

1 **Electronic Supplement to “The thallium isotope composition of carbonaceous**
2 **chondrites – New evidence for live ^{205}Pb in the early solar system”**

3
4 **Procedure for the correction of measured Pb concentrations for terrestrial**
5 **contamination**

6
7 The measured Pb concentrations and isotope compositions (Table 2, main text) of the
8 carbonaceous chondrites may be affected by terrestrial Pb contamination. All samples were
9 therefore screened for such contamination and appropriate corrections were applied, where
10 appropriate, using the procedures described below.

11
12 *Screening for terrestrial Pb contamination*

13 As discussed in the manuscript, samples without significant Pb contamination define a
14 correlation of increasing (more radiogenic) $^{206}\text{Pb}/^{204}\text{Pb}$ with decreasing Tl contents (Fig. 1,
15 main text). There are four samples, which clearly deviate from this correlation with
16 radiogenic $^{206}\text{Pb}/^{204}\text{Pb}$ ratios of between 11.70 and 16.66: Cold Bokkeveld CM2, Allende
17 Smithsonian CV3, Colony CO3, and NWA 801 CR2 (Table 2, main text). Only these four
18 meteorites were corrected for the presence of terrestrial Pb, whilst all others were assumed to
19 be essentially uncontaminated, containing only primitive indigenous Pb.

20
21 *Correction for terrestrial Pb contamination*

22 The procedure for the correction of terrestrial Pb contamination assumed simple, two-
23 component mixing, whereby the Pb present in the samples (Pb_s) as analyzed by mass
24 spectrometry (Table 2, main text), is a mixture of primitive indigenous meteorite Pb_i and
25 recently admixed terrestrial Pb_t .

26 The calculations assumed that the terrestrial contaminant Pb_t can feature a range of Pb
27 isotope compositions, as defined by the relationships that were used by Göpel et al. (1985) to
28 evaluate the Pb contamination of iron meteorites:

$$29 \quad {}^{207}Pb/{}^{204}Pb = 0.0898 \times {}^{206}Pb/{}^{204}Pb + 13.92 \pm 0.30 \quad (S1)$$

$$30 \quad {}^{208}Pb/{}^{204}Pb = 0.697 \times {}^{206}Pb/{}^{204}Pb + 25.3 \pm 0.5 \quad (S2)$$

31 The relationships are valid for ${}^{206}Pb/{}^{204}Pb$ values of between 16.0 and 19.5, to delineate a
32 modern terrestrial Pb field, which includes data for major ore deposits, industrial pollutants,
33 and laboratory blanks from reagents and sample processing (Göpel et al., 1985).

34 The following approach was adopted to correct for terrestrial Pb contamination:

35 1. A suitable primitive indigenous (uncontaminated) Pb_i isotope composition was first
36 chosen for each meteorite sample. This composition is obviously unknown and an
37 appropriate composition or range of compositions must therefore be estimated. Based on
38 the correlation of Fig. 1, this was achieved by applying the Pb isotope composition of a
39 similar but uncontaminated meteorite (Table 2), as follows:

40 (i) The measured Pb_s isotope compositions of Cold Bokkeveld CM2 (${}^{206}Pb/{}^{204}Pb \approx 13.9$)
41 and Colony CO3 (${}^{206}Pb/{}^{204}Pb \approx 16.7$) are clearly strongly contaminated with
42 terrestrial Pb_t (Fig. 1, Table 2). For these meteorites, the isotope composition of Pb_i
43 was assumed to be best approximated by the data obtained for Murchison CM2 and
44 Kainsaz CO3, respectively (Table 2).

45 (ii) Allende Smithsonian CV3 and NWA 801 CR2 feature less radiogenic (less
46 contaminated) Pb isotope compositions with ${}^{206}Pb/{}^{204}Pb_s$ ratios of about 11.7 to 11.8
47 (Table 2, Fig. 1). The indigenous Pb_i isotope compositions of these samples were
48 therefore assumed to be intermediate between (and identical within error to) the
49 measured Pb_s values (which implies no contamination with $Pb_i = Pb_s$) and the less

50 radiogenic results obtained for Allende NHM CV3 and EET 92042 CR2,
51 respectively (Table 2).

52 As an example, the indigenous $^{206}\text{Pb}/^{204}\text{Pb}_{i \text{ All-Smith}}$ ratio of Allende Smithsonian was
53 thus obtained as follows. The difference in measured $^{206}\text{Pb}/^{204}\text{Pb}_s$ between Allende
54 Smithsonian and Allende NHM is given by

$$55 \quad \Delta^{206}\text{Pb}/^{204}\text{Pb} = ^{206}\text{Pb}/^{204}\text{Pb}_{s \text{ All-Smith}} - ^{206}\text{Pb}/^{204}\text{Pb}_{s \text{ All-NHM}} \quad (\text{S3})$$

56 With this

$$57 \quad ^{206}\text{Pb}/^{204}\text{Pb}_{i \text{ All-Smith}} = ^{206}\text{Pb}/^{204}\text{Pb}_{s \text{ All-NHM}} + (\Delta^{206}\text{Pb}/^{204}\text{Pb})/2 \quad (\text{S4})$$

58 This estimate of $^{206}\text{Pb}/^{204}\text{Pb}_{i \text{ All-Smith}}$ was then assigned an uncertainty of
59 $\pm(\Delta^{206}\text{Pb}/^{204}\text{Pb})/2$.

60 Similar calculations were then performed to determine the remaining isotope ratios
61 (and uncertainties) of $\text{Pb}_{i \text{ All-Smith}}$ and the isotope composition of $\text{Pb}_{i \text{ EET}}$.

62 2. If terrestrial Pb contamination is indeed correctly described by two-component mixing
63 between Pb_i and Pb_t , then the measured Pb_s isotope ratios of the samples should fall on
64 appropriate mixing lines that connect Pb_i and Pb_t , in diagrams of $^{207}\text{Pb}/^{204}\text{Pb}$ vs.
65 $^{206}\text{Pb}/^{204}\text{Pb}$ and $^{208}\text{Pb}/^{204}\text{Pb}$ vs. $^{206}\text{Pb}/^{204}\text{Pb}$. This assumption was used to determine the
66 distinct Pb_t isotope composition for each sample, as this is given by the intersection of
67 (i) a tie line that connects and extends from the Pb isotope data for Pb_i and Pb_s with
68 (ii) the line that defines the permissible range of Pb isotope compositions for Pb_t
69 (Equations S1, S2; Göpel et al., 1985)

70 in plots of both $^{207}\text{Pb}/^{204}\text{Pb}$ vs. $^{206}\text{Pb}/^{204}\text{Pb}$ and $^{208}\text{Pb}/^{204}\text{Pb}$ vs. $^{206}\text{Pb}/^{204}\text{Pb}$. Two separate
71 (but consistent) mixing diagrams/relationships were thus evaluated to obtain $^{206}\text{Pb}/^{204}\text{Pb}_i$,
72 $^{207}\text{Pb}/^{204}\text{Pb}_i$, and $^{208}\text{Pb}/^{204}\text{Pb}_i$.

73 3. At this stage the Pb isotope compositions Pb_s , Pb_i , and Pb_t were all known for a sample.
74 The lever rule was then applied to calculate the mol fraction X_i of indigenous meteoritic

75 Pb_i that must be present in each sample, to account for the measured isotope composition
76 of Pb_s by mixing of Pb_i and Pb_t.

77 4. The calculated values of X_i were then employed to determine the indigenous Pb
78 concentrations [Pb]_i of the samples:

79 (i) The uncorrected Pb abundances [Pb]_s (which include terrestrial contamination) were
80 first determined with the ID technique, from the ²⁰⁸Pb/²⁰⁶Pb ratios that were
81 measured for the *spiked* sample aliquots. The ID equations employed the measured
82 ²⁰⁶Pb/²⁰⁴Pb_s, ²⁰⁷Pb/²⁰⁴Pb_s, and ²⁰⁸Pb/²⁰⁴Pb_s ratios (determined for the unspiked sample
83 aliquots) to define the natural Pb isotope compositions of the samples.

84 (ii) The [Pb]_i contents shown in Table 2 (main text) were then calculated as:

$$85 \quad [Pb]_i = [Pb]_s \times X_i \quad (S5)$$

86 The inferred [Pb]_i abundances were combined with the measured Tl contents for the
87 calculation of ²⁰⁴Pb/²⁰³Tl_i (Table 2).

88 5. The uncertainties that are quoted for [Pb]_i and ²⁰⁴Pb/²⁰³Tl_i (Table 2, main text) take into
89 account the uncertainty in the isotope compositions (i) of Pb_t, as defined by the errors of
90 the y-offset for Equations (S1) and (S2) (Göpel et al., 1985) and (ii) of Pb_i as applied for
91 the correction of the Allende Smithsonian and NWA 801 data (see step 1.ii. above).
92 These errors are combined with the uncertainties from the blank correction (Section
93 3.2.2, main text) and the mass spectrometric uncertainties of the Pb isotope compositions
94 determined for the spiked and unspiked sample aliquots (Sections 3.3.2 and 3.3.3, main
95 text) to derive the total uncertainties for [Pb]_i and ²⁰⁴Pb/²⁰³Tl_i (Table 2).

96

97 **Reference**

98 Göpel, C., Manhès, G., and Allègre, C. J., 1985. U-Pb systematics in iron meteorites:
99 Uniformity of primordial lead. *Geochim. Cosmochim. Acta* 49, 1681-1695.