LEAD-THALLIUM CHRONOLOGY OF IIAB AND IIIAB IRON METEORITES AND THE SOLAR SYSTEM INITIAL ABUNDANCE OF LEAD-205. R. Andreasen[1,2], M. Rehkämper[1], G. K. Benedix[3], K. J. Theis[4], M. Schönbächler[4], C. L. Smith[3], [1]Department of Earth Science and Engineering, Imperial College London, Prince Consort Road, London SW7 2AZ, United Kingdom, [2]Department of Earth and Atmospheric Science, University of Houston, 312 SR1, Houston TX 77204, USA (randreas@central.uh.edu), [3]Department of Mineralogy, Natural History Museum, Cromwell Road, London SW7 5BD, United Kingdom, [4]School of Earth, Atmospheric, and Environmental Sciences, University of Manchester, Oxford Road, Manchester M13 9PL, United Kingdom.

Introduction: The short-lived ^{205}Pb-^{205}Tl decay system ($t_{½}$= 15.1 My) is the only radioactive decay system in which the parent nuclide (^{205}Pb) is an s-process only nuclide, and thus the Solar System initial abundance of ^{205}Pb provides unique constraints on the amount of AGB star material that was injected into the Solar Nebula shortly before its collapse. The Pb-Tl system is furthermore one of few decay systems that can be used to date metal crystallization and hence provide independent age constraints on the core crystallization and thermal history of planetary bodies. Utilization of the Pb-Tl system is complicated by the fact that neither parent nor daughter element has a stable isotope ratio that can be used for internal mass bias correction. In addition natural variations Tl isotope composition are small—a few tens of ε-units or parts per ten thousand. However, the use of Multi-Collector Inductively Coupled Plasma Mass Spectrometry (MC-ICP-MS) and external normalization negate these issues, and evidence of the presence of live ^{205}Pb in the early Solar System has been found in non-magmatic IAB iron meteorites [1] and carbonaceous chondrites [2]. Another issue is the widespread contamination of meteorites with anthropogenic Pb, which is difficult to correct for due to the large spread in terrestrial Pb isotope ratios. This makes it especially difficult to correct for terrestrial Pb contamination in samples with low Pb concentrations and samples where the initial Pb isotope composition is not well known.

Sample preparation: Ten IIAB iron meteorites (including 5 separate samples of Sikhote-Alin) and 6 IIIAB iron meteorites were analyzed. In order to minimize the problem of terrestrial lead contamination all sawn surfaces were polished with silicon carbide paper. Repeated leaching with 0.5 M HBr was found to be effective for removