9,$
675 0.95 and 1 for diagram (a) and $P_i/P_{i\text{ sat}} = 0.95, 0.98$ and 1 for diagram (b), respectively. The
676 kinetic isotope fractionation factor is equal to $\sqrt{64/66}$. The equilibrium isotope fractionation
677 factor was taken from Ducher et al. (2016) corresponding to an equilibrium between ZnO(solid)
678 and Zn(gas).

679
680
681 Figure 7 Histograms of Sn concentrations in H chondrites, L chondrites, and LL chondrites. Data
682 source are Friedrich et al. (2003), Friedrich et al. (2004), Friedrich et al. (2014) for the L and LL
683 chondrites and Creech and Moynier (2019) and this study. The data shows no difference
684 between the three ordinary chondrite groups.

685
686 Figure 8 $\delta^{124/116}\text{Sn}$ versus Sn concentrations in ordinary chondrites (data from Table 1). The
687 unequilibrated chondrites are marked with a black circling. The only positive $\delta^{124/116}\text{Sn}$ are
688 found for equilibrated chondrites.

689
690 Figure 9 Sn volatility in ordinary chondrites calculated at 1 atm. The Sn speciation was
691 calculated for an H chondrite bulk composition and the main volatile species is SnCl_2 . The other
692 species are SnCl_4 or SnS but are too low to be shown in this diagram. FCC and BCC indicates two
693 allotropes of the iron alloy. The calculations were made for three Cl concentrations: 11 ppm, 80
694 ppm, 400 ppm corresponding to the minimum, the mean and the maximum concentrations,
695 respectively. The minimum and the maximum Cl concentrations are from the literature (Sharp
696 et al. 2013; Clay et al. 2017).

697 Figure 10 (a) Calculated metal-silicate partition coefficient for tin as a function of temperature
698 (in K) and pressure corresponding to the mantle melting curve of Andrault et al. (2011) (after
699 Righter et al., 2017). The model curves were calculated for two evolution models. The fixed fO_2

700 cases correspond to IW-2, while the variable fO_2 values range from IW-4 to IW-1.6, as might be
701 expected during a heterogeneous accretion scenario. The light green boxes illustrate the
702 temperature of the magma ocean.

703
704 Figure 11 Calculated difference of $\delta^{124/116}\text{Sn}$ between the BSE and the bulk Earth expressed as
705 $\Delta^{124/116}\text{Sn}_{\text{BSE}-\text{Bulk Earth}}$ as a function of temperature corresponding to the two fO_2 evolution
706 models. (a): model curves calculated assuming Sn(IV) is the dominant species in the silicate
707 melt, (b) : model curves calculated assuming Sn(II) is the dominant species in the silicate melt.
708 In both cases, $\Delta^{124/116}\text{Sn}_{\text{BSE}-\text{Bulk Earth}}$ at temperatures > 3000 K is below our measurement
709 precision of 0.11 ‰ (2 s.d.).

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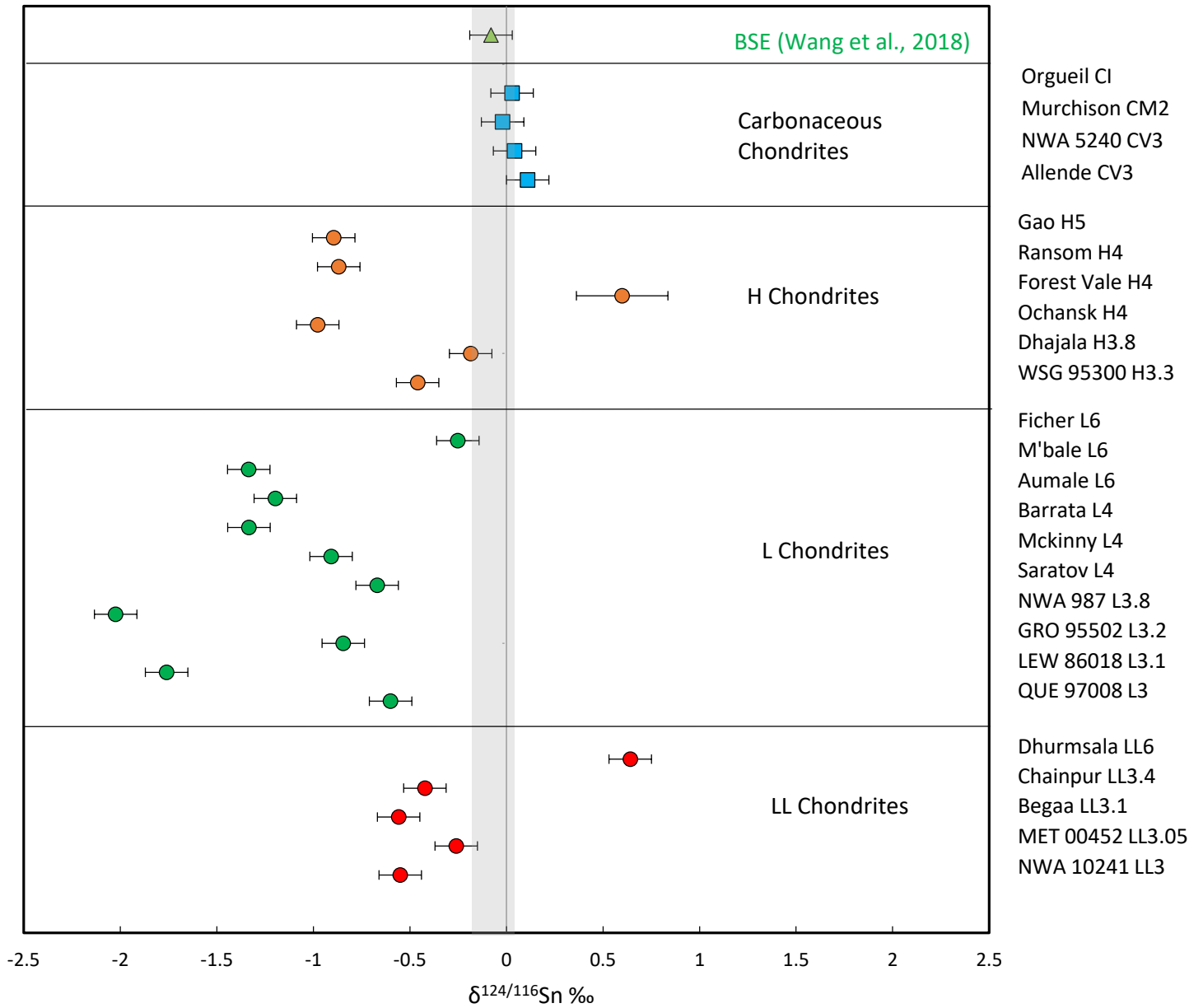
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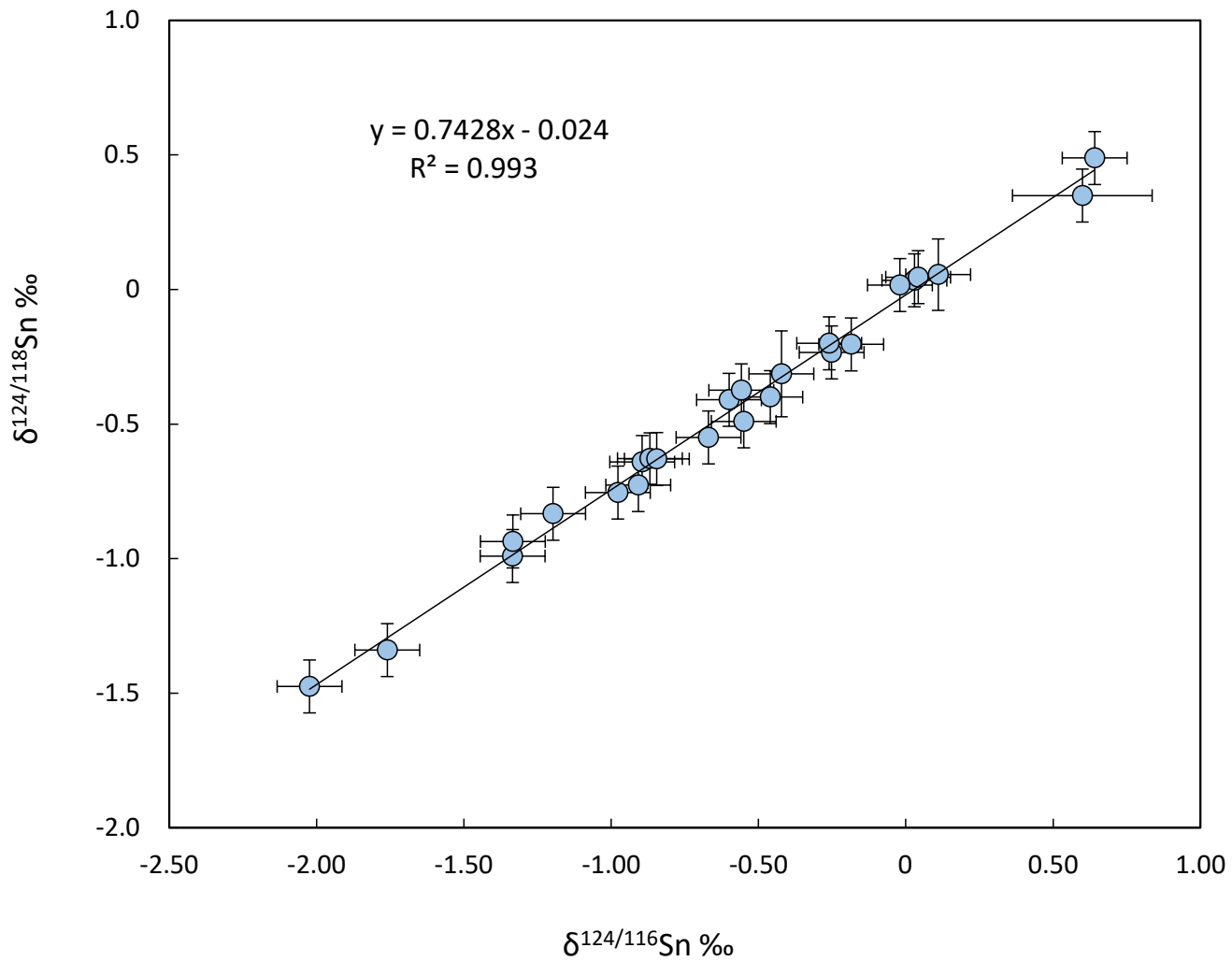


Figure 3

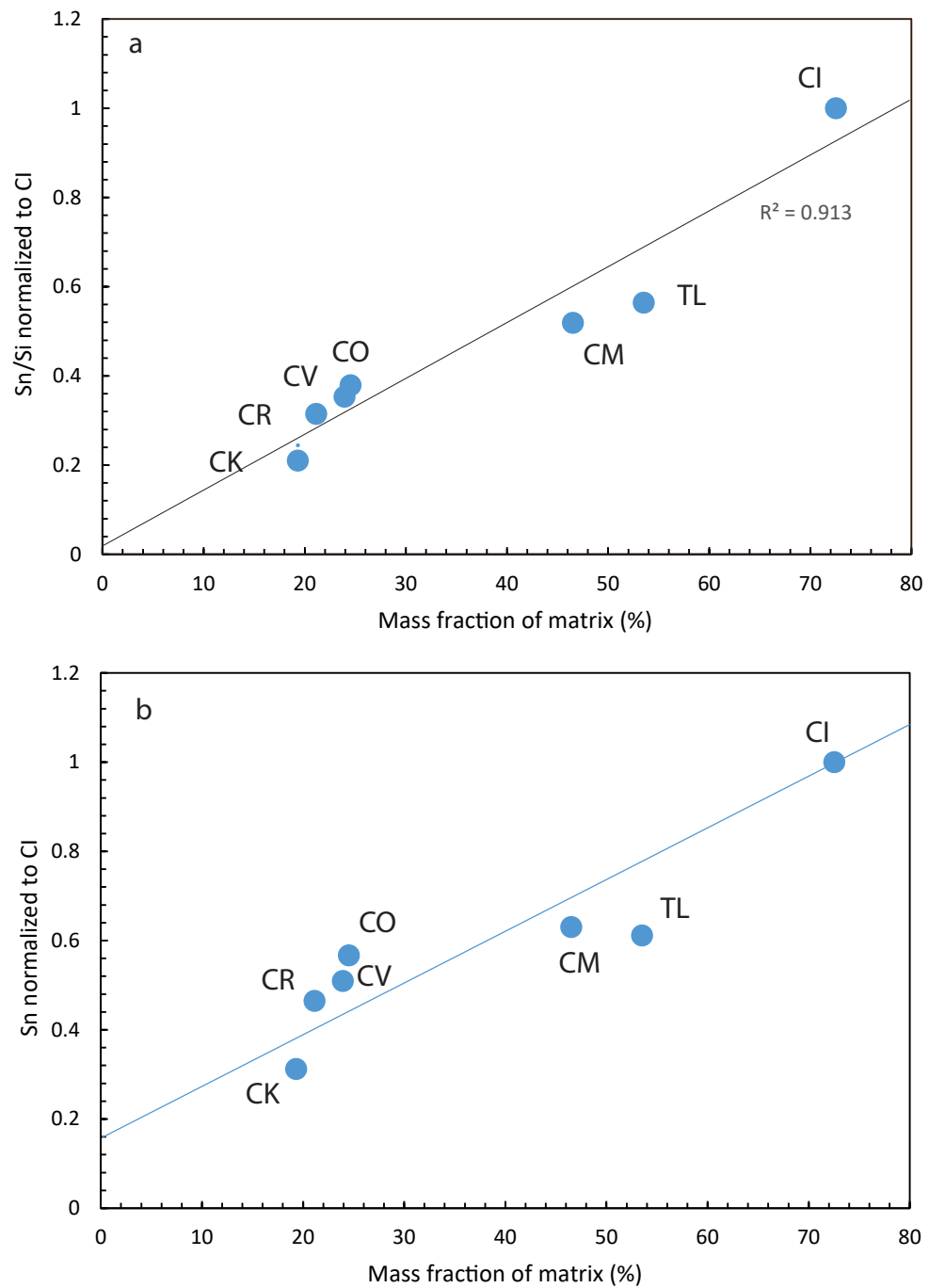


Figure 4

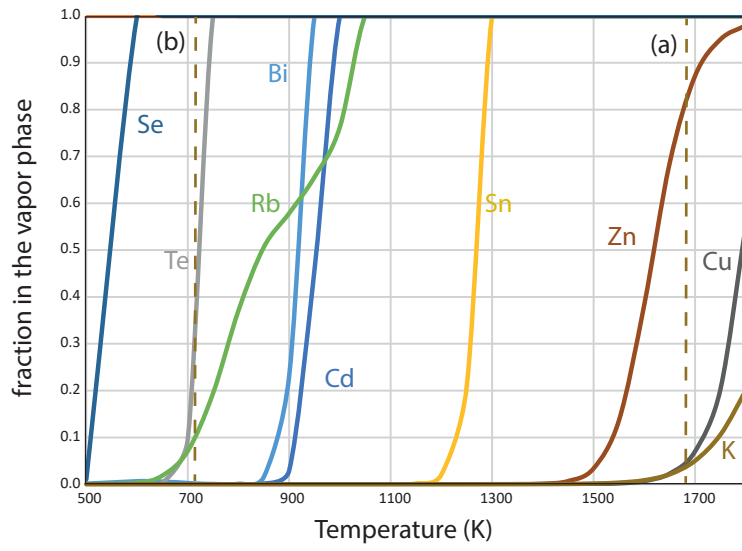


Figure 5

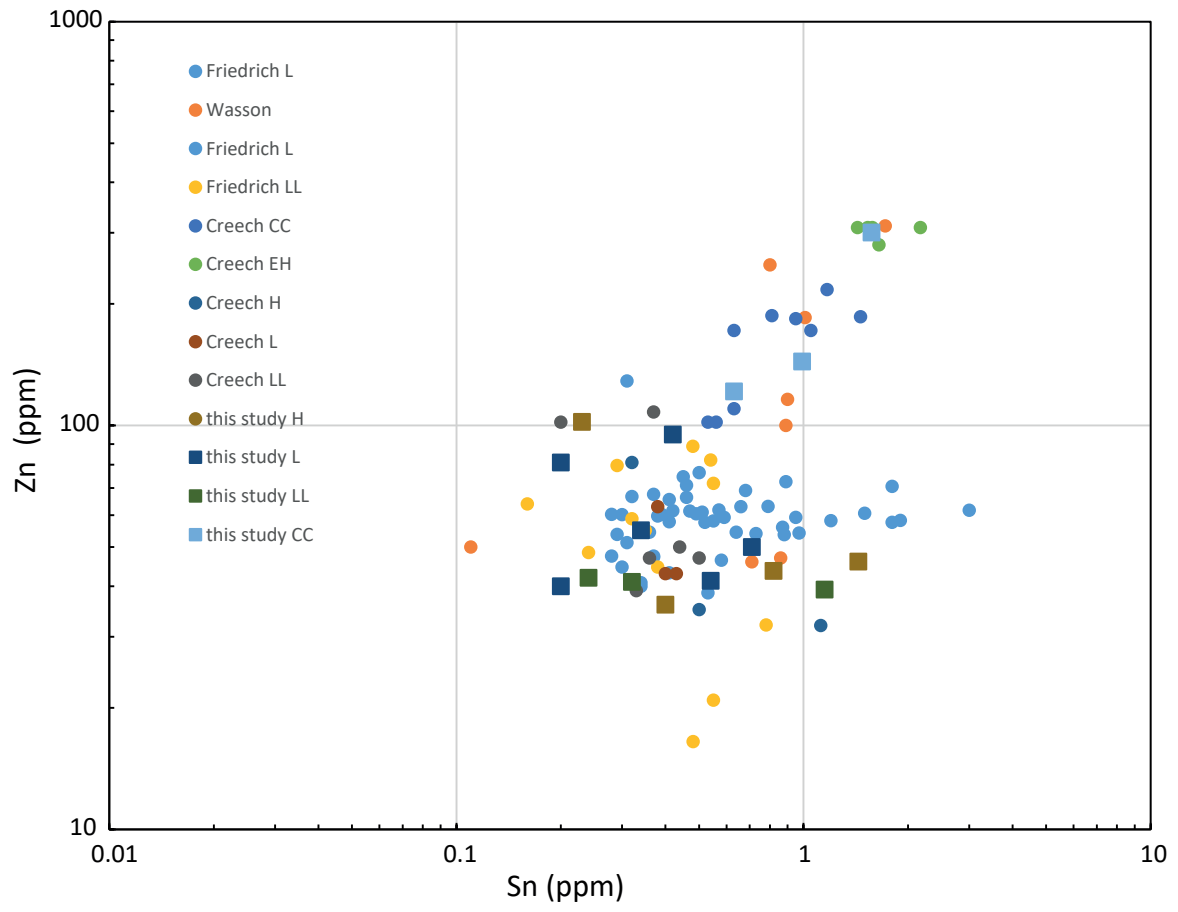
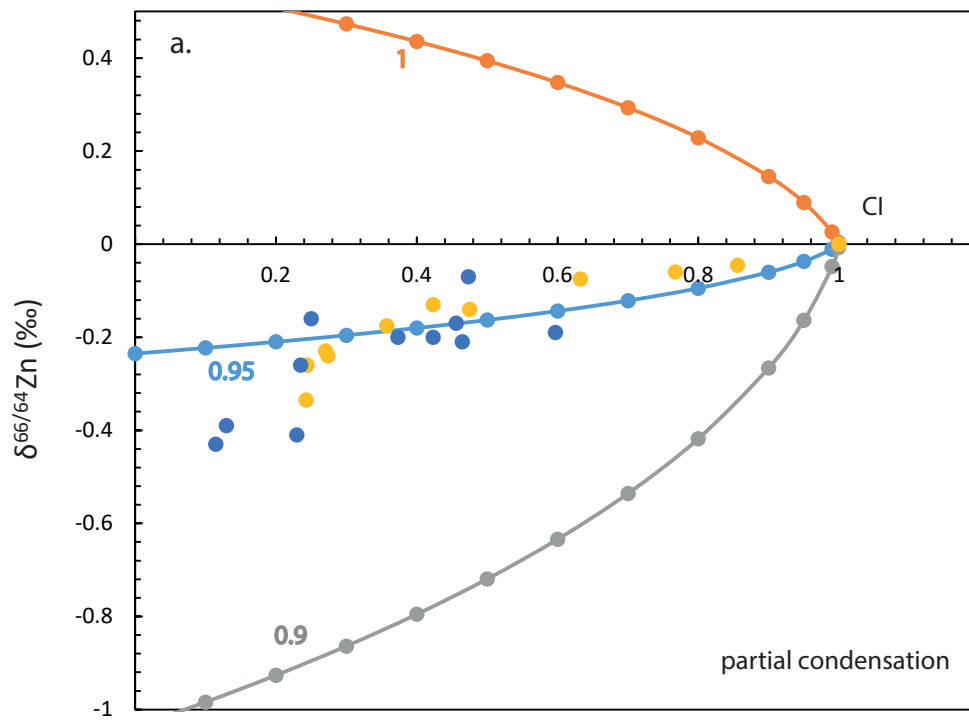
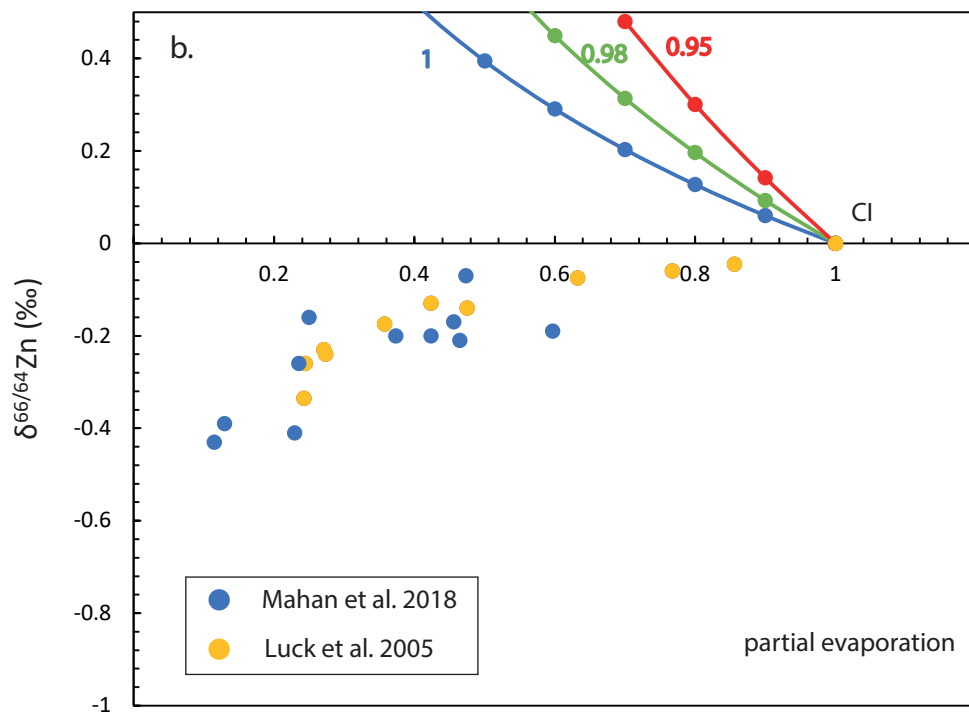


