

Titanium isotope source relations and the extent of mixing in the proto-Solar nebula examined by Independent Component Analysis.

Robert C. J. Steele and Patrick Boehnke

Department of Earth, Planetary, and Space Sciences, University of California, Los Angeles, CA 90095

February 11, 2015

ABSTRACT

The Ti isotope variations observed in hibonites represent some of the largest isotope anomalies observed in the Solar System. Titanium isotope compositions have previously been reported for a wide variety of different early Solar System materials, including calcium, aluminium rich inclusions (CAIs) and CM hibonite grains, some of the earliest materials to form in the Solar System, and bulk meteorites which formed later. These data have the potential to allow mixing of material to be traced between many different regions of the early Solar System. We have used Independent Component Analysis to examine the mixing end-members required to produce the compositions observed in the different datasets. The ICA yields results identical to a linear regression for the bulk meteorites. The components identified for hibonite suggest that most of the grains are consistent with binary mixing from one of three highly anomalous nucleosynthetic sources. Comparison of these end-members show that the sources which dominate the variation of compositions in the meteorite parent body forming regions was not present in the region in which the hibonites formed. This suggests that the source which dominates variation in Ti isotope anomalies between the bulk meteorites was not present when the hibonite grains were forming. One explanation is that the bulk meteorite source may not be a primary nucleosynthetic source but was created by mixing two or more of the hibonite sources. Alternatively, the hibonite sources may have been diluted during subsequent nebula processing and are not a dominant Solar System signatures.

Subject headings: astrochemistry; meteorites, meteors, meteoroids; methods: statistical; nuclear reactions, nucleosynthesis, abundances; protoplanetary disks; supernovae: general

1. Introduction

1 Isotope anomalies in primitive meteorites
2 and their components can be used to examine
3 the nucleosynthetic origins of the Solar Sys-
4 tem (e.g. Lee et al. 1978). The various ma-
5 terials we sample in meteorites, for example
6 CAIs, chondrules and achondrites, formed in

7 different parts of the proto-Solar nebula, and
8 at different times. Therefore, these samples
9 naturally record some of the variation in iso-
10 topic compositions present in the early So-
11 lar System. Investigation of the differences
12 in anomalies between these distinct materials
13 can be used to examine their genetic relation-
14 ships and the processes by which the different

15 sources were mixed together to yield the vari-
16 ation in compositions we observe today.

17 Isotope anomalies in refractory elements
18 with more than four isotopes, for example Ti,
19 Ni and Mo, are highly correlated through the
20 bulk meteorites, both chondrites and achon-
21 drites, and the population of normal CAIs
22 (e.g. Trinquier et al. 2009; Steele et al. 2010,
23 2011, 2012; Burkhardt et al. 2011). This sys-
24 tematic variation occurs in most early Solar
25 System objects. There are, however, a few no-
26 table exceptions to this systematic variation,
27 most interestingly the FUN (fractionated with
28 unknown nuclear effects) CAIs and the CM hi-
29 bonite grains. These are relatively poorly un-
30 derstood populations of refractory inclusions
31 found in primitive meteorites.

32 Hibonite, the focus of this study, is a highly
33 refractory minerals thought to have been one
34 of the first minerals to have formed in the So-
35 lar System either as a refractory residue or
36 by condensation from a nebula gas (Ireland
37 et al. 1988). Hibonite grains from the CM
38 carbonaceous chondrites (CCs) exhibit some
39 of the largest isotope anomalies of any mate-
40 rials thought to have formed within the So-
41 lar System. The evidence that the hibonite
42 grains are of Solar origin stems from their O
43 isotope compositions within the normal range
44 for Solar System objects (Fahey et al. 1987a;
45 Ireland et al. 1992; Liu et al. 2009). How-
46 ever, they exhibit ^{50}Ti enrichments of up to
47 27 % (Ireland 1990) and deficits of up to 7
48 % (Hinton et al. 1987). Several populations
49 of hibonite grains have been documented in
50 the literature (Ireland et al. 1988; Marhas
51 et al. 2002), though they can broadly be split
52 into two groups based on their petrology and
53 their isotopic compositions. The first group is
54 petrologically characterised by platy hibonite
55 crystal fragments (PLACs from Ireland et al.
56 1988) and blue aggregates (BAGs from Ire-
57 land et al. 1988). This group shows the large
58 mass-independent isotope anomalies in ele-
59 ments such as Ti and Ca described above and

60 no evidence for live short-lived radionuclides
61 (SLRs) such ^{26}Al (Ireland et al. 1988; Ireland
62 1990). The second group, comprised of spinel
63 and hibonite rich spherules (SHIBs from Ire-
64 land et al. 1988), show much smaller mass-
65 independent isotope anomalies but do show
66 evidence for live SLRs for example canonical,
67 or supra-canonical ^{26}Al (Ireland et al. 1988;
68 Ireland 1990; Liu et al. 2009).

69 The large variation of mass-independent
70 isotope anomalies found in hibonites are
71 thought to represent either pure, or only
72 slightly diluted, presolar signatures (e.g. Ire-
73 land 1990). However, it is not known if the
74 events which produced the hibonites occurred
75 within the Solar System at some time be-
76 fore the large scale chemical and isotopic ho-
77 mogenisation. Alternatively, the hibonites
78 may have formed outside the Solar System,
79 possibly closer to the source of the isotopic
80 anomalies, for example around a star in some
81 region of the molecular cloud which had sim-
82 ilar oxygen isotope compositions. Therefore,
83 it is clearly of great importance to assess the
84 relationship of the anomalies observed in the
85 hibonites to those observed in bulk meteorites
86 and other Solar System materials. Examining
87 these relationships will offer insight into
88 whether the hibonites represent purer samples
89 of the isotopic precursors of the bulk Solar
90 System and sampled by the bulk meteorites
91 or if they represent a previously unsampled
92 reservoir. An example of this type of finding
93 is the recent discovery of oxide grains highly
94 enriched in ^{54}Cr which are thought to be the
95 carrier phase of mass-independent Cr isotope
96 anomalies in the Solar System (Dauphas et al.
97 2010; Qin et al. 2011). The alternative, that
98 the hibonites formed outside the Solar Sys-
99 tem, possibly prior to Solar System formation,
100 would mean that hibonite grains do not have
101 isotopic significance for the bulk Solar Sys-
102 tem. Rather they represent a new population
103 of presolar grain that is physically an order
104 of magnitude larger than the previous largest
105 population (with exception of certain very rare

106 examples e.g., Zinner et al. 2010). However,
107 previous presolar grain populations have very
108 large O isotope variations, several orders of
109 magnitude larger than those observed in CM
110 hibonites (Fahey et al. 1987b; Zinner 2003)

111 In order to examine the relationships be-
112 tween hibonite grains and bulk meteorites
113 common data must be examined to find simi-
114 larities. With the exception of O, Ti is the ele-
115 ment which has been most extensively studied
116 in a wide variety of early Solar System mate-
117 rials, including hibonite, grains FUN and nor-
118 mal CAIs and bulk meteorites. Though O has
119 been more extensively studied, it is not suit-
120 able for this purpose for two reasons. Firstly,
121 the origins of O isotope variations are debated
122 and may have non-unique sources (Thiemens
123 1999; Lyons and Young 2005; Lyons et al.
124 2009). Moreover, there has clearly been sub-
125 sequent mixing between these different Solar
126 System reservoirs. Secondly, O only has three
127 isotopes, so only one mass-independent ratio,
128 which limits the scope for analysing sources.
129 For these reasons we focus on Ti.

130 The first studies of Ti were in the FUN
131 CAIs by the ANU and Caltech groups (Hey-
132 degger et al. 1979; Niemeyer and Lugmair
133 1980). These rare inclusions exhibited large
134 anomalies in Ti, ± 40 ‰ on ^{50}Ti , where ‰
135 is the ϵ unit defined as the parts per ten thou-
136 sand difference to a terrestrial standard. Sev-
137 eral other elements show similar variation, for
138 example 150 ‰ on ^{48}Ca (Lee et al. 1978) and
139 290 ‰ on ^{58}Fe (Völkening and Papanastas-
140 siou 1989). Interestingly, these large anoma-
141 lies on ^{58}Fe are unique in the Solar System as
142 no other objects have been found to show iron
143 isotopic variation (e.g. Dauphas et al. 2008).
144 Even larger anomalies were found in the hi-
145 bonite grains from CM chondrites (e.g. Fa-
146 hey et al. 1985), see above. Smaller, but still
147 significant, Ti isotope anomalies were subse-
148 quently observed in the normal population of
149 CAIs (e.g. Niemeyer and Lugmair 1981) and
150 in bulk meteorites (e.g. Leya et al. 2008).

151 In general, the early studies of Ti isotopes
152 in chondritic inclusions and hibonites grains
153 showed large anomalies on the neutron rich
154 isotope ^{50}Ti and smaller anomalies on the
155 other isotopes. However, no coherent corre-
156 lations have been observed in the Ti isotope
157 compositions from these samples. These stud-
158 ies were focused on examining the input of
159 neutron-rich isotopes to the early Solar Sys-
160 tem, principally because of the early identifi-
161 cation of anomalies on ^{48}Ca in FUN CAIs (Lee
162 et al. 1978), which led to the almost universal
163 use of normalization to $^{46}\text{Ti}/^{48}\text{Ti}$ for the mass-
164 dependent fraction correction in early studies.
165 Fahey et al. (1987a) examined the number of
166 sources required to produce the variation in
167 Ti isotope compositions by fitting the data to
168 a plane with a least squares algorithm simi-
169 lar to the more conventional York regression
170 for 2-d correlation. Fahey et al. (1987a) con-
171 cluded that at least 4, possibly more, sources
172 were needed to reproduce the observed com-
173 positions.

174 In contrast, the more recent, high preci-
175 sion data for bulk meteorites show a strik-
176 ing correlation between $\epsilon^{50}\text{Ti}_{47}$ and $\epsilon^{46}\text{Ti}_{49}$
177 where the subscripts denote the normalizing
178 ratio, see section 2.1 below (Leya et al. 2008,
179 2009; Trinquier et al. 2009; Zhang et al. 2011,
180 2012). This correlation was shown by Qin
181 et al. (2011) and Steele et al. (2012) to be con-
182 sistent with input from the O/Ne zone of a
183 type II supernova (SN II). Mass-independent
184 isotopic variation between different bulk me-
185 teorite groups likely represents much more
186 anomalous Ti by mass than the much larger
187 anomalies seen in the hibonite grains.

188 One obvious hypothesis for the origin of
189 Ti isotope variation in the Solar System is
190 that one of the sources present in the hibonite
191 grains is also the source represented by the
192 variation observed in the bulk meteorites. Re-
193 lationships between hibonites and other me-
194 teorite groups were investigated by previous
195 studies, for example Fahey et al. (1987a).

196 Since these early investigations more higher
197 quality data have been reported, especially in
198 the bulk meteorite populations, therefore this
199 subject is worth revisiting. The aim of this
200 paper is to test the hypothesis that one of the
201 sources present in the compositions of the hi-
202 bonites and FUN CAIs is the same source ob-
203 served in the bulk meteorites. This will en-
204 able us to examine the reasons why multiple
205 sources are observed in the meteoritic inclu-
206 sions (hibonites and FUN CAIs) while only
207 one appears to be sampled by the bulk mete-
208 orites.

209 **2. Methods**

210 **2.1. Renormalization**

211 Mass-independent isotope anomalies re-
212 quire normalisation for mass-dependent frac-
213 tionation. This is normally achieved using one
214 isotope ratio to calculate and remove the ex-
215 pected fractionation on the other ratios, a pro-
216 cess termed ‘internally normalising’. See Rus-
217 sell et al. (1978), McCulloch and Wasserburg
218 (1978) and Young et al. (2002) for discussions
219 of fractionation corrections and the choice of
220 fractionation ‘laws’. Many of the hibonite
221 data were original presented normalized to
222 $^{46}\text{Ti}/^{48}\text{Ti}$ by the exponential law. These data
223 have been renormalized to $^{47}\text{Ti}/^{49}\text{Ti}$ to make
224 them comparable with the more recent bulk
225 data for which $\epsilon^{50}\text{Ti}_{49}$ and $\epsilon^{46}\text{Ti}_{49}$ anomalies
226 have been observed. It is important for the
227 Independent Component Analysis (ICA) (see
228 below) that appropriate uncertainties prop-
229 agated through the normalization. To en-
230 sure renormalized uncertainties are appropri-
231 ate they have been modelled using a Monte
232 Carlo simulation by varying the data around
233 their original uncertainties and propagating
234 this through the full renormalization. This
235 was repeated 10000 times to fully map the
236 parameter space and the renormalized uncer-
237 tainty is then the standard deviation (s.d.) of
238 these repeats. The code used for the renor-
239 malization was written using R (R Core Team

240 2013) and is available on request. The renor-
241 malized data are given an appendix table and
242 are plotted in figure 1.

