# NEW EVIDENCE FROM CARBONACEOUS CHONDRITES FOR THE PRESENCE OF LIVE $^{205}$Pb IN THE EARLY SOLAR SYSTEM.

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**Introduction:** The nuclide $^{205}$Pb decays to $^{205}$Tl with a half-life of 15 Myrs. This decay system is of particular interest because $^{205}$Pb is the only short-lived radionuclide that is solely produced by the *s*-process and hence it provides unique constraints on the sites and timing of *s*-process nucleosynthesis [1]. The $^{205}$Pb-$^{205}$Tl decay system may furthermore be useful for dating volatile loss and planetary differentiation in the early solar system because Pb and Tl are highly volatile elements with different condensation temperatures and because Pb is somewhat more siderophile/chalcophile than Tl.

A number of astrophysical models have predicted that significant amounts of $^{205}$Pb were present in the early solar system [e.g. 2] but cosmochemical evidence that substantiated these predictions remained elusive for more than 40 years. The first conclusive evidence for the presence of live $^{205}$Pb in the early solar system was presented by Nielsen et al. [3], who obtained a $^{205}$Pb-$^{205}$Tl isochron for seven metal fragments from the IAB iron meteorites Toluca and Canyon Diablo (Fig. 1). Based on this isochron, Nielsen et al. [3] inferred initial solar system values of $^{205}Pb/^{204}Pb_{SS,0} \approx (1 - 2) \times 10^{-4}$ and $\varepsilon^{205}Tl_{SS,0} = -2.8 \pm 1.7$ ($\varepsilon^{205}$Tl is the relative deviation of the $^{205}Tl/^{203}Tl$ isotope ratio of a sample from the NIST SRM 997 Tl isotope standard in parts per $10^4$).

Nielsen et al. [3] also observed that troilites from the same iron meteorites (denoted by grey symbols in Fig. 1) did not conform to the isochron relationship defined by the metals. In studies of extinct radionuclides it has been standard practice to use the least radiogenic isotope ratio measured as the best approximation of the solar system initial. This would imply, however, that $\varepsilon^{205}Tl_{SS,0}$ may be as low as -20, which is difficult to reconcile with the IAB metal isochron.

The troilite data are hence more plausibly explained by the kinetic stable isotope fractionation that may occur during sub-solidus diffusion of Tl between metal and sulphide [3]. Thallium has only two stable isotopes, and it is thus difficult to distinguish stable from radiogenic isotope effects. In this study, we have therefore analysed volatile-rich carbonaceous chondrites to obtain independent constraints on the initial Tl isotope composition of the solar system.

**Methods:** Eight carbonaceous chondrites from 5 groups (CI, CM, CV, CO, CR) were analyzed in this study. About 150 to 1000 mg of meteorite were dissolved for analysis with HF-$HNO_3$-HCl using Parr digestion bombs. Separation of Tl and Pb was achieved by anion-exchange chromatography using procedures modified from [4].

All samples were analysed for Tl and Pb isotope compositions and concentrations by multiple collector ICP-MS using external normalization to an admixed element for mass bias correction [4]. The Pb isotope compositions of the meteorites were determined primarily to correct the measured $^{204}$Pb concentrations for terrestrial contamination.

All Tl and Pb concentrations will eventually be determined at high precision by isotope dilution. Here we report only preliminary results for $^{204}Pb/^{203}Tl$ and some of these data were acquired by comparing the measured ion beam intensities for samples with those obtained for standard solutions of known concentration. These results have an estimated accuracy of ±30%, based on a comparison with isotope dilution data.

**Results**: An evaluation of the Pb isotope results with methods modified from [3] indicate that terrestrial contamination contributes less than 15% to the total measured Pb concentrations of five chondrites. More pervasive Pb contamination was determined for the chondrites NWA 801 (~30% terrestrial Pb), Cold Bokkeveld (~40%) and Colony (~90%).

Eight Tl isotope analyses of individual powder splits of the CV3 chondrite Allende yielded an average result of $\varepsilon^{205}Tl = -3.1 \pm 0.5$ (all uncertainties given are 2sd). Based on this, we have adopted an uncertainty of ±0.5 ε-units for all Tl isotope data reported in this study.

Five of the meteorites, including Cold Bokkeveld (CM2), Leoville (CV3), Kainsaz (CO3), Murchison (CM2), and Orgueil (CI1), display a very limited range of Tl isotope compositions of between $\varepsilon^{205}Tl = -1.7$ for Murchison and +0.7 for Kainsaz. These results are essentially identical within error and they yield an average value of $\varepsilon^{205}Tl = -1.3 \pm 0.8$.

The CR2 chondrite NWA 801 exhibits the most unradiogenic isotope ratio of $\varepsilon^{205}Tl = -4.0$, whereas Colony (CO3) displays the heaviest Tl isotope composition with $\varepsilon^{205}Tl = +1.2$. As the latter sample is highly weathered [5] and characterized by the most severe Pb contamination, it is possible that its unusual Tl isotope signature may reflect terrestrial processes. We will hence not consider this sample in the following discussion.

The preliminary concentration data indicate that the $^{204}Pb/^{203}Tl$ displays only limited variation in carbonaceous chondrites, with results that vary between 0.56 ± 0.06 for NWA 801 and 1.45 ± 0.02 for Murchison.

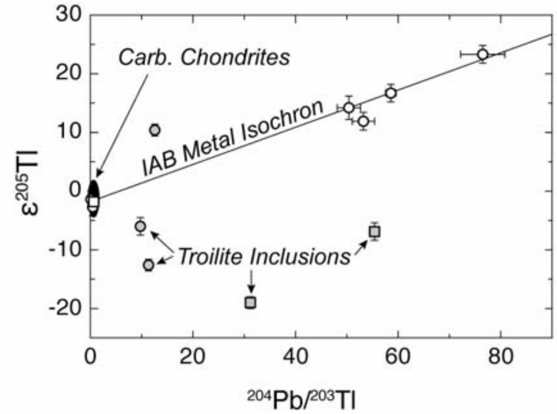


Fig. 1. Isochron diagram for the extinct $^{205}Pb$-$^{205}Tl$ decay system with previously published results [3] for metal and sulfide samples (open and grey symbols, respectively) of the IAB irons Canyon Diablo (squares) and Toluca (circles). The new data for carbonaceous chondrites plots within the small black field.

**Discussion:** The new data that were acquired for carbonaceous chondrites do not display a straightforward correlation in a $^{205}Pb$-$^{205}Tl$ isochron diagram (Fig. 1). Hence there is no isochron that provides a straightforward estimate of the initial Tl isotope composition of the solar system.

A robust estimate of $\varepsilon^{205}Tl_{SS,0}$ can nonetheless be extracted from the results because the carbonaceous chondrites exhibit ubiquitously low Pb/Tl ratios (Fig. 1). Simple modeling to estimate the effects of radiogenic ingrowth of $^{205}Tl$ for the chondrites utilised the measured $^{204}Pb/^{203}Tl$ ratios of the samples, and an initial $^{205}Pb$ abundance of $^{205}Pb/^{204}Pb_{SS,0} \approx (1 - 2) \times 10^{-4}$, as inferred from the IAB isochron [3]. It was furthermore assumed that the meteorite parent bodies may have formed up to 5 Myr after CAI's [6].

With these parameters, it can be inferred that radiogenic ingrowth is only able to increase the measured Tl isotope compositions of the carbonaceous chondrites by less than 0.4 ε-units, which is smaller than the uncertainty of the analytical data. The effect of radiogenic ingrowth will be less then 1 ε-unit even for an unrealistically high value of $^{205}Pb/^{204}Pb_{SS,0} = 7 \times 10^{-4}$. Based on the modeling, the chondrite results indicate an initial solar system Tl isotope composition of $\varepsilon^{205}Tl_{SS,0} = -2.5 \pm 1.5$, which is essentially identical to the value inferred from the IAB metal isochron [3].

The observation that the Pb-Tl data obtained for carbonaceous chondrites are not in accord with a simple isochronous relationship most likely reflects either nucleosynthetic isotope anomalies or stable isotope fractionations that are associated with late-stage mobilisation of Tl (and Pb).

It is conceivable that the small disturbances of the Pb-Tl systematics of carbonaceous chondrites reflect incomplete mixing of distinct nucleosynthetic reservoirs with different initial $^{205}Pb$ abundances or Tl isotope compositions. We consider this to be an unlikely scenario, however, because both $^{203}Tl$ and $^{205}Tl$ are predominantly *s*-process nuclides that are not expected to exhibit nucleosynthetic isotope anomalies [7]. It is furthermore significant that other moderately and highly volatile elements such as Zn, Cd and Te [8, 9, 10] do not display evidence of nucleosynthetic isotope anomalies in bulk carbonaceous chondrites.

Hence, it is more likely that the minor disturbances of the Pb-Tl systematics of carbonaceous chondrites reflect secondary stable isotope effects. Stable isotope fractionations generate Tl isotope variations of 30 ε-units in terrestrial environments [11] and the large offsets between sulphides and metal samples from IAB iron meteorites (Fig. 1) are also attributed to such processes. Recent analyses have furthermore revealed significant Cd isotope variability amongst some groups of carbonaceous chondrites (particularly CV and CO chondrites) that appears to reflect isotope fractionation by parent body processes [12]. This indicates that the observed Tl isotope variations are most likely due to stable isotope fractionations that were generated by either partial volatilisation/evaporation or aqueous alteration on the chondrite parent bodies. The same processes may also fractionate the elemental Pb/Tl ratios of the chondrite samples and produce additional scatter in the data.

**Conclusion:** The new Tl isotope results for carbonaceous chondrites strongly support the conclusion that the IAB iron meteorite isochron (Fig. 1) was indeed generated by *in situ* decay of $^{205}Pb$ to $^{205}Tl$. The small deviations of the chondrite data from an isochron relationship are thought to reflect secondary stable isotope fractionation processes on the meteorite parent bodies.

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