Figure 6



fraction of zinc in solid



fraction of zinc in solid

Figure 7

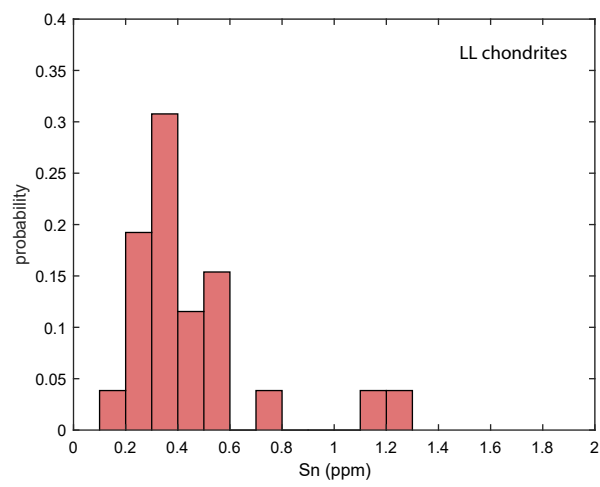
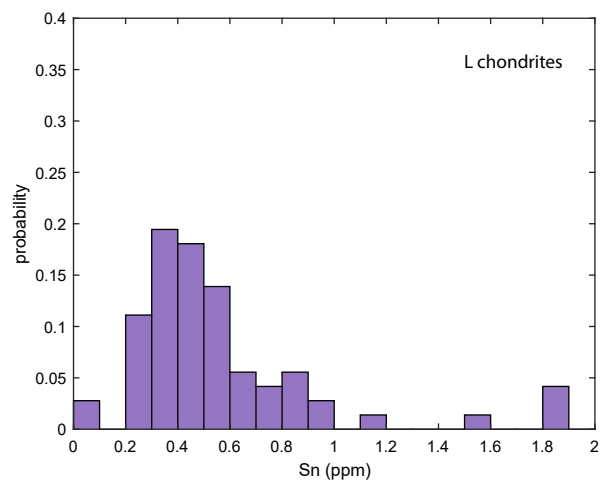
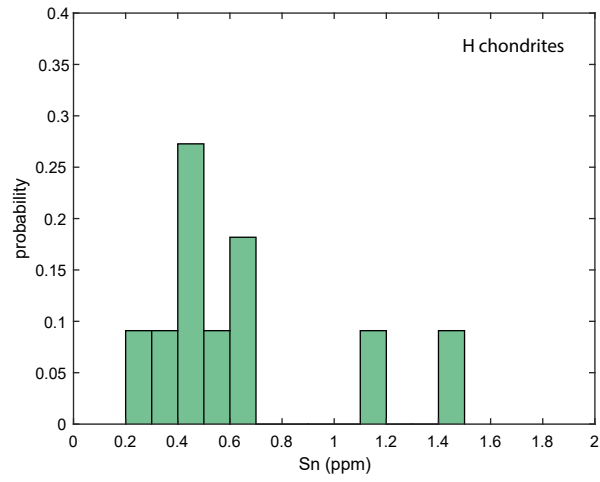