243 **2.2. Independent Component Analysis**

244 Independent Component Analysis is a
245 method of blind source separation by which a
246 multivariate dataset can be deconvolved into
247 independent subcomponents (Comon 1994).
248 That is, a dataset, in this case Ti isotope
249 data from hibonites, can be examined for the
250 sources which mixed to produce the observed
251 variation. Independent Component Analysis
252 is related to the more commonly used princi-
253 pal component analysis (PCA) but differs in
254 that the components identified do not have to
255 be orthogonal, thus making it a more flexible
256 tool. Independent Component Analysis relies
257 on two assumptions, firstly that the sources
258 are statistically independent from each other,
259 and secondly that mixing between sources is
260 linear. Independent Component Analysis al-
261 gorithms work by finding solutions which min-
262 imise the shared information between compo-
263 nents and maximising the non-Gaussianity of
264 the individual components. The ICA algo-
265 rithm returns a component, or vector, along
266 which a significant proportion of the varia-
267 tion observed in the dataset as a whole can
268 be described. Where significant variation re-
269 mains, more components can be added until
270 the dataset can be fully described. The com-
271 ponents describe a slope which is a mixing
272 line to a composition which represents one of
273 the sources required to produce the variation
274 in the data. These vectors, or components,
275 may share a source at one end, or all sources
276 may be independent, so there must be at least
277 $n+1$ sources where n is the number of compo-
278 nents. Therefore, three vectors may represent
279 between 4 and 6 isotopic source compositions.
280 To describe the vector, the ICA reports a point
281 and the vector is the slope to the origin in n
282 dimensions. These points maybe in either pos-
283 itive or negative fields because the ICA only
284 determines the vector and the sign is assigned

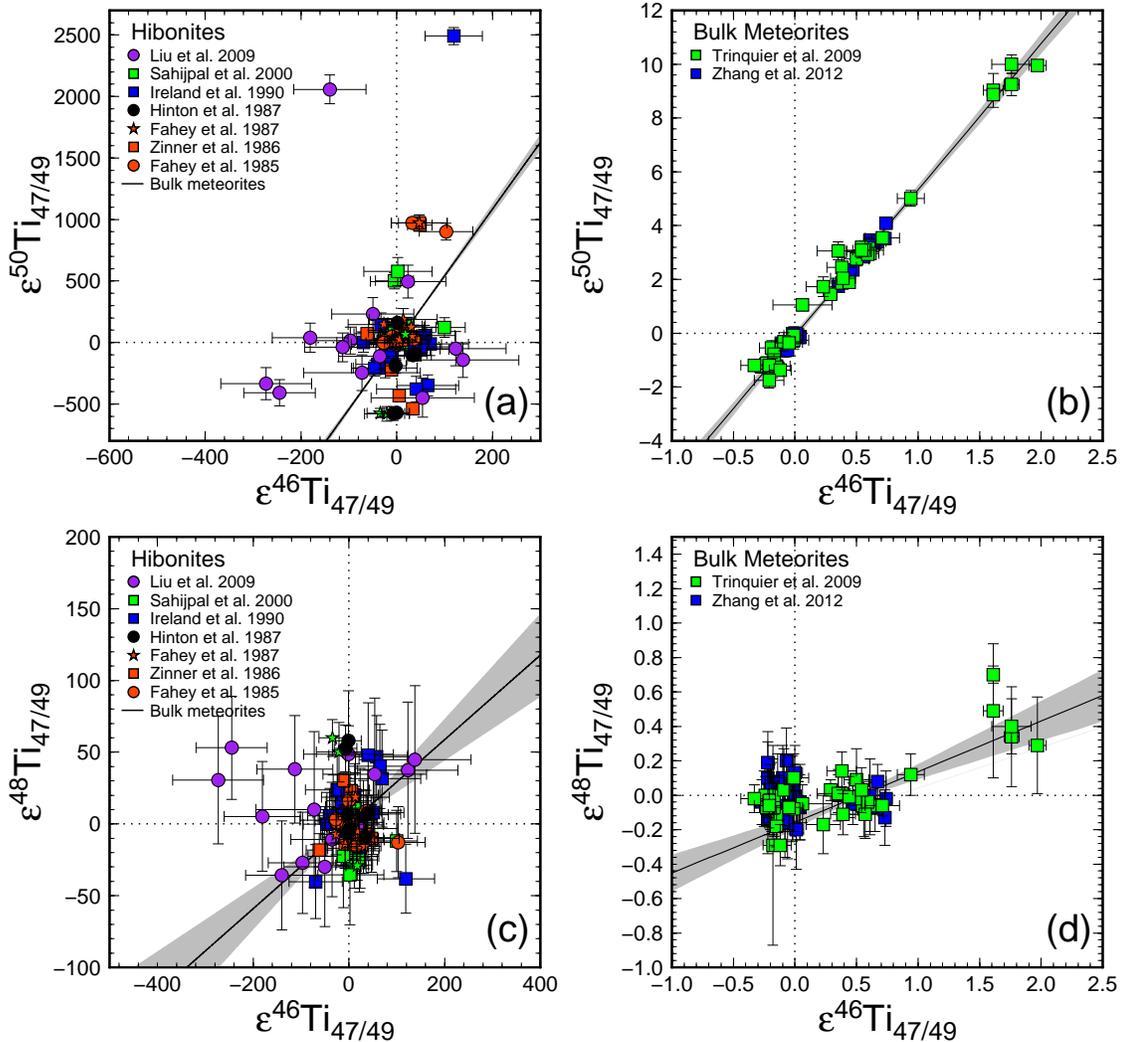


Fig. 1.— Figures showing previous Ti isotope compositions for hibonites (a) and (c) (from Fahey et al. 1985; Zinner et al. 1986; Fahey et al. 1987a; Hinton et al. 1987; Ireland 1990; Sahijpal et al. 2000; Liu et al. 2009), bulk meteorites and normal CAIs (b) and (d) (from Trinquier et al. 2009; Zhang et al. 2011, 2012). Also shown on both pairs of plots are extrapolations, with error envelopes, of the slopes from bulk meteorites and normal CAIs determined by York Regression (York 1969; Mahon 1996).

285 randomly. For a full description of ICA see
 286 Hyvärinen and Oja (2000).

287 There have been two previous studies in
 288 geochemistry and cosmochemistry which have
 289 used ICA for investigating source relation-
 290 ships. These looked at the distribution of
 291 mantle sources from the isotopic compositions
 292 of MORBs and OIBs (Iwamori and Albarède
 293 2008) and the mixing relations in the HED

294 suite of meteorites by examining major ele-
 295 ment abundances (Usui and Iwamori 2013). It
 296 is not clear that the requirement for straight
 297 line mixing is satisfied in the study of Iwamori
 298 and Albarède (2008) as they include isotope
 299 data from several different elements. Where
 300 different elements are used mixing will only
 301 approximate straight line mixing if the denomi-
 302 nator isotopes are of the same concentration.

303 It is important to note that in this study mix-
304 ing is linear in Ti isotope space as all the ratios
305 use the same denominator isotope.

306 We have used the FastICA algorithm of Hy-
307 varinen (1999); Hyvärinen and Oja (2000) im-
308 plemented in an R (R Core Team 2013) script
309 by Marchini et al. (2013) to determine the in-
310 dependent components of Ti isotope data. A
311 limitation of the traditional ICA is that un-
312 certainties are not taken into account. This
313 is because for the situations in which it has
314 normally been used, the noise on the data
315 does not come from measurement uncertain-
316 ties, rather from real variation in the data.

317 We have extended the of traditional ICA
318 by combining it with a Monte Carlo simula-
319 tion of the uncertainties, or bootstrap (Efron
320 1981), of the data in order to gain a statisti-
321 cal measure of the uncertainty of the indepen-
322 dent components determined by the FastICA
323 algorithm. The isotopic data were varied ran-
324 domly around their averages with a Gaussian
325 distribution and an ICA performed for each
326 iteration, this was repeated 20000 times to
327 achieve a robust measure of the uncertainty.
328 We have used clustering algorithms to sort
329 the data into the individual components (Fritz
330 et al. 2012; Cuesta-Albertos et al. 1997). Dur-
331 ing this process 1 % of the models were dis-
332 carded (e.g. tclust $\alpha = 0.01$) because they did
333 not fit into any of the clusters. This loss of
334 data is taken into account in the eventual un-
335 certainty. The clustering algorithms were op-
336 timised to use equal weights to ensure that the
337 clusters contained equal numbers of points.
338 The R scripts used to perform the ICA are
339 available on request.

340 The results of these iterations produce a
341 number of vectors which describe a mixing line
342 towards the source and are illustrated by the
343 distribution of points in figure 2. The vectors
344 form clusters, the number of which is twice
345 the number of components used because the
346 vector may be either positive or negative from
347 the origin; each pair of clusters describes the

348 distribution of one component. The variation
349 within a cluster of vectors describes the uncer-
350 tainty in the component and includes contri-
351 butions from the analytical uncertainty of the
352 data and the uncertainty inherent in the ICA
353 algorithm. This gives a robust measure of the
354 uncertainty of the components determined by
355 the ICA and allows components from different
356 sample populations to be compared.

357 2.3. Data used for the ICA

358 We have performed the ICA on both the hi-
359 bonite and bulk meteorite datasets. We have
360 used hibonite data for SHIBs, PLACs and
361 BAGs from as many studies possible includ-
362 ing measurements from: Fahey et al. (1985);
363 Zinner et al. (1986); Fahey et al. (1987a); Hin-
364 ton et al. (1987); Ireland (1990); Sahijpal et al.
365 (2000); Liu et al. (2009). The data from these
366 studies were collected over 25 years on a vari-
367 ety of of ion probes, however, they all provide
368 measurements from the standard Madagascar
369 hibonite which are in agreement warranting
370 their inclusion in this metadataset.

371 Several recent studies have measured the Ti
372 isotope compositions of bulk meteorites and
373 normal CAIs (Leya et al. 2008, 2009; Trin-
374 quier et al. 2009; Zhang et al. 2011, 2012).
375 However, not all of these studies are appro-
376 priate to include as a metadataset. The re-
377 cent Ti studies of bulk meteorites and nor-
378 mal CAIs all show broadly consistent results
379 finding correlated anomalies on $\epsilon^{50}\text{Ti}_{49}$ and
380 $\epsilon^{46}\text{Ti}_{49}$, with very little variation on $\epsilon^{48}\text{Ti}_{49}$.
381 Trinquier et al. (2009); Zhang et al. (2011,
382 2012) report very high precision Ti isotope
383 compositions for a wide range of bulk mete-
384 orites, both chondrites and achondrites. In
385 addition Trinquier et al. (2009) report Ti iso-
386 tope compositions for the normal population
387 of CAIs. Though the earlier studies of Leya
388 et al. (2008, 2009) show broadly similar re-
389 sults to the more recent studies, they are of
390 lower precision and so would hamper our abil-
391 ity to determine the slopes of mixing. Leya

et al. (2008) also raise concerns over large non-terrestrial blanks introduced to some of their samples during digestion in Teflon bombs which may affect the accuracy of some data. Therefore we do not include the results of Leya et al. (2008, 2009) in our meta-dataset. Correlations produced by the studies of Trinquier et al. (2009); Zhang et al. (2011, 2012) are very consistent. In $\epsilon^{46}\text{Ti}_{49}$ vs. $\epsilon^{50}\text{Ti}_{49}$ they produce slopes of 5.48 ± 0.27 and 5.23 ± 0.22 for Trinquier et al. (2009) and Zhang et al. (2011, 2012) respectively. A “new” York regression (Mahon 1996) based on this combined dataset yields 5.37 ± 0.15 .

2.4. ICA Data Presentation and Mixing angles

In order to compare the sources of different sample populations, the mixing vectors of each cluster, and the variation in these vectors within each cluster, must be compared. The data are presented initially by simply plotting the points returned by the ICA. In order that the components obtained from different sample populations maybe compared on the same diagram, the data for each point, in each individual dimension, has been normalized to the sum of the standard deviations of each dimension. This was achieved using

$$\gamma(^j\text{Ti}_{49}^{47}) = \frac{i(^j\text{Ti}_{49}^{47})}{\sigma(^j\text{Ti}_{49}^{47}) + \sigma(^k\text{Ti}_{49}^{47}) + \sigma(^l\text{Ti}_{49}^{47})}, \quad (1)$$

where $\gamma(^j\text{Ti}_{49}^{47})$ is the renormalized component composition for the i th iteration of the ICA in the dimension of $(^j\text{Ti}_{49}^{47})$ (where isotope ^jTi which was normalized for mass-dependent fractionation using the $^{47}\text{Ti}/^{49}\text{Ti}$ ratio), j , k and l represent the isotopes of Ti, $i(^j\text{Ti}_{49}^{47})$ is the raw composition of the i th iteration of the ICA in the dimension of $(^j\text{Ti}_{49}^{47})$ and $\sigma(^j\text{Ti}_{49}^{47})$, $\sigma(^k\text{Ti}_{49}^{47})$ and $\sigma(^l\text{Ti}_{49}^{47})$ is the standard deviation over all iterations of the ICA in the dimensions of $(^j\text{Ti}_{49}^{47})$, $(^k\text{Ti}_{49}^{47})$ and $(^l\text{Ti}_{49}^{47})$, respectively. Thus the total variation of each

sample population can be presented in the same, albeit artificial range. Importantly though, the angular information is retained, so the slopes, or angles, of the components hold through this normalization.

In order to quantitatively compare the components obtained for different sample populations the vectors must be obtained from the slopes. Previous studies have used the approach of comparing slopes of mixing in order to compare data sets (e.g. Dauphas et al. 2004; Steele et al. 2012). In these previous cases the dataset being compared were bulk meteorite data and modelled nucleosynthetic data for different astrophysical environments, not different populations of measurements but the concepts are the same. The use of slopes has some associated problems for comparison of different datasets. The major problem with slopes is that when they approach the vertical or horizontal the slope loses linearity and tends to zero or infinity. This reduces the ability to graphically resolve the difference between a slopes multiple slopes of different magnitudes. More seriously, however, if the uncertainty of a slope crosses either the vertical axis it will artificially encompass both zero and infinity making statistical comparison between different more complicated.

The issue of infinities may be simplified by using the angles of mixing (where $\theta = \arctan(m) \cdot 180/\pi$) rather than slopes of mixing (m). Angles of mixing have the advantage that they are linear and contain only one singularity (360 to 0, where 0 is the positive limb of the x axis) compared to the four of slopes. This means that for comparison of two mixing lines (and possibly more) a position of the singularity may be chosen such none of the uncertainties are affected. In this study mixing lines are presented as angles of mixing and not slopes. This method could be useful in other areas, for example the comparison of mixing of nucleosynthetic environments in to the Solar System.

478 **3. Results of the ICA**

479 The results of the ICA are presented in ta-
480 ble 1 and figures 2 (a), (b), (c), (d) and 3.

	Average	-2 s.d.	+2 s.d.
$\epsilon^{50}\text{Ti}_{49}/\epsilon^{46}\text{Ti}_{49}$			
Bulk {a}	79.784	0.323	0.347
Bulk {b}	79.467	0.519	0.442
Hibonite {1}	88.673	1.126	0.962
Hibonite {2}	24.342	56.666	31.094
Hibonite {3}	101.968	18.662	26.189
$\epsilon^{50}\text{Ti}_{49}/\epsilon^{48}\text{Ti}_{49}$			
Bulk {a}	86.069	2.059	1.146
Bulk {b}	92.264	1.492	4.145
Hibonite {1}	91.040	0.459	0.463
Hibonite {2}	100.075	60.164	43.414
Hibonite {3}	106.891	6.463	18.400

Table 1: Results of the ICA on the hibonite and bulk meteorite populations. The results are presented as the angle ($\theta^\circ = \arctan(m) \cdot 180/\pi$, where m is the slope) of mixing vectors to the returned components. The uncertainties are 95 % confidence intervals derived from the spread of the ICA components returned by the bootstrap. Note the errors are the deviation in angle and are not symmetrical.

481 **3.1. ICA components**

482 Figures 2 (a), (b) and (c) show the ICA
483 components presented as $\gamma(^j\text{Ti}_{49})$ normalized
484 data show the hibonite and bulk meteorite
485 data.

486 As expected the results of the ICA for the
487 bulk meteorite data, shown in red colours in
488 figure 2(a), show a very clear correlation be-
489 tween $\gamma(^{50}\text{Ti}_{49})$ and $\gamma(^{46}\text{Ti}_{49})$. In the figures
490 2 (b) and (c), which include the other dimen-
491 sion $\gamma(^{48}\text{Ti}_{49})$, the components are not corre-
492 lated. The divergence from a single correla-
493 tion, and the presence of well defined compo-
494 nents, shows that variation in $\epsilon^{48}\text{Ti}_{49}$ is ob-
495 served and the mixing vectors of the sources
496 can be examined by the ICA.

497 **3.2. ICA angles**

498 The single, tight, correlation observed be-
499 tween $\epsilon^{46}\text{Ti}_{49}$ and $\epsilon^{50}\text{Ti}_{49}$ in the bulk mete-
500 orites can be used as a test of the accuracy of
501 the ICA method for determining the mixing
502 slopes of the sources and also the uncertainty
503 estimation by our bootstrap method, see fig-
504 ure 3. Regardless of any variation in $\epsilon^{48}\text{Ti}_{49}$, a
505 projection of the plane present in the bulk me-
506 teorites on to the $\epsilon^{50}\text{Ti}_{49}$ vs. $\epsilon^{46}\text{Ti}_{49}$ axis will
507 be a single correlation. This holds for line fit-
508 ting algorithms, such as the least squares York
509 regression, and ICA. Therefore, the slopes and
510 errors obtained by York regression and ICA
511 can be compared. This offers a demonstra-
512 tion of the accuracy of the angle obtained by
513 the ICA and the precision of this method re-
514 lative to the uncertainty of the original data.
515 A “new” York regression on the combined
516 dataset of Trinquier et al. (2009) and Zhang
517 et al. (2012) yields a slope of 5.37 ± 0.15 , the
518 angle obtained from this slope is $79.45 \pm 0.29^\circ$.
519 This value is very close to the average value ob-
520 tained by the ICA of $79.63 \pm 0.42^\circ$, see figure
521 3. Moreover, the uncertainties are also very
522 similar, with the uncertainty slightly larger as
523 expected as it includes a potential contribu-
524 tion from the ICA algorithm. This displays
525 the ability of ICA to return accurate and pre-
526 cise vectors of source mixing.

527 **4. Discussion**

528 The results of the ICA show clear differ-
529 ences between character of the sources repre-
530 sented in the hibonite population versus the
531 bulk meteorites and the normal CAIs, see fig-
532 ure 2. The most obvious explanation for this
533 would be that the sources represented by the
534 hibonite and bulk meteorites populations are
535 different. The implications of this explanation
536 are discussed in detail in section 4.3. How-
537 ever, there are several other possible reasons
538 why the components represented by these two
539 populations might be different. Firstly, the
540 sources represented by the bulk meteorites and

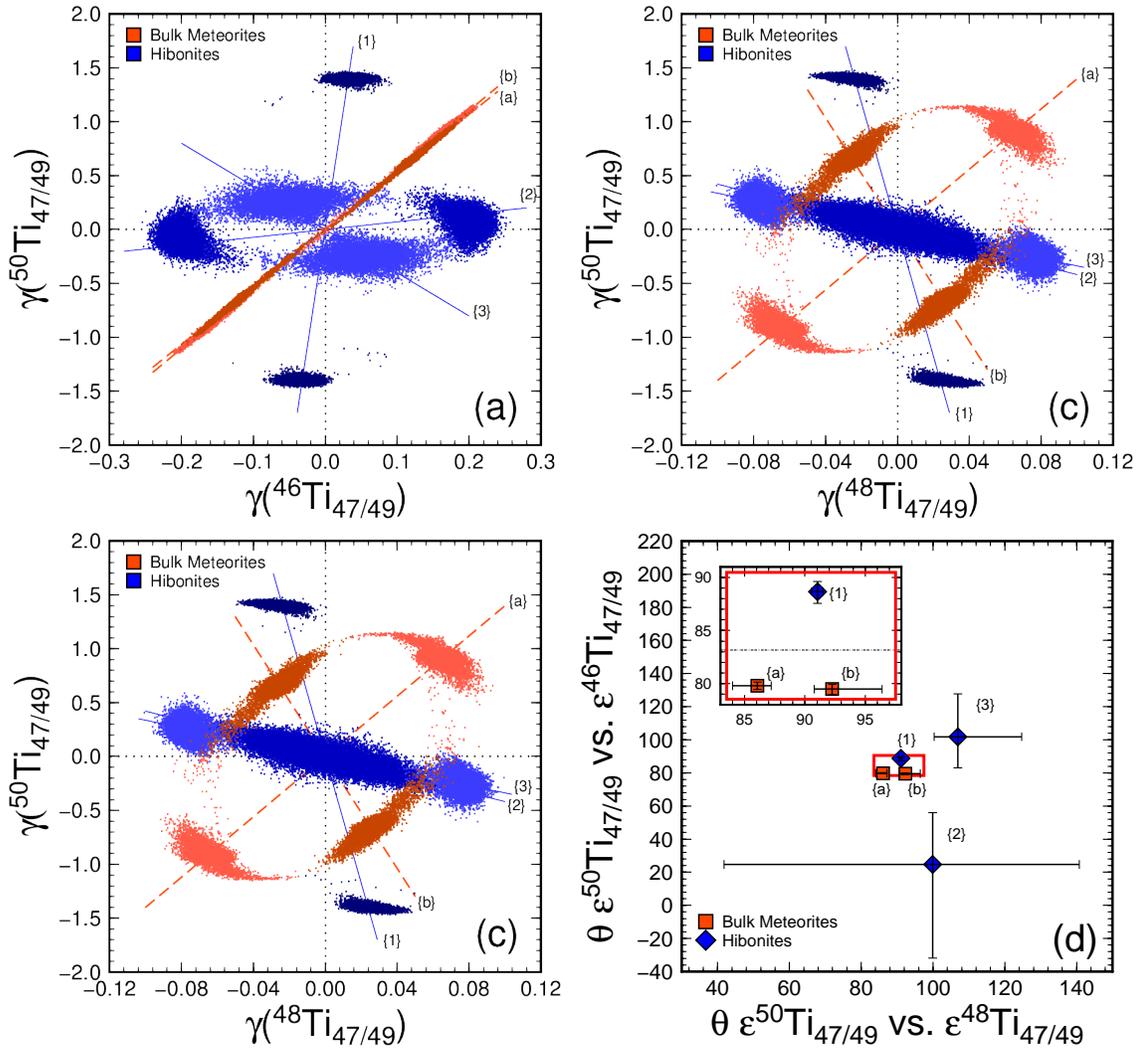


Fig. 2.— Figures showing the results of the ICA bootstrap for both the hibanite and bulk meteorite data. Plots (a), (b) and (c) show all the raw compositions of the ICA bootstrap iterations with the exception, in the case of the hibanites, of 1 % of components which are so far away from an average component composition that they cannot be grouped. The individual components are highlighted by different shades and labelled with {1}, {2}, {3}, {a} and {b}. Plot (d) shows the slopes of mixing of the different components and their uncertainties. By plotting $\theta \epsilon(^{50}\text{Ti}_{47/49})$ vs. $\epsilon(^{46}\text{Ti}_{47/49})$ against $\theta \epsilon(^{50}\text{Ti}_{47/49})$ vs. $\epsilon(^{48}\text{Ti}_{47/49})$ we using all Ti isotope dimensions. These plots show that the dominant sources present in the bulk meteorite data are not represented in the hibanite data as all hibanite component are resolved from both bulk meteorite components in both dimensions. (The only exception is the hibanite component {3} in $\theta \epsilon(^{50}\text{Ti}_{47/49})$ vs. $\epsilon(^{48}\text{Ti}_{47/49})$ which is very poorly defined and has essentially 100 % uncertainties.) The errors are the 95 % confidence intervals from the distribution of the bootstrapped ICA components. The dashed line in the inset figure is the lower error bound of the hibanite component {3}.

541 normal CAIs might be mixtures of the hi-
 542 bonite sources. Secondly, the hibanite popula-
 543 tion may represent more sources than the 4 to
 544 6 that can be examined by a three dimensional

545 ICA. Lastly, a combination of these possibil-
 546 ities occurred and the 3 hibanite components
 547 represent more than 6 sources and that the
 548 bulk meteorites compositions are mixtures be-

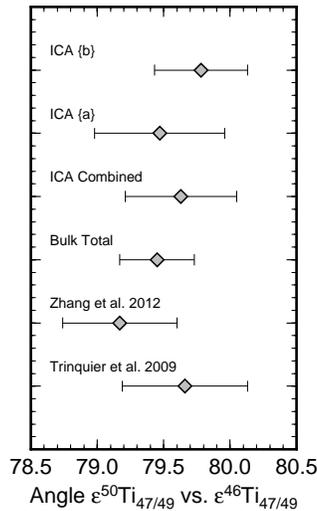


Fig. 3.— Figures a comparison between the estimates of angle of the bulk meteorite Ti isotope data in $\epsilon^{50}\text{Ti}_{47/49}$ vs. $\epsilon^{46}\text{Ti}_{47/49}$ estimated by York regression and ICA. The results of the two estimations are very similar which displays the power of ICA to estimate the slopes of correlations within data. ICA{a} and ICA{b} are the two components which are determined for the bulk meteorite population.

549 tween some of these sources. The likelihood
 550 and implications of these more complex sce-
 551 narios are discussed below.

552 4.1. Number of Sources

553 The simplest explanation of the isotopic
 554 variation observed by the ICA is that the hi-
 555 bonites represent between 4 and 6 sources and
 556 the bulk meteorites and normal CAIs repre-
 557 sent between 3 and 4. These are the sim-
 558 plest explanations because they represent the
 559 fewest number of anomalous sources in the So-
 560 lar System that can entirely explain the varia-
 561 tion we observe. However, more complex mod-
 562 els which involve more sources or mixing of
 563 sources are possible.

564 4.1.1. Number of sources represented by the 565 hibonites

566 The hibonite system is under-constrained
 567 in that there are at least as many required
 568 components as dimensions. It is very diffi-
 569 cult to examine how many components are
 570 required by an under-constrained system, and
 571 impossible to conclusively test. However,
 572 there are certain arguments and lines of evi-
 573 dence that suggest that the hibonite data, at
 574 the current level of precision, are only show-
 575 ing evidence for three components. Firstly,
 576 four sources can describe any three dimen-
 577 sional data array; akin to a point lying on a
 578 mixing line between two sources, there is no
 579 mathematical requirement for more sources
 580 to explain the spread of the data. Secondly,
 581 the components we have identified closely re-
 582 semble the data with the largest variation
 583 is in $\gamma(^{50}\text{Ti}_{47/49})$ followed by $\gamma(^{46}\text{Ti}_{47/49})$ then
 584 $\gamma(^{48}\text{Ti}_{47/49})$. Moreover, the angles of these com-
 585 ponents fit with the angles of the original data
 586 such that the errors all but a few data overlap
 587 with the mixing vectors of the components,
 588 see figure 4 and further discussion in section
 589 4.2.2. This demonstrates that the components
 590 are fully explaining the data without simply
 591 describing a cube around them. The most
 592 striking feature is that most data only require
 593 input from one component. Most hibonite
 594 data show evidence of binary mixing from one
 595 of three highly anomalous sources. Thirdly,
 596 the ICA tries to explain the variation in a
 597 dataset by making the sources as statistically
 598 independent as possible. Therefore, we can
 599 examine the possibility of unrepresented com-
 600 ponents by looking at the distribution of the
 601 three components we can identify.