Figure 8

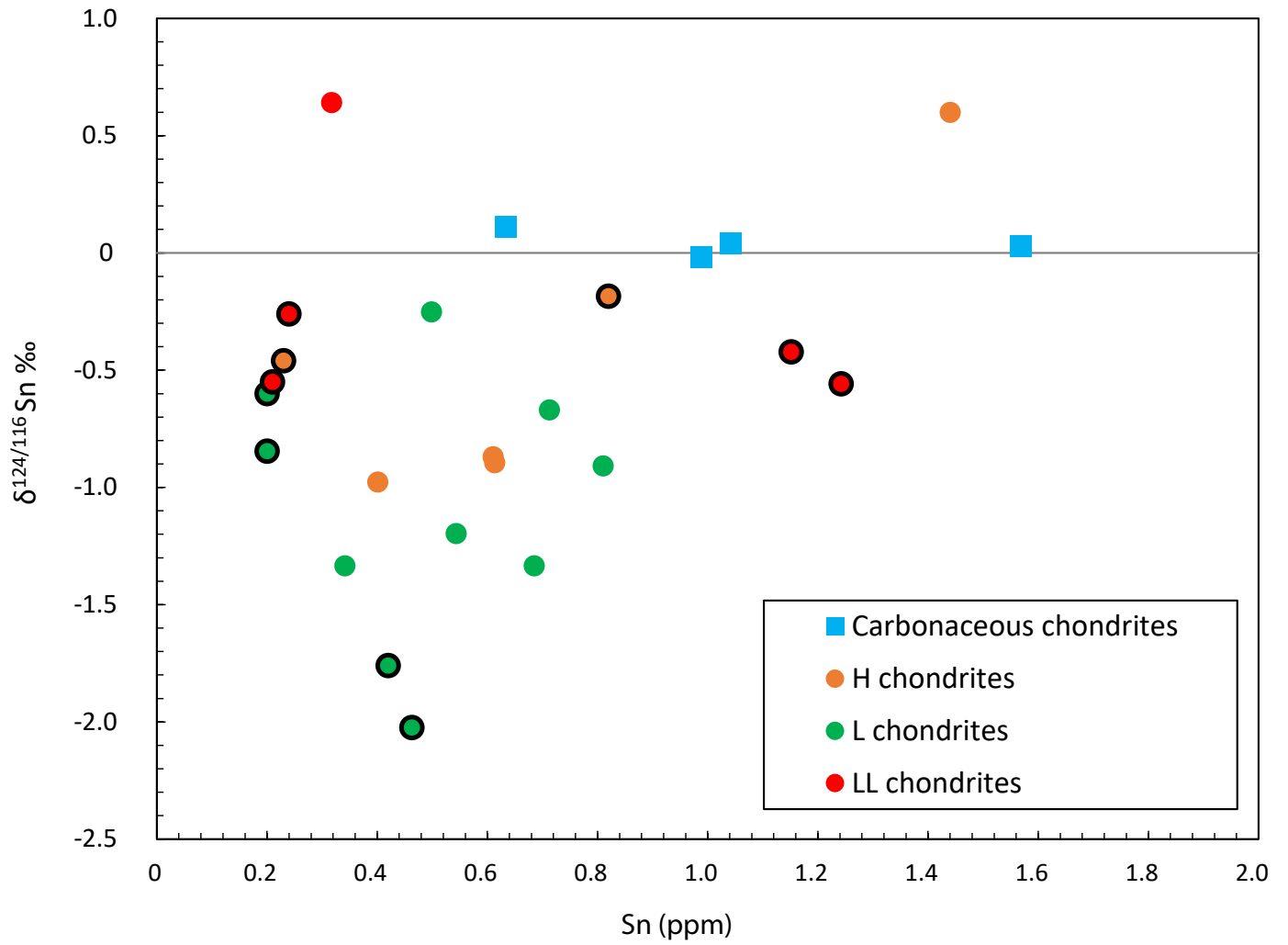


Figure 9

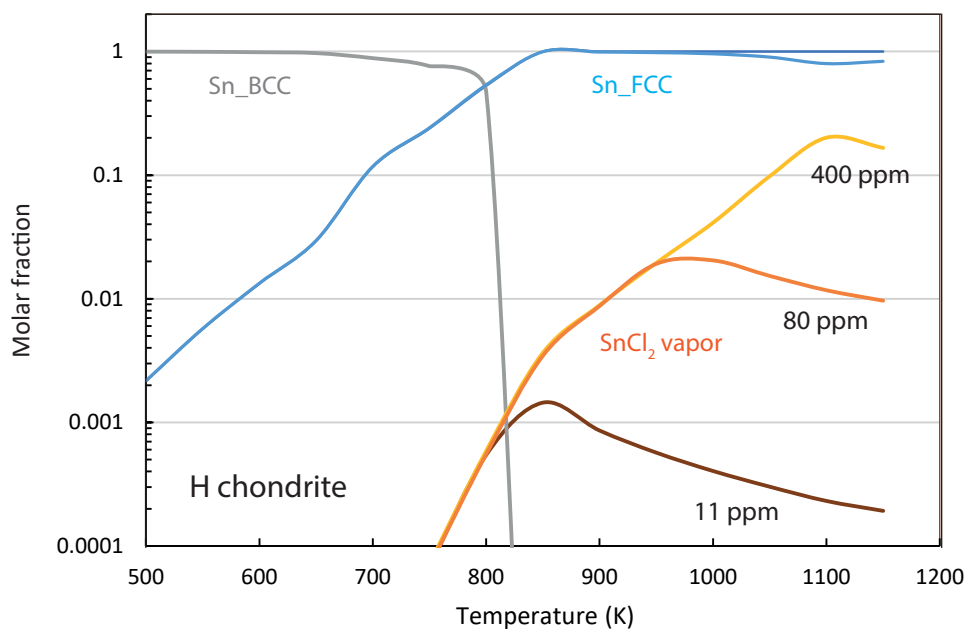


Figure 10

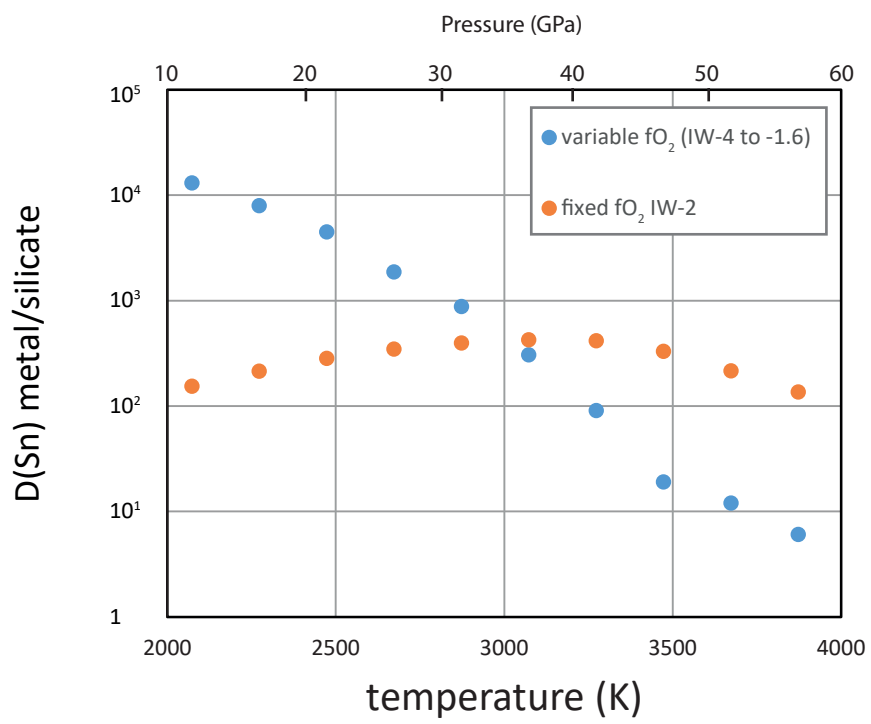


Figure 11

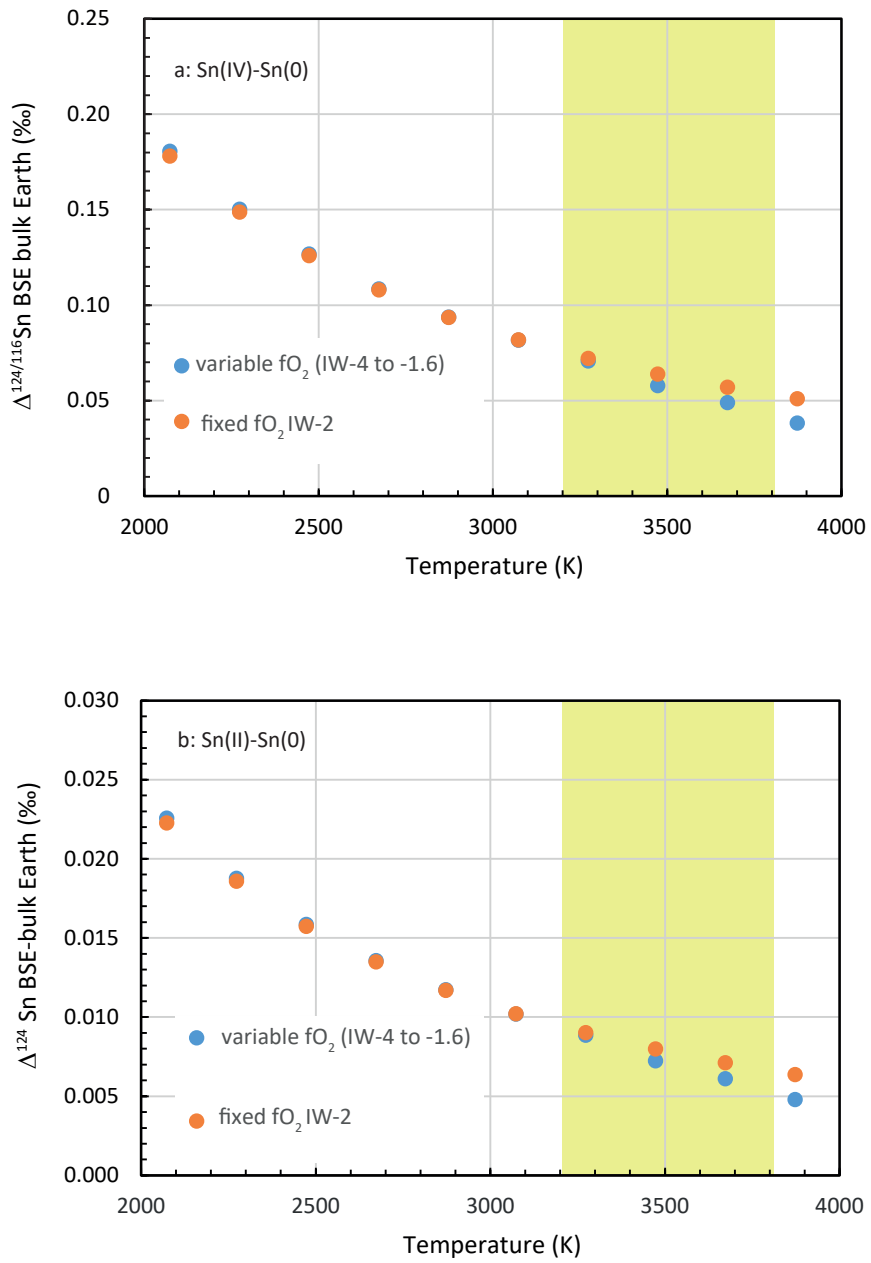


Table 1. Sn isotope compositions and concentrations in chondrite samples analyzed in this study.

Sample Name	Origin	Shock level	$\delta^{124}\text{Sn}$ (‰)	2 s.d. (‰)	$\delta^{124/118}\text{Sn}$ (‰)	2 s.d. (‰)	Sn (ppm)	Sample mass (mg)	N*
<i>Carbonaceous chondrites</i>									
Orgueil C11	France (fall)		0.03	0.003	0.03	0.01	1.57±0.13	29.1	2
Murchison CM2	Australia (fall)		-0.02	0.05	0.02	0.05	0.99±0.08	51.5	2
NWA 5240 CV3	Northwest Africa		0.04	0.01	0.05	0.02	1.04±0.08	59.0	2
Allende CV3	Mexico (fall)		0.11	0.07	0.06	0.13	0.63±0.01	148.4-151.5	11
Average CC			0.04	0.11					
<i>Ordinary chondrites</i>									