602 As shown in figure 2, the vector of compo-
 603 nent {1} is very different from the other two,
 604 this makes it very statistically resolved as this
 605 is a function of the angular separation and the
 606 distribution of point along the component vec-
 607 tor. The angles of components {2} and {3} are
 608 much more similar. As the ICA hierarchically

609 tries to assign components based on statisti-
610 cal significance, a 4th, unresolved component,
611 must be at least as similar to one of the other
612 components as {2} and {3} are to each other
613 or else it would have been chosen. If a 4th
614 component were to be present and unrepres-
615 ented it would most likely lie in the plane of
616 the {2} and {3} components as this is where
617 several of the data which are not within error
618 of a component vector reside. It is possible
619 that if such a component exists it could be
620 represented by the SHIB hibonite population,
621 which are currently largely within error of ter-
622 restrial values.

623 A higher precision Ti isotope study of hi-
624 bonite would significantly help resolve issues
625 surrounding the number of sources. However,
626 as the data stand with the PLAC population
627 essentially showing all the variation, for the
628 reasons described above, the most likely expla-
629 nation is three components and four sources.
630 We discuss the compositions of these sources
631 in more detail below, see section 4.2.

632 4.1.2. *Number of sources represented by the* 633 *bulk meteorites*

634 The variation observed in the bulk mete-
635 orites is well described by two components, or
636 at least three sources. This finding is in agree-
637 ment with previous studies which show that
638 there must be more than two sources in the
639 bulk meteorite and normal CAI system (Trin-
640 quier et al. 2009; Williams et al. 2014). There
641 is a hint in the bulk meteorite and normal CAI
642 data that the third anomalous source is only
643 present in the CAIs. This is because a York
644 regression on the bulk meteorite population,
645 without the normal CAI data, yield a slope
646 of zero in $\epsilon^{50}\text{Ti}_{47/49}$ vs. $\epsilon^{48}\text{Ti}_{47/49}$. Moreover, an
647 ICA on the bulk meteorites alone shows no
648 variation in the $\gamma(^{48}\text{Ti}_{47/49})$ dimension. This
649 is supported by Williams et al. (2014) who
650 have reported evidence for a third source in
651 chondrules and CAIs in the absolute Ti iso-
652 tope compositions. Absolute ratios include

653 both natural mass-dependent fractionation
654 and mass-independent anomalies, thus show
655 the true location and magnitude of the anom-
656 alies. Absolute ratios have previously been re-
657 ported for Ti in CAIs (Niederer et al. 1985)
658 and Ni in bulk meteorites (Steele et al. 2012).
659 Interestingly, Trinquier et al. (2009) found ev-
660 idence for a third source in leachates from CI
661 CC which depart from the single correlation
662 of the bulk meteorites. However, the carrier
663 phase of this variation is likely much more
664 homogeneously distributed between different
665 chondrite parent bodies and so is not observed
666 in bulk dissolutions of meteorites as has been
667 observed for other elements (e.g. Schön-
668 bächler et al. 2005; Trinquier et al. 2008). The varia-
669 tion from the leachates, however, does super-
670 ficially look more like the variation seen in the
671 hibonites, with significantly more variation in
672 $\epsilon^{50}\text{Ti}_{47/49}$ creating a near vertical trend, see fig
673 3 of Trinquier et al. (2009).

674 4.1.3. *Bulk meteorites as mixtures of hi-* 675 *bonite sources*

676 One further possibility is that the compo-
677 nents observed in the bulk meteorites and nor-
678 mal CAIs are mixtures of the sources repre-
679 sented by the hibonites. It is important to
680 note that the bulk meteorite population is
681 over-described as it is fully described by fewer
682 components than dimensions. The maximum
683 number of sources is 4 and the most likely is 3.
684 The 3 most likely sources are an average, old
685 molecular cloud population and two anom-
686 alous sources. The identities of the sources are
687 discussed in more detail in section 4.2. The
688 two anomalous sources could both be hibonite
689 sources or one could be an independent presol-
690 ar carrier phase. This scenario is possibly
691 somewhat less likely because it requires an ex-
692 tra stage of discrete mixing between the for-
693 mation of hibonites and the formation of the
694 normal CAIs and bulk meteorites.

4.2. Source Compositions

Firstly, it is important to note that the ICA returns a vector along which the source composition lies. Therefore, it is not possible to say exactly what the source composition is, just the angle of its mixing line. However, by using the angles of the mixing lines we can identify compatible nucleosynthetic sources in the same way that York regressions on data have been used to examine nucleosynthetic sources in the past (e.g. Dauphas et al. 2004; Steele et al. 2012).

4.2.1. Source compositions of bulk meteorite components

The two components which the bulk meteorites and normal CAIs require are identical within error in $\gamma(^{50}\text{Ti}_{49})$ vs. $\gamma(^{46}\text{Ti}_{49})$ and have angles identical to a York regression in the same dimensions on the original data of Trinquier et al. (2009), see figure 2. This suggests binary mixing in $\epsilon^{46}\text{Ti}_{49}$ and $\epsilon^{50}\text{Ti}_{49}$ to produce this line. This source has been previously suggested to be input from the O/Ne zone of an SN II (Qin et al. 2011; Steele et al. 2012). The compositions produced by the O/Ne zones of SN II vary with the mass of the pre supernova star. Few of the models of Rauscher et al. (2002) match exactly the slopes required to explain Ti isotope variation in the Solar System, they are however, all close. Titanium-50 and ^{46}Ti are thought to be dominantly produced in different nucleosynthetic environments. Therefore the finding that by including anomalies on the normalising isotopes the correct slopes of mixing may be produced is itself important. An alternative hypothesis to explain the meteorite correlation is simultaneous mixing of two distinct nucleosynthetic sources, one with $\epsilon^{46}\text{Ti}_{49}$ anomalies and one with $\epsilon^{50}\text{Ti}_{49}$ anomalies, within the Solar System due to some common process (Trinquier et al. 2009). The ICA analysis is consistent with either interpretation.

In the other Ti axis, $\gamma(^{48}\text{Ti}_{49})$, however,

there is slight variation between the two components. Note that the scale of the $\gamma(^{48}\text{Ti}_{49})$ axis on figure 2(c) significantly magnifies the apparent difference and the two components are in fact only divergent by 6 degrees. The component {a} shows a positive correlation with an angle of $86.1^{+1.2}_{-2.1}$, while component {b} is, in fact, almost vertical, so showing almost no influence on ^{48}Ti , with an angle of $92.3^{+4.7}_{-1.5}$. The positive correlation in component {a} is just visible in the original meteorite and normal CAI data, see figure 1(d). However, the apparent correlation is only present in the CAI data. This may suggest that the CAIs are sampling a source which imparts a very slight anomaly in $\epsilon^{48}\text{Ti}_{49}$ and is not present in the bulk meteorite population. This inference may be supported by recent absolute Ti isotope measurements of Williams et al. (2014) who found evidence for correlated ^{46}Ti , ^{47}Ti and ^{50}Ti anomalies in the bulk meteorites while the CAIs plotted off this trend in ^{47}Ti . It is not clear if this can explain the deviation in $\epsilon^{48}\text{Ti}_{49}$ observed in the previous measurements, but is supporting evidence that the CAIs may be sampling a subtly different set of sources. An ICA performed on the bulk meteorites alone yields one component identical in angle in $\gamma(^{50}\text{Ti}_{49})$ vs. $\gamma(^{46}\text{Ti}_{49})$ to the two component ICA and with zero variation in $\gamma(^{48}\text{Ti}_{49})$ which may support this hypothesis further. The major conclusions of this study are not affected by this debate because the variation in $\gamma(^{50}\text{Ti}_{49})$ vs. $\gamma(^{46}\text{Ti}_{49})$ is not affected and the variation in $\gamma(^{48}\text{Ti}_{49})$ is very small and always resolved from the hibonite components. However, the hint that the CAIs, while dominated by the same anomalous sources as the bulk meteorites, may sample an additional anomalous source is very interesting. This finding could have significant implications for early Solar System mixing and warrants further study.

782 *4.2.2. Source compositions of the hibonite*
783 *components*

784 The nucleosynthetic origins of the hibonite
785 sources have been less well studied in a quan-
786 titative sense and the overall lack of preci-
787 sion in the components we have identified does
788 not makes this endeavour much easier. In-
789 terestingly, the errors of most of the hibonite
790 grains overlap with at least one of the identi-
791 fied components, for example in $\epsilon^{50}\text{Ti}_{49}^{47}$ vs.
792 $\epsilon^{46}\text{Ti}_{49}^{47}$ see figure 4. This suggests the hibonite
793 grains may mostly be exhibiting binary mixing
794 to one anomalous nucleosynthetic source.
795 In the rare cases of the remaining data which
796 lie a long way from any of the component vec-
797 tors, these compositions may be produced by
798 mixing two of the anomalous nucleosynthetic
799 sources. Mixing two sources is not an unex-
800 pected result as the ICA is designed to decon-
801 volve multivariate data. Even so, a finding
802 of binary mixing in the majority of the hi-
803 bonites presents a simplified case which may
804 offer more easily testable predictions for mix-
805 ing in other isotope systems.

806 As discussed above, the components of the
807 hibonites are not equally well defined. The
808 most precisely defined component is compo-
809 nent {1} with a angle of $88.7_{-1.1}^{+1.0}$ in $\gamma(^{50}\text{Ti}_{49}^{47})$
810 vs. $\gamma(^{46}\text{Ti}_{49}^{47})$ and $91.0_{-0.5}^{+0.5}$ in $\gamma(^{50}\text{Ti}_{49}^{47})$ vs.
811 $\gamma(^{48}\text{Ti}_{49}^{47})$. These angles may be well enough
812 defined to examine the nucleosynthetic signif-
813 icance, however, it is not clear where these
814 signatures may be produced.

815 As with previous studies (e.g. Steele et al.
816 2012) we are looking for a signature that de-
817 scribes the characteristic of a distinct nucle-
818 osynthetic environment. The most significant
819 finding is a feature which is consistent among
820 most, if not all, models. For example the
821 ^{58}Ni anomaly found in the Si/S zone of all
822 masses SN II (Steele et al. 2012). We have
823 compared the slopes of the hibonite compo-
824 nents to the same range of supernova mod-
825 els as previous studies: for type Ia super-
826 nova (SN Ia), Woosley (1997); Travaglio et al.

827 (2004); Iwamoto et al. (1999); Hashimoto
828 (1995); Maeda et al. (2010); Travaglio et al.
829 (2011); for SN II, Iwamoto et al. (1999);
830 Umeda and Nomoto (2002); Nomoto et al.
831 (1997); Rauscher et al. (2002); Hashimoto
832 (1995); for asymptotic giant branch star
833 (AGB), models were kindly provided by
834 Gallino and Davis (pers. comm. 2009); and
835 for individual shells of SN II we used mod-
836 els from Meyer et al. (1995) Rauscher et al.
837 (2002). Bulk homogenous SN II are not a
838 likely candidate, both because it is unrealis-
839 tic they would present a single homogenised
840 signature, and also this signature does not
841 produce the high $\epsilon^{50}\text{Ti}_{49}^{47}$ anomalies required.
842 Large ^{50}Ti excesses are produced in the inner
843 regions of SN II suggesting this as a possi-
844 ble candidate. However, these excesses are
845 accompanied with significant deficits in ^{47}Ti
846 which in the $^{47}\text{Ti}/^{49}\text{Ti}$ normalization acts to
847 reduce the apparent ^{50}Ti anomaly, and in-
848 crease the apparent ^{46}Ti anomaly, such that
849 the slope in $\epsilon^{50}\text{Ti}_{49}^{47}$ vs. $\epsilon^{46}\text{Ti}_{49}^{47}$ is always too
850 low. Finally, large excesses in ^{50}Ti , not ac-
851 companied by excesses of ^{47}Ti or ^{46}Ti , are
852 a general, if not universal, feature of SN Ia.
853 This suggestion is further supported by the
854 large positive ^{48}Ca anomalies which are cor-
855 related with ^{50}Ti anomalies (e.g. Zimmer et al.
856 1986). Large ^{48}Ca anomalies, in absence of
857 ^{46}Ca anomalies, are strong evidence for input
858 from an SN Ia (Meyer et al. 1996). This pos-
859 sibly makes SN Ia the most likely source for
860 the high ^{50}Ti component in the hibonites, but
861 this is by no means conclusive.

862 The other two components are not nearly
863 as well defined and no nucleosynthetic signif-
864 icance can be determined due to the large un-
865 certainties on their compositions. One of the
866 most intriguing aspects of the hibonite grains
867 is that they have both large positive and large
868 negative anomalies in for example, $\epsilon^{50}\text{Ti}_{49}^{47}$,
869 see figure 1. These components represent the
870 sources of hibonite grains which exhibit nega-
871 tive anomalies in $\epsilon^{50}\text{Ti}_{49}^{47}$ and $\epsilon^{46}\text{Ti}_{49}^{47}$. These
872 large positive and negative anomalies are dif-

873 ficult to reconcile with the very small range
 874 in bulk meteorites in the context binary mix-
 875 ing. However, in the context of binary mix-
 876 ing between multiple sources these anomalies
 877 are more easily understood. Broadly, compo-
 878 nent {2} constitutes low $\gamma(^{50}\text{Ti}_{47/49})$, slight
 879 $\gamma(^{46}\text{Ti}_{47/49})$ and high $\gamma(^{48}\text{Ti}_{47/49})$, while compo-
 880 nent {3} is made up of low $\gamma(^{50}\text{Ti}_{47/49})$, high
 881 $\gamma(^{46}\text{Ti}_{47/49})$ and low $\gamma(^{48}\text{Ti}_{47/49})$. These two compo-
 882 nents are not well defined enough to quan-
 883 titatively discuss the nucleosynthetic signifi-
 884 cance, however, qualitative assessments may
 885 be made. Component {2} is consistent with
 886 input from material from low mass (~ 13 - 15
 887 M_{\odot}) SN II models, while the sources of compo-
 888 nent {3} are consistent with several mid-
 889 mass (~ 25 - $30 M_{\odot}$) SN II (Hashimoto 1995;
 890 Umeda and Nomoto 2002). Even though the
 891 sources cannot be definitively identified, these
 892 components are still useful for discussing po-
 893 tential mixing environments within the Solar
 894 System as they are helpful for interpreting the
 895 hibonite data, see section 4.3

896 4.3. Implications for mixing and sources 897 within the Solar System

898 The ICA of the hibonite data returns the
 899 compositions of three components which most
 900 likely reflect mixing of four sources. Most
 901 of the data cluster around zero, which sug-
 902 gests that one of the sources is represented by
 903 this composition. Interestingly, this compo-
 904 sition is compatible with the composition of
 905 one of the bulk meteorite sources. This may
 906 be close in composition one of the end of the
 907 bulk meteorite correlation and could represent
 908 the bulk composition of the presolar molecular
 909 cloud, see section 4.3.1 below. That one of the
 910 sources of the two different sample sets may be
 911 the same is interesting and also may present
 912 a more likely scenario because it reduces the
 913 number of required sources. However, we must
 914 examine the likelihood that such a source may
 915 exist.

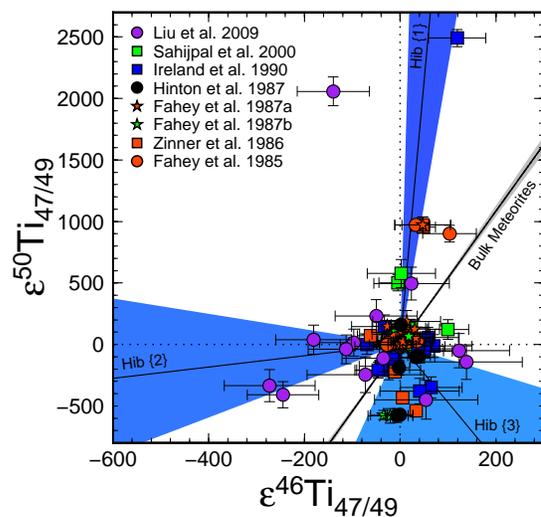


Fig. 4.— Figures showing the Ti isotope compositions for hibonite grains and the components determined from the ICA. The data, with only a few exceptions, are within error of one of the components. This suggests that there is no requirement for a further components. The compositions of few point that lie outside of error of all the components can be created by mixing two of the components.

916 4.3.1. Material in proto-planetary disks, 917 molecular clouds and the interstellar 918 medium

919 The identity of the material of this shared
 920 composition is likely the average of highly
 921 processed material from the parent molecular
 922 cloud. The journey of stellar condensates from
 923 stars and supernovae to proto-planetary disks
 924 is not a simple one. There are many stages
 925 of processing and mechanisms by which they
 926 may be altered and destroyed.

927 The Solar System, and proto-planetary
 928 disks in general, form from a mixture of
 929 material from many different generations of
 930 stars likely integrating material over billions
 931 of years of galactic evolution. Indeed, it has
 932 been estimated that the timescale for a grain
 933 from a stellar sources to traverse the interstel-
 934 lar medium (ISM) and be incorporated into a
 935 proto-planetary disk is on the order of a 1 Ga
 936 (Dwek and Scalo 1980; Jones and Nuth 2011).

937 However, due to physical processing and ir-
938 radiation in the interstellar medium it is an
939 oversimplification to assume that the material
940 in a molecular cloud contains the original iso-
941 topic signatures of stars integrated over 1 or
942 2 Ga.

943 Material in molecular clouds may have
944 experienced several cycles through the dif-
945 fuse interstellar medium into molecular clouds
946 without necessarily being incorporated into a
947 proto-planetary disk, or a star, to be repro-
948 cessed. Lifetimes of refractory grains have
949 been estimated at $4 \times 10^8 - 20 \times 10^8$ years de-
950 pending on how much time the grains spend in
951 molecular clouds where stars are more likely
952 to form (see, Barlow 1978; Jones 2004, for
953 discussions). Interestingly, Molster and Wa-
954 ters (2003) note that crystalline silicates are
955 only observed around supernovae and young
956 stars and proto-planetary disks - where they
957 are likely to be forming. This suggests from
958 an observational standpoint that fresh ma-
959 terial does not last long in the ISM. Car-
960 rez et al. (2002) performed laboratory exper-
961 iments to investigate the effects of He irra-
962 diation of olivine as an analogue to radia-
963 tion processing in interstellar medium finding
964 that both chemical and structural changes oc-
965 cur in olivine culminating in amorphitization.
966 While, from astronomical observations, Kem-
967 per et al. (2004) find that the timescale of
968 amorphitization in the ISM is on the order of
969 5-9 Ma.

970 Therefore, in most cases the lifetime of
971 grains will be less than the timescale from pro-
972 duction in supernova to processing in proto-
973 planetary disk. It follows, then, that many
974 grains will not survive their journey through
975 the interstellar medium to be incorporated
976 into a proto-planetary disk. Rather these
977 grains will become amorphitized and eventu-
978 ally destroyed. This material will remain in
979 the ISM and form new grains. Jones and Nuth
980 (2011) estimate that the majority (90-95 %) of
981 grains observed in the ISM would have formed

982 in *in situ* and so will not represent pure stellar
983 compositions. The action of this grain refor-
984 mation will be to homogenise the isotopic com-
985 positions of the many generations and types
986 of stars that have given material to the ISM.
987 From this argument alone it seems logical
988 that material from the majority of old stellar
989 sources will have been homogenised to yield
990 a single, averaged, composition in the early
991 Solar System and not a multitude of grains
992 with highly exotic compositions integrating
993 stellar sources over billions of years. Moreover,
994 as Molster and Waters (2003) describe, crys-
995 talline grains are only observed around young
996 stars (proto-planetary disks) and evolved stars
997 (e.g. winds from AGB) but not in the ISM.
998 This suggests that the grains that are forming
999 in the ISM are amorphous and may be more
1000 volatile and so may have been more easily fur-
1001 ther homogenised by higher temperature pro-
1002 cessing in the proto-Solar nebula.

1003 Younger grains from more recent events
1004 ($\ll 1$ Ga to ~ 1 Ma prior to the formation of
1005 the proto-Solar nebula), however, are more
1006 likely to survive intact and to remain crys-
1007 talline. Therefore, more recent events may
1008 still present grains with large anomalies to
1009 the early Solar System. Due to the lifetime
1010 of grains ($5-20 \times 10^8$ years, Barlow 1978) it
1011 is unlikely they will have been destroyed to
1012 form new phases so will not reflect the ho-
1013 mogenised isotopic compositions of the host
1014 molecular cloud or ISM. However, to a vari-
1015 able degree they will likely have experienced
1016 processing in the ISM or molecular cloud and
1017 this may affect their ability to contribute to
1018 isotopic heterogeneity in the proto-planetary
1019 disk.

1020 An important point to clarify here is what
1021 the anomalies that are measured mean in
1022 terms of grains, or carrier phases. To present
1023 an isotopic anomaly in the early Solar System,
1024 grains must be heterogeneously distributed.
1025 For example, large anomalies are present in
1026 CC leachates in Zr isotopes (Schönbächler

et al. 2005), however, much smaller anomalies are present in the bulk meteorites. This is because the carrier phase of Zr isotopes anomalies was relatively homogeneously distributed between the different chondrite parent bodies. For the bulk meteorites, normal CAI and hibonite populations of interest to this study, the anomalies represent different abundances of carrier phases. Therefore, the effectiveness of a grain to transfer an anomaly from a stellar source to a proto-planetary disk relies on its ability to either remain, or become, heterogeneously distributed through the nebula. Factors that may affect this are: volatility, as volatile grains are effectively mixed as they enter the gas phase; crystallinity, as more crystalline phases may be more refractory but also effectively sorted by physical processes such as photophoresis (Krauss and Wurm 2005; Wurm et al. 2010); size; density or chemistry. Clearly some of the factors are more important than others; if the grains are chemically unstable or volatile, size and density sorting will be less significant. Therefore, we can examine the effects of processing in the molecular cloud and ISM on some of these important factors for the preservation of anomalies.

4.3.2. *Relationships between hibonite and bulk meteorite sources*

We find that the sources of the hibonite Ti isotope anomalies are not related to the sources of the bulk meteorite anomalies, see section 3.1 above. This is an interesting finding because while the hibonites represent the largest Ti isotope anomalies in the Solar System, the bulk meteorites constitute much more material and so actually contain a much greater mass of anomalous Ti. Therefore, it might have been expected that these two repositories of anomalous Ti share a common source.

There are two end-member explanations for why the hibonite sources and the bulk meteorites sources are not the same:

1. The dominant source in the bulk meteorites is not a primary nucleosynthetic source(s) as previously thought (e.g. Leya et al. 2008, 2009; Qin et al. 2011; Steele et al. 2012) but rather is made within the Solar System by mixing one or more of the hibonite sources, or some other sources, at some point after hibonite formation but before CAI formation as normal CAIs sample the same sources as the bulk meteorites (Trinquier et al. 2009).
2. The hibonite sources, while highly concentrated in the hibonite forming regions, are not dominant in the Solar System and have been diluted to extinction during subsequent mixing.

The first scenario poses the very interesting prospect of being able to directly examine the timescales of early Solar System mixing in the early Solar System. By dating hibonite formation we may find the final time at which primary nucleosynthetic sources were sampled. This may then be compared with the age of CAI formation, after which time full Solar System mixing of the primary sources had occurred. This comparison will yield information about the time taken for large scale homogenisation of the primary nucleosynthetic sources. As discussed above, and in previous studies (e.g. Ireland 1990), the hibonites with the largest anomalies are the PLACs, these then might represent the earliest samples. The SHIBs have much smaller, or no, stable isotope anomalies coupled with variable ^{26}Al abundances (Sahijpal and Goswami 1998). The SHIBs may simply represent hibonite formation contemporaneously with the PLACs, but in a region with lower abundance of anomalous Ti carriers and higher abundances of ^{26}Al carriers (Liu et al. 2012). Alternatively, the SHIBs may represent an intermediate stage of mixing between the PLAC sources and the bulk meteorite sources. Possibly the carrier phase for the bulk meteorite

1116 variation.

1117 While it is certainly a possibility that the
1118 SHIBs represent an intermediate stage of mix-
1119 ing, the current level of precision on Ti iso-
1120 tope compositions the hibonite population are
1121 not high enough to determine if this is the
1122 case. However, we can place limits on the ex-
1123 tent of mixing that would have had to occur.
1124 The precision of the hibonite measurements
1125 would place the bulk meteorite source com-
1126 position only +200 ‰ in $\epsilon^{50}\text{Ti}_{49}$ and + 50
1127 ‰ in $\epsilon^{46}\text{Ti}_{49}$. In order to create the varia-
1128 tion in $\epsilon^{50}\text{Ti}_{49}$ from SHIBs with this compo-
1129 sition would require hibonite abundances be-
1130 tween 50 and 200 times those observed in CM
1131 hibonites, depending on Ti concentration in
1132 the hibonites (using concentration and abun-
1133 dance data from Ireland et al. 1988; Was-
1134 son and Kallemeyn 1988). Under nebula con-
1135 ditions, outside the CAI forming region, hi-
1136 bonite is a physically, chemically and ther-
1137 mally stable mineral, therefore, if hibonite
1138 grains had been present in these abundances it
1139 seems unlikely it would be depleted so sig-
1140 nificantly in all chondrite groups, especially
1141 in primitive groups such as CM chondrites.
1142 Moreover, other chondrites contain even lower
1143 abundances of hibonite, so would require even
1144 more complete reprocessing. Therefore, we fa-
1145 vor the second model in which the hibonite
1146 sources, although highly anomalous in the hi-
1147 bonite forming region, are not dominant on a
1148 bulk Solar System scale (e.g. Boss 2008).