Gao-Guenie H5	Burkina faso (fall)	S1-2	-0.89	0.06	-0.64	0.03	0.61±0.05	150.5	2
Ransom H4	United States	–	-0.87	0.01	-0.63	0.10	0.61±0.05	78.2	2
Forest Vale H4	Australia (fall)	S2	0.60	0.24	0.35	0.01	1.44±0.12	57.1	2
Ochansk H4	Russia (fall)	S3	-0.98	0.01	-0.75	0.01	0.40±0.03	79.5	1
Dhajala H3.8	India (fall)	S1	-0.19	0.02	-0.20	0.02	0.82±0.07	40.2	2
WSG 95300 H3.3	Antarctica	S2	-0.46	0.05	-0.40	0.05	0.23±0.02	307.4	2
Fisher L6	United States (fall)	S5	-0.25	0.003	-0.23	0.02	0.50±0.04	139.8	2
Mbale L6	Uganda	S6	-1.33	0.01	-0.99	0.01	0.68±0.05	149.6	1
Aumale L6	Algeria (fall)	S2	-1.20	0.02	-0.83	0.04	0.54±0.04	53.2-68.1	2
Barratta L4	Australia	S4	-1.33	0.01	-0.94	0.01	0.34±0.03	81.8	2
Mckinney L4	United States	S4-5, S6	-0.91	0.04	-0.73	0.09	0.81±0.06	74.3	2
Saratov L4	Russia (fall)	S2	-0.67	0.02	-0.55	0.02	0.71±0.04	75.2	4
NWA 987 L3.8	Northwest Africa	S5	-2.02	0.03	-1.48	0.02	0.46±0.04	79.3	2