1149 The second scenario, in which the sources
1150 hibonite of Ti isotope anomalies are diluted
1151 to extinction during the final stages of ho-
1152 mogenisation in the protosolar nebula, makes
1153 the hibonite source compositions less signifi-
1154 cant for the bulk composition of the Solar Sys-
1155 tem. However, these sources record nucleosyn-
1156 thetic information not present in other early
1157 Solar System samples and so may provide a
1158 tantalising, fine scale, view of the astrophys-
1159 ical birth environment of the Solar System.
1160 They may record the compositions of stellar

1161 environments which, while not dominant, con-
1162 tributed material to the Solar System while it
1163 was forming; they provide a more complete
1164 view of all of the stellar sources to the proto-
1165 Solar Nebula.

1166 The sources hibonite Ti isotope anom-
1167 alies were not homogenised into the molecu-
1168 lar cloud composition, therefore they must be
1169 younger than around 100 Ma, see above sec-
1170 tion 4.3.1. These sources were likely some
1171 combination of SN Ia and SN II. The grains
1172 in material ejected during these events would
1173 still be crystalline after ~ 100 Ma and so would
1174 have remained relatively impervious to ther-
1175 mal processing during the early collapse of the
1176 proto-Solar nebula. This relatively recently
1177 synthesised material, may have been concen-
1178 trated in clumps as it hasn't had as much
1179 time to processed and mixed into the molecu-
1180 lar cloud, so could survive in relatively con-
1181 centrated clumps in the proto-solar nebula.
1182 These clumps may be distributed throughout
1183 the proto-solar nebula, but the only samples
1184 we derive from this time were in the hibonite
1185 forming region. It is in these clumps that the
1186 hibonites would form, partially mixing with
1187 homogenised molecular cloud material to give
1188 the compositions we observe in the hibonites
1189 today. Subsequently, these clumps would be
1190 dynamically mixed into the rest of the Solar
1191 System (e.g. Boss 2008; Ciesla 2009).

1192 There are three mechanisms by which the
1193 highly anomalous material in these clump
1194 would not dominate the bulk meteorite com-
1195 positions which formed later. Firstly, the
1196 sources may have been in such low concentra-
1197 tion in the bulk proto-solar nebula that they
1198 were simply be diluted to extinction. Sec-
1199 ondly, they may have been somewhat amor-
1200 phitized by radiation damage in the molecular
1201 cloud and more susceptible to the thermal pro-
1202 cessing than the bulk meteorite source. Lastly,
1203 the majority of the material may have been
1204 lost to the proto-Sun during infall and that
1205 the hibonite grains were propelled back out

1206 into the Solar System on stellar winds or in
1207 bipolar outflow jets (e.g. Shu et al. 2001).

1208 In addition to large stable isotope anom-
1209 lies, the PLACs, which best represent the
1210 hibonite sources, are devoid of evidence for
1211 live ^{26}Al . Therefore, the hibonites may have
1212 formed from material devoid of ^{26}Al , either
1213 dominantly form a supernova zone which does
1214 not produce ^{26}Al or from material older than
1215 $\gtrsim 5$ Ma. A second possibility is that the hi-
1216 bonite grains formed contemporaneously with
1217 CAIs and experienced a thermal event in the
1218 Solar System ~ 5 Ma after CAI formation
1219 which would have erased any signature of ^{26}Al .
1220 However, if the hibonite and normal popula-
1221 tion of CAIs formed contemporaneously it is
1222 difficult understand why the hibonite grains
1223 have such large stable isotope anomalies.

1224 4.3.3. *Why is the bulk meteorite source dom-* 1225 *inant?*

1226 There is evidence that a wide variety of dif-
1227 ferent nucleosynthetic sources were present in
1228 the proto-Solar nebula. However, only one of
1229 these sources dominates the Ti isotope varia-
1230 tion in the bulk meteorites. Taking again the
1231 example of Ti, the O/Ne zone from a SN II
1232 appears to be the dominant source in the bulk
1233 meteorites, and so the bulk Solar System. It
1234 is possible that this source is dominant simply
1235 because it was the most concentrated. How-
1236 ever, due to processing in the ISM, material
1237 from older stellar sources may be more read-
1238 ily homogenised in the early Solar system, see
1239 section 4.3.1 above. In this way material from
1240 more recent nucleosynthetic sources may be
1241 more resistant to mixing in the early Solar
1242 System. Therefore, the dominant sources in
1243 the bulk meteorites and normal CAIs may not
1244 be the most concentrated but the freshest, or
1245 more recently synthesised. With further stud-
1246 ies it may be possible to test this hypothesis
1247 by examining potential correlations between
1248 the sources of stable isotope anomalies (like
1249 Ti) and the sources of SLRs (like ^{26}Al).

1250 5. Conclusions

1251 We have examined mixing relationships
1252 in the early Solar System by comparing the
1253 sources of Ti isotope variation. Using Inde-
1254 pendent Component Analysis we have com-
1255 pared the sources of Ti isotope variation in
1256 hibonite grains and bulk meteorites. We find
1257 the variation in bulk meteorites represents
1258 mixing of three sources, while the hibonite
1259 populations are consistent with mixing of
1260 four sources. The ICA shows that the highly
1261 anomalous sources of bulk meteorite and hi-
1262 bonite Ti isotope variation are not related.
1263 One source which is consistent between both
1264 data sets has a composition close to the ter-
1265 restrial ratio. This source may be represented
1266 by molecular cloud material, highly processed
1267 and homogenised by radiation and thermal
1268 events significantly before the start of the So-
1269 lar System.

1270 The finding that the anomalous nucleosyn-
1271 thetic sources represented by the variation in
1272 hibonite and bulk meteorites are not related
1273 has significant implications for mixing in the
1274 early Solar System. One possible explanation
1275 is that the sources which dominate the vari-
1276 ation in the bulk meteorites are not primary
1277 nucleosynthetic sources but rather were cre-
1278 ated by mixing two or more hibonite sources.
1279 However, this requires a stage of full proto-
1280 Solar nebula mixing from which we derive no
1281 samples. An alternative hypothesis is that the
1282 hibonite sources, although dominant in the
1283 hibonite forming regions, are not significant
1284 sources for isotope anomalies in the bulk Solar
1285 System. This may be because the sources were
1286 of low concentration after major homogenisa-
1287 tion had occurred. Another, possibly more
1288 likely, hypothesis is that is that the carrier
1289 phases of the hibonite Ti isotope anomalies,
1290 while abundant, were not physically or chem-
1291 ically robust enough to survive the vigorous
1292 mixing in the proto-Solar nebula.

1293 **Acknowledgements**

1294 We would like to thank Kevin McKeegan
1295 and Ming-Chang Liu for detailed and insight-
1296 ful comments, thorough discussion and en-
1297 couragement. We would also like to thank
1298 Andrew Davis and Levke Kööp for reviewing
1299 this manuscript and their detailed comments
1300 which greatly improved the manuscript. We
1301 would like to thank Eric Feigelson for his ed-
1302 itorial handling. This work was funded by
1303 UCLA and NASA Cosmochemistry.

1304 **REFERENCES**

- 1305 Barlow, M. J., 1978. The destruction and
1306 growth of dust grains in interstellar space.
1307 I - Destruction by sputtering. *Monthly No-*
1308 *tices of the Royal Astronomical Society* 183,
1309 367–395.
- 1310 Boss, A. P., 2008. Mixing in the solar neb-
1311 ular: Implications for isotopic heterogene-
1312 ity and large-scale transport of refractory
1313 grains. *Earth and Planetary Science Letters*
1314 268 (1–2), 102–109.
- 1315 Burkhardt, C., Kleine, T., Oberli, F., Pack,
1316 A., Bourdon, B., Wieler, R., 2011. Molyb-
1317 denum isotope anomalies in meteorites:
1318 Constraints on solar nebula evolution and
1319 origin of the earth. *Earth and Planetary*
1320 *Science Letters* 312 (3–4), 390–400.
- 1321 Carrez, P., Demyk, K., Cordier, P., Gengem-
1322 bre, L., Grimblot, J., D’Hendecourt, L.,
1323 Jones, A. P., Leroux, H., 2002. Low-energy
1324 helium ion irradiation-induced amorphiza-
1325 tion and chemical changes in olivine: In-
1326 sights for silicate dust evolution in the in-
1327 terstellar medium. *Meteoritics & Planetary*
1328 *Science* 37 (11), 1599–1614.
- 1329 Ciesla, F. J., 2009. Two-dimensional transport
1330 of solids in viscous protoplanetary disks.
1331 *Icarus* 200 (2), 655–671.
- 1332 Comon, P., 1994. Independent component
1333 analysis, a new concept? *Signal Process-*
1334 *ing* 36 (3), 287–314.
- 1335 Cuesta-Albertos, J., Gordaliza, A., Matrán,
1336 C., et al., 1997. Trimmed k -means: an at-
1337 tempt to robustify quantizers. *The Annals*
1338 *of Statistics* 25 (2), 553–576.
- 1339 Dauphas, N., Cook, D. L., Sacarabany, A.,
1340 Fröhlich, C., Davis, A. M., Wadhwa, M.,
1341 Pourmand, A., Rauscher, T., Gallino, R.,
1342 2008. Iron-60 Evidence for Early Injection
1343 and Efficient Mixing of Stellar Debris in the
1344 Protosolar Nebula. *The Astrophysical Jour-*
1345 *nal* 686 (1), 560–569.
- 1346 Dauphas, N., Davis, A., Marty, B., Reis-
1347 berg, L., 2004. The cosmic molybdenum-
1348 ruthenium isotope correlation. *Earth and*
1349 *Planetary Science Letters* 226 (3), 465–475.
- 1350 Dauphas, N., Remusat, L., Chen, J. H.,
1351 Roskosz, M., Papanastassiou, D. A.,
1352 Stodolna, J., Guan, Y., Ma, C., Eiler,
1353 J. M., 2010. Neutron-rich Chromium Iso-
1354 tope Anomalies in Supernova Nanoparti-
1355 cles. *The Astrophysical Journal* 720, 1577–
1356 1591.
- 1357 Dwek, E., Scalo, J. M., 1980. The evolution
1358 of refractory interstellar grains in the so-
1359 lar neighborhood. *The Astrophysical Jour-*
1360 *nal* 239, 193–211.
- 1361 Efron, B., 1981. Nonparametric estimates of
1362 standard error: the jackknife, the bootstrap
1363 and other methods. *Biometrika* 68 (3), 589–
1364 599.
- 1365 Fahey, A., Goswami, J. N., McKeegan, K. D.,
1366 Zinner, E., 1985. Evidence for extreme Ti-
1367 50 enrichments in primitive meteorites. *The*
1368 *Astrophysical Journal, Letters* 296, L17–
1369 L20.
- 1370 Fahey, A. J., Goswami, J. N., McKeegan,
1371 K. D., Zinner, E. K., 1987a. ^{26}Al , ^{244}Pu ,
1372 ^{50}Ti , REE, and trace element abundances

- 1373 in hibonite grains from CM and CV me-
1374 teorites. *Geochimica et Cosmochimica Acta*
1375 51 (2), 329–350.
- 1376 Fahey, A. J., Goswami, J. N., McKeegan,
1377 K. D., Zinner, E. K., 1987b. O-16 excesses
1378 in Murchison and Murray hibonites - A
1379 case against a late supernova injection ori-
1380 gin of isotopic anomalies in O, Mg, Ca, and
1381 Ti. *The Astrophysical Journal, Letters* 323,
1382 L91–L95.
- 1383 Fritz, H., Garcia-Escudero, L. A., Mayo-Iscar,
1384 A., 2012. tclust: An r package for a trim-
1385 ming approach to cluster analysis. *Journal*
1386 *of Statistical Software* 47 (12), 1–26.
- 1387 Gallino, R., Davis, A., 2009. Personal commu-
1388 nication.
- 1389 Hashimoto, M., 1995. Supernova nucleosyn-
1390 thesis in massive stars. *Progress of Theo-*
1391 *retical Physics* 94 (5), 663–736.
- 1392 Heydegger, H. R., Foster, J. J., Compston,
1393 W., 1979. Evidence of a new isotopic
1394 anomaly from titanium isotopic ratios in
1395 meteoric materials. *Nature* 278 (5706), 704–
1396 707.
- 1397 Hinton, R. W., Davis, A. M., Scatena-Wachel,
1398 D. E., 1987. Large negative Ti-50 anomalies
1399 in refractory inclusions from the Murchison
1400 carbonaceous chondrite - Evidence for in-
1401 complete mixing of neutron-rich supernova
1402 ejecta into the solar system. *The Astrophys-*
1403 *ical Journal* 313, 420–428.
- 1404 Hyvarinen, A., 1999. Fast and robust fixed-
1405 point algorithms for independent com-
1406 ponent analysis. *Neural Networks, IEEE*
1407 *Transactions on* 10 (3), 626–634.
- 1408 Hyvärinen, A., Oja, E., 2000. Independent
1409 component analysis: algorithms and appli-
1410 cations. *Neural Networks* 13 (4–5), 411–430.
- 1411 Ireland, T. R., 1990. Presolar isotopic and
1412 chemical signatures in hibonite-bearing re-
1413 fractory inclusions from the Murchison car-
1414 bonaceous chondrite. *Geochimica et Cos-*
1415 *mochimica Acta* 54 (11), 3219–3237.
- 1416 Ireland, T. R., Fahey, A. J., Zinner,
1417 E. K., 1988. Trace-element abundances
1418 in hibonites from the murchison carbona-
1419 ceous chondrite: Constraints on high-
1420 temperature processes in the solar nebula.
1421 *Geochimica et Cosmochimica Acta* 52 (12),
1422 2841–2854.
- 1423 Ireland, T. R., Zinner, E. K., Fahey, A. J.,
1424 Esat, T. M., 1992. Evidence for distilla-
1425 tion in the formation of hal and related
1426 hibonite inclusions. *Geochimica et Cos-*
1427 *mochimica Acta* 56 (6), 2503–2520.
- 1428 Iwamori, H., Albarède, F., 2008. Decoupled
1429 isotopic record of ridge and subduction
1430 zone processes in oceanic basalts by in-
1431 dependent component analysis. *Geochem-*
1432 *istry, Geophysics, Geosystems* 9 (4).
- 1433 Iwamoto, K., Brachwitz, F., Nomoto, K.,
1434 Kishimoto, N., Umeda, H., Hix, W. R.,
1435 Thielemann, F., 1999. Nucleosynthesis in
1436 chandrasekhar mass models for type IA su-
1437 pernovae and constraints on progenitor sys-
1438 tems and burning-front propagation. *Astro-*
1439 *physical Journal, Supplement Series* 125,
1440 439–462.
- 1441 Jones, A. P., May 2004. Dust Destruction Pro-
1442 cesses. In: Witt, A. N., Clayton, G. C.,
1443 Draine, B. T. (Eds.), *Astrophysics of Dust*.
1444 Vol. 309 of *Astronomical Society of the Pa-*
1445 *cific Conference Series*. p. 347.
- 1446 Jones, A. P., Nuth, J. A., 2011. Dust destruc-
1447 tion in the ism: a re-evaluation of dust life-
1448 times. *A&A* 530.
- 1449 Kemper, F., Vriend, W. J., Tielens, A. G.
1450 G. M., 2004. The absence of crystalline sili-
1451 cates in the diffuse interstellar medium. *The*
1452 *Astrophysical Journal* 609 (2), 826–837.

- 1453 Krauss, O., Wurm, G., 2005. Photophoresis and the pile-up of dust in young circumstellar disks. *The Astrophysical Journal* 630 (2), 1088.
- 1454
1455
1456
- 1457 Lee, T., Papanastassiou, D. A., Wasserburg, G. J., 1978. Calcium isotopic anomalies in the Allende meteorite. *The Astrophysical Journal, Letters* 220, L21–L25.
- 1458
1459
1460
- 1461 Leya, I., Schönbachler, M., Wiechert, U., Krähenbühl, U., Halliday, A. N., 2008. Titanium isotopes and the radial heterogeneity of the solar system. *Earth and Planetary Science Letters* 266, 233–244.
- 1462
1463
1464
1465
- 1466 Leya, I., Schönbachler, M., Krähenbühl, U., Halliday, A. N., 2009. New Titanium Isotope Data for Allende and Efremovka CAIs. *The Astrophysical Journal* 702 (2), 1118–1126.
- 1467
1468
1469
1470
- 1471 Liu, M.-C., Chaussidon, M., Göpel, C., Lee, T., 2012. A heterogeneous solar nebula as sampled by cm hibonite grains. *Earth and Planetary Science Letters* 327–328 (0), 75–83.
- 1472
1473
1474
1475
- 1476 Liu, M.-C., McKeegan, K. D., Goswami, J. N., Marhas, K. K., Sahijpal, S., Ireland, T. R., Davis, A. M., 2009. Isotopic records in cm hibonites: Implications for timescales of mixing of isotope reservoirs in the solar nebula. *Geochimica et Cosmochimica Acta* 73 (17), 5051–5079.
- 1477
1478
1479
1480
1481
1482
- 1483 Lyons, J., Young, E., 2005. CO self-shielding as the origin of oxygen isotope anomalies in the early solar nebula. *Nature* 435 (7040), 317–20.
- 1484
1485
1486
- 1487 Lyons, J. R., Bergin, E. A., Ciesla, F. J., Davis, A. M., Desch, S. J., Hashizume, K., Lee, J. E., 2009. Timescales for the evolution of oxygen isotope compositions in the solar nebula. *Geochimica et Cosmochimica Acta* 73 (17), 4998–5017.
- 1488
1489
1490
1491
1492
- 1493 Maeda, K., Röpke, F. K., Fink, M., Hillebrandt, W., Travaglio, C., Thielemann, F., 2010. Nucleosynthesis in two-dimensional delayed detonation models of type Ia supernova explosions. *The Astrophysical Journal* 712 (1), 624–638.
- 1494
1495
1496
1497
1498
- 1499 Mahon, K., 1996. The new “York” regression: Application of an improved statistical method to geochemistry. *International Geology Review* 38 (4), 293–303.
- 1500
1501
1502
- 1503 Marchini, J., Heaton, C., Ripley, B., 2013. fastica: Fastica algorithms to perform ica and projection pursuit. R package version, 1–2.
- 1504
1505
- 1506 Marhas, K. K., Goswami, J. N., Davis, A. M., 2002. Short-lived nuclides in hibonite grains from murchison: Evidence for solar system evolution. *Science* 298 (5601), 2182–2185.
- 1507
1508
1509
- 1510 McCulloch, M. T., Wasserburg, G. J., 1978. Barium and neodymium isotopic anomalies in the Allende meteorite. *The Astrophysical Journal, Letters* 220, L15–L19.
- 1511
1512
1513
- 1514 Meyer, B. S., Krishnan, T. D., Clayton, D. D., 1996. ^{48}Ca production in matter expanding from high temperature and density. *The Astrophysical Journal* 462, 825–839.
- 1515
1516
1517
- 1518 Meyer, B. S., Weaver, T. A., Woosley, S. E., 1995. Isotope source table for a 25 M_{\odot} supernova. *Meteoritics* 30, 325–334.
- 1519
1520
- 1521 Molster, F., Waters, L., 2003. The mineralogy of interstellar and circumstellar dust. In: Henning, T. (Ed.), *Astromineralogy*. Vol. 609 of *Lecture Notes in Physics*. Springer Berlin Heidelberg, pp. 121–170.
- 1522
1523
1524
1525
- 1526 Niederer, F. R., Papanastassiou, D. A., Wasserburg, G. J., 1985. Absolute isotopic abundances of Ti in meteorites. *Geochimica et Cosmochimica Acta* 49 (3), 835–851.
- 1527
1528
1529
- 1530 Niemeyer, S., Lugmair, G. W., 1980. Ti isotope anomalies in an “un-fun” Allende inclusion. *Meteoritics* 15 (4), 341.
- 1531
1532

- 1533 Niemeyer, S., Lugmair, G. W., 1981. Ubiquitous isotopic anomalies in Ti from normal Allende inclusions. *Earth and Planetary Science Letters* 53 (2), 211–225.
- 1534
1535
1536
- 1537 Nomoto, K., Hashimoto, M., Tsujimoto, T., Thielemann, F. K., Kishimoto, N., Kubo, Y., Nakasato, N., 1997. Nucleosynthesis in type II supernovae. *Nuclear Physics A* 616, 79–90.
- 1538
1539
1540
1541
- 1542 Qin, L., Nittler, L. R., Alexander, C. M. O., Wang, J., Stadermann, F. J., Carlson, R. W., 2011. Extreme ^{54}Cr -rich nanooxides in the CI chondrite Orgueil – Implication for a late supernova injection into the Solar System. *Geochimica et Cosmochimica Acta* 75 (2), 629–644.
- 1543
1544
1545
1546
1547
1548
- 1549 R Core Team, 2013. R: A Language and Environment for Statistical Computing. R Foundation for Statistical Computing, Vienna, Austria.
- 1550
1551
1552
- 1553 Rauscher, T., Heger, A., Hoffman, R. D., Woosley, S. E., 2002. Nucleosynthesis in Massive Stars with Improved Nuclear and Stellar Physics. *The Astrophysical Journal* 576 (1), 323–348.
- 1554
1555
1556
1557
- 1558 Russell, W., Papanastassiou, D., Tombrello, T., 1978. Ca isotope fractionation on the Earth and other Solar System materials. *Geochimica et Cosmochimica Acta* 42, 1075–1090.
- 1559
1560
1561
1562
- 1563 Sahijpal, S., Goswami, J. N., 1998. Refractory phases in primitive meteorites devoid of 26Al and 41Ca: representative samples of first solar system solids? *The Astrophysical Journal Letters* 509 (2), L137.
- 1564
1565
1566
1567
- 1568 Sahijpal, S., Goswami, J. N., Davis, A. M., 2000. K, Mg, Ti and Ca isotopic compositions and refractory trace element abundances in hibonites from CM and CV meteorites: implications for early solar system processes. *Geochimica et Cosmochimica Acta* 64 (11), 1989–2005.
- 1569
1570
1571
1572
1573
1574
- 1575 Schönbachler, M., Rehkämper, M., Fehr, M. A., Halliday, A. N., Hattendorf, B., Günther, D., 2005. Nucleosynthetic zirconium isotope anomalies in acid leachates of carbonaceous chondrites. *Geochimica et Cosmochimica Acta* 69 (21), 5113–5122.
- 1576
1577
1578
1579
1580
- 1581 Shu, F., Shang, H., Gounelle, M., Glassgold, A., Lee, T., 2001. The origin of chondrules and refractory inclusions in chondritic meteorites. *The Astrophysical Journal* 548 (2), 1029–1050.
- 1582
1583
1584
1585
- 1586 Steele, R. C. J., Coath, C. D., Regelous, M., Russell, S., Elliott, T., 2010. Correlated Neutron Rich Ni Isotope Anomalies in Chondritic and Iron Meteorites. *Lunar and Planetary Institute Science Conference Abstracts* 41, 1984.
- 1587
1588
1589
1590
1591
- 1592 Steele, R. C. J., Coath, C. D., Regelous, M., Russell, S., Elliott, T., 2012. Neutron-poor nickel isotope anomalies in meteorites. *The Astrophysical Journal* 758 (1), 59.
- 1593
1594
1595
- 1596 Steele, R. C. J., Elliott, T., Coath, C. D., Regelous, M., 2011. Confirmation of mass-independent Ni isotopic variability in iron meteorites. *Geochimica et Cosmochimica Acta* 75 (24), 7906–7925.
- 1597
1598
1599
1600
- 1601 Thiemens, M. H., 1999. Mass-independent isotope effects in planetary atmospheres and the early solar system. *Science* 283 (5400), 341–345.
- 1602
1603
1604
- 1605 Travaglio, C., Hillebrandt, W., Reinecke, M., Thielemann, F., 2004. Nucleosynthesis in multi-dimensional SN Ia explosions. *Astronomy and Astrophysics* 425, 1029–1040.
- 1606
1607
1608
- 1609 Travaglio, C., Röpke, F. K., Gallino, R., Hillebrandt, W., 2011. Type Ia supernovae as sites of the p-process: Two-dimensional models coupled to nucleosynthesis. *The Astrophysical Journal* 739 (2), 93.
- 1610
1611
1612
1613
- 1614 Trinquier, A., Birck, J. L., Allègre, C. J., Göpel, C., Ulfbeck, D., 2008. ^{53}Mn – ^{53}Cr
- 1615

- 1616 systematics of the early Solar System re-
1617 visited. *Geochimica et Cosmochimica Acta*
1618 72 (20), 5146–5163.
- 1619 Trinquier, A., Elliott, T., Ulfbeck, D., Coath,
1620 C., Krot, A. N., Bizzarro, M., 2009. Ori-
1621 gin of nucleosynthetic isotope heterogene-
1622 ity in the Solar protoplanetary disk. *Science*
1623 324 (5925), 374–376.
- 1624 Umeda, H., Nomoto, K., 2002. Nucleosynthe-
1625 sis of zinc and iron peak elements in popu-
1626 lation III type II supernovae: Comparison
1627 with abundances of very metal poor halo
1628 stars. *The Astrophysical Journal* 565, 385–
1629 404.
- 1630 Usui, T., Iwamori, H., 2013. Mixing relations
1631 of the howardite-eucrite-diogenite suite: A
1632 new statistical approach of independent
1633 component analysis for the dawn mission.
1634 *Meteoritics & Planetary Science* 48 (11),
1635 2289–2299.
- 1636 Völkening, J., Papanastassiou, D. A., 1989.
1637 Iron isotope anomalies. *The Astrophysical*
1638 *Journal, Letters* 347, L43–L46.
- 1639 Wasson, J. T., Kallemeyn, G. W., 1988.
1640 Compositions of chondrites. *Philosophical*
1641 *Transactions of the Royal Society AA325*,
1642 535 – 544.
- 1643 Williams, N., Schönbächler, M., Fehr, M.,
1644 Akram, A., Parkinson, I., et al., 2014. Dif-
1645 ferent heterogeneously distributed titanium
1646 isotope components in solar system mate-
1647 rials and mass-dependent titanium isotope
1648 variations. In: Lunar and Planetary Insti-
1649 tute Science Conference Abstracts. Vol. 45.
1650 p. 2183.
- 1651 Woosley, S. E., 1997. Neutron-rich Nucleosyn-
1652 thesis in Carbon Deflagration Supernovae.
1653 *The Astrophysical Journal* 476 (2), 801–
1654 810.
- 1655 Wurm, G., Teiser, J., Bischoff, A., Haack, H.,
1656 Roszjar, J., 2010. Experiments on the pho-
1657 tophoretic motion of chondrules and dust
1658 aggregates—indications for the transport
1659 of matter in protoplanetary disks. *Icarus*
1660 208 (1), 482–491.
- 1661 York, D., 1969. Least squares fitting of a
1662 straight line with correlated errors. *Earth*
1663 *and Planetary Science Letters* 5, 320–324.
- 1664 Young, E. D., Galy, A., Nagahara, H., 2002.
1665 Kinetic and equilibrium mass-dependent
1666 isotope fractionation laws in nature and
1667 their geochemical and cosmochemical sig-
1668 nificance. *Geochimica et Cosmochimica*
1669 *Acta* 66 (6), 1095–1104.
- 1670 Zhang, J., Dauphas, N., Davis, A. M., Leya,
1671 I., Fedkin, A., 2012. The proto-earth as a
1672 significant source of lunar material. *Nature*
1673 *Geosci* 5 (4), 251–255.
- 1674 Zhang, J., Dauphas, N., Davis, A. M., Pour-
1675 mand, A., 2011. A new method for mc-
1676 icpms measurement of titanium isotopic
1677 composition: Identification of correlated
1678 isotope anomalies in meteorites. *J. Anal.*
1679 *At. Spectrom.* 26, 2197–2205.
- 1680 Zinner, E., 2003. Presolar grains. In: Holland,
1681 H. D., Turekian, K. K. (Eds.), *Treatise on*
1682 *Geochemistry*. pp. 17–39.
- 1683 Zinner, E., Jadhav, M., Gyngard, F., Nittler,
1684 L., 2010. Bonanza: Isotopic anatomy of a
1685 large pre-solar sic grain of type x. *Meteo-*
1686 *ritics and Planetary Science Supplement*
1687 73, 5137.
- 1688 Zinner, E. K., Fahey, A. J., McKeegan, K. D.,
1689 Goswami, J. N., Ireland, T. R., 1986. Large
1690 Ca-48 anomalies are associated with Ti-
1691 50 anomalies in Murchison and Murray hi-
1692 bonites. *The Astrophysical Journal, Letters*
1693 311, L103–L107.