GRO 95502 L3.2	Antarctica	S3	-0.85	0.01	-0.63	0.05	0.20±0.03	156.8	2
LEW 86018 L3.1	Antarctica	S2	-1.76	0.03	-1.34	0.02	0.42±0.03	157.2	2
QUE 97008 L3.05	Antarctica	S3	-0.60	0.05	-0.41	0.09	0.20±0.03	303.2	2
Dhurmsala LL6	India (fall)	S3	0.64	0.01	0.49	0.01	0.32±0.03	78.5	1
Chainpur LL3.4	India (fall)	S1	-0.42	0.04	-0.31	0.16	1.15±0.09	69.2	3
Begaa (NWA 4910) LL3.1	Northwest Africa	S2	-0.56	0.02	-0.37	0.02	1.24±0.10	51.1	2
MET 00452 LL3.05	Northwest Africa	–	-0.26	0.02	-0.20	0.003	0.24±0.03	308.1	2
NWA 10241 LL3	Antarctica	S3	-0.55	0.001	-0.49	0.02	0.21±0.02	253.0	2

*The number of replicates N are the sum of measurement replicates. Allende(CV3) and Saratov(L4) have four and two full chemistry replicates, respectively, with three or two measurements for each full replicate. The uncertainties of these two samples are also for full chemistry replicates given as 2 s.d.