This 2-column preprint was prepared with the AAS
L^AT_EX macros v5.2.

1694 Appendix: Table of Renormalized Hibonite Ti Isotope Compositions

Sample Name	Type	Reference	$\epsilon^{46}\text{Ti}_{49}^{47}$	2 s.e.	$\epsilon^{48}\text{Ti}_{49}^{47}$	2 s.e.	$\epsilon^{50}\text{Ti}_{49}^{47}$	2 s.e.
7-76	SHIB	Ireland 1990	-6.1	64.6	2.0	29.8	36.9	87.1
7-143	SHIB	Ireland 1990	8.1	8.7	2.0	4.7	28.1	14.5
7-290	SHIB	Ireland 1990	39.6	68.8	6.9	31.1	-57.4	91.5
7-373	SHIB	Ireland 1990	-9.8	64.9	7.6	27.7	-4.8	79.2
7412t	PLAC	Ireland 1990	5.4	7.1	14.6	4.1	40.4	14.0
7-505	SHIB	Ireland 1990	-4.3	44.3	12.1	19.6	25.8	57.8
7-551	SHIB	Ireland 1990	-1.1	59.7	3.0	26.9	33.0	83.7
7-644	PLAC	Ireland 1990	69.5	80.5	31.5	33.8	-13.0	93.3
7-658	PLAC	Ireland 1990	58.0	63.9	46.7	28.7	55.9	84.4
7-664	SHIB	Ireland 1990	6.3	8.9	21.6	5.2	22.3	17.5
7-734	SHIB	Ireland 1990	11.9	49.7	20.1	22.0	25.9	63.4
7-789	SHIB	Ireland 1990	5.8	5.5	16.1	3.3	11.9	12.7
7-821	SHIB	Ireland 1990	20.9	51.4	-0.6	23.3	58.7	69.1
7-953	SHIB	Ireland 1990	9.4	9.0	16.6	5.2	16.4	18.0
7-980	PLAC	Ireland 1990	-10.1	10.7	23.7	6.4	-110.0	18.1
7-981	PLAC	Ireland 1990	-16.1	7.1	19.7	4.1	-112.0	12.8
7-A84	SHIB	Ireland 1990	-8.1	38.9	-2.0	18.0	24.9	52.9
7-A95	SHIB	Ireland 1990	30.2	59.9	-9.6	24.8	-53.7	69.6
8-47	SHIB	Ireland 1990	12.6	40.8	7.5	18.7	7.6	53.4
849	SHIB	Ireland 1990	50.9	64.2	7.9	29.0	-34.3	77.8
8-65	SHIB	Ireland 1990	5.1	44.7	-1.0	19.5	19.1	59.0
8-66	PLAC	Ireland 1990	-24.3	66.6	23.8	27.9	-208.0	80.3
13-02	SHIB	Ireland 1990	-11.0	45.0	-9.0	20.6	28.9	69.1
13-03	SHIB	Ireland 1990	4.2	29.4	-6.0	14.2	20.1	43.3
13-04	SHIB	Ireland 1990	-16.5	31.2	12.1	14.9	29.6	45.0
13-13	PLAC	Ireland 1990	119.0	59.7	-38.2	24.0	2491.0	68.9
13-23	BAG	Ireland 1990	-40.2	45.8	5.7	19.1	-178.0	50.6
13-24	SHIB	Ireland 1990	-33.0	69.0	2.7	31.4	146.0	93.5
13-25	PLAC	Ireland 1990	64.8	64.6	40.1	29.3	-350.0	85.9
13-33	SHIB	Ireland 1990	-69.5	55.5	-40.2	25.8	0.0	80.6
13-37	SHIB	Ireland 1990	-10.5	43.4	-9.5	19.5	13.5	54.0
13-51	PLAC	Ireland 1990	40.5	82.6	47.8	36.5	-379.0	104.0
13-60	SHIB	Ireland 1990	-6.6	37.2	-3.5	16.7	-24.5	46.3
13-61	SHIB	Ireland 1990	23.7	39.6	-17.1	17.3	-5.4	48.5
1412	PLAC	Ireland 1990	-46.1	48.9	-0.3	22.3	-210.0	63.5
14-14	SHIB	Ireland 1990	-4.1	33.6	2.0	14.8	44.9	43.1
Mur-A1	—	Zinner 1986	-61.9	27.3	-18.3	11.6	70.9	32.1
Mur-H7	—	Zinner 1986	15.5	12.6	-15.1	9.1	58.2	31.4
Mur-H8	—	Zinner 1986	-7.9	41.0	-14.0	16.8	-1.9	48.7
Mur-20	—	Zinner 1986	4.7	7.0	23.1	4.1	-433.0	13.9
Mur-70	—	Zinner 1986	-10.7	6.5	30.3	4.8	-222.0	15.6
Mur-170	—	Zinner 1986	33.2	6.8	-6.6	3.6	-539.0	12.0
My-H3	—	Zinner 1986	48.1	25.8	-11.2	11.7	955.0	35.5
My-H4	—	Zinner 1986	47.0	58.5	-10.2	23.6	971.0	65.0
CH-B2	PLAC	Sahijpal 2000	-4.8	39.4	-17.0	18.4	502.0	65.1
CH-B7	PLAC	Sahijpal 2000	99.8	43.2	-11.8	21.4	121.0	79.9
CH-A5	SHIB	Sahijpal 2000	11.1	38.3	-28.6	16.3	-15.8	47.9
CH-B5	PLAC	Sahijpal 2000	21.7	37.6	-25.1	22.3	138.0	65.1
All-3529-42	—	Sahijpal 2000	13.6	53.4	10.5	25.1	22.6	72.1
CH-A3	SHIB	Sahijpal 2000	-11.3	73.8	-22.5	35.9	86.4	109.0
CH-B3	PLAC	Sahijpal 2000	7.8	46.5	-11.6	22.0	92.5	75.7
CH-A4	SHIB	Sahijpal 2000	2.1	71.2	-35.5	34.9	577.0	115.0
Mur-S15	SHIB	Liu 2009	-96.9	71.9	-27.1	35.2	15.3	110.0
Mur-P1	PLAC	Liu 2009	23.5	78.8	-3.1	41.8	496.0	133.0
Mur-P2	PLAC	Liu 2009	-50.1	85.7	-29.8	41.8	232.0	133.0
Mur-P6	PLAC	Liu 2009	-113.0	79.0	38.1	37.4	-39.8	117.0
Mur-P7	PLAC	Liu 2009	-140.0	75.4	-35.8	38.0	2058.0	116.0
Mur-P8	PLAC	Liu 2009	-181.0	79.4	5.2	38.2	38.6	119.0
Mur-P9-spot1	PLAC	Liu 2009	-273.0	94.8	30.5	44.4	-335.0	130.0
Mur-P9-spot2	PLAC	Liu 2009	-245.0	74.4	53.1	35.9	-410.0	107.0
Mur-B1-spot1a	BAG	Liu 2009	123.0	105.0	37.4	47.6	-50.2	139.0

Mur-B1-spot1-1a	BAG	Liu 2009	138.0	117.0	44.9	51.4	-143.0	140.0
Mur-B1-spot2a	BAG	Liu 2009	54.1	108.0	34.5	53.3	-449.0	159.0
Mur-B1-spot2-1a	BAG	Liu 2009	-72.6	122.0	9.9	54.4	-247.0	145.0
Mur-B1-spot3	BAG	Liu 2009	-35.3	85.9	-10.9	39.9	-113.0	110.0
Mur-B1-spot3-1	BAG	Liu 2009	-0.9	94.1	48.5	44.4	-16.0	126.0
MUR-AL-a	—	Fahey 1985	7.1	65.9	1.0	26.2	82.1	73.7
MUR-AL-B	—	Fahey 1985	-9.6	29.0	-0.5	12.8	48.4	35.7
MUR-H7	—	Fahey 1985	15.5	25.2	-15.1	10.3	58.2	27.7
MY-H3-a	—	Fahey 1985	32.7	29.0	-10.1	13.0	971.0	41.5
MY-H3-b	—	Fahey 1985	103.0	56.3	-12.8	24.7	902.0	68.1
ATP-1	—	Fahey 1985	-25.4	23.8	1.1	10.2	10.6	29.8
ATP-2-a	—	Fahey 1985	-27.5	36.9	3.1	16.3	-1.5	52.1
ATP-2-b	—	Fahey 1985	-0.3	46.9	16.1	18.3	17.8	49.3
ATP-3-a	—	Fahey 1985	15.4	35.1	-7.1	15.5	21.3	44.6
ATP-3-b	—	Fahey 1985	35.5	32.1	8.9	14.6	25.3	42.5
C-1	FUN	Niederer 1980	-10.6	1.6	0.6	0.7	-39.0	1.8
C-2	FUN	Niederer 1980	-11.8	1.8	1.2	0.8	-37.9	2.8
EK-1-4-1	FUN	Niederer 1980	-13.2	4.4	-15.5	2.5	17.6	9.7
EK-1-4-2	FUN	Niederer 1980	-9.8	3.4	-14.3	1.7	16.1	4.8
EK-1-4-3	FUN	Niederer 1980	-7.7	11.3	-18.0	6.4	15.1	19.3
EK-1-4-4	FUN	Niederer 1980	-6.1	5.6	-16.4	2.6	10.0	7.3
EK-1-4-5	FUN	Niederer 1980	-8.8	4.8	-16.3	2.1	14.8	5.4
DJ-1	—	Hinton 1987	-2.7	22.3	6.5	15.2	-190.0	58.9
BB-5	—	Hinton 1987	-7.1	23.8	52.6	16.3	-578.0	56.9
BB-5-c	—	Hinton 1987	-0.6	26.7	58.1	10.3	-573.0	25.6
Gr-1-r	—	Hinton 1987	39.1	23.5	7.4	12.3	-93.8	53.8
Gr-1-c	—	Hinton 1987	34.3	37.9	-9.7	16.0	-105.0	44.7
SH-7	—	Hinton 1987	1.6	20.3	-3.5	7.7	160.0	17.3
Mur-H9	—	Fahey 1987a	-26.8	32.0	-7.4	14.1	147.0	42.8
My-CH1	—	Fahey 1987a	-2.0	23.4	-4.0	10.2	30.0	29.5
MY-IP	—	Fahey 1987a	5.0	47.3	3.0	19.8	-1.0	56.0
CB-H2	—	Fahey 1987a	-1.1	24.2	5.0	10.9	-3.1	36.1
CB-H4	—	Fahey 1987a	17.4	35.3	-5.1	16.0	133.0	48.2
HAL-Ha	—	Fahey 1987a	13.4	71.0	18.6	30.4	188.0	87.0
HAL-Hb	—	Fahey 1987a	30.0	52.1	6.4	23.9	124.0	73.3
DA2-12	—	Fahey 1987a	32.3	35.0	-15.6	15.5	49.9	45.0
ATP-1b	—	Fahey 1987a	5.5	20.5	4.5	8.5	23.5	23.8
MUR-H!	Hib	Fahey 1987b	17.7	23.7	-29.1	10.3	65.2	30.3
BB-5-H	Hib	Fahey 1987b	-34.7	27.6	59.9	12.8	-574.0	37.4
BB-5-C	Corr	Fahey 1987b	-20.8	46.8	50.1	20.7	-581.0	58.4

Table 2:: Table showing Ti isotope compositions of hibonites renormalized from literature. Errors were obtained using a Monte Carlo simulation as outlined in the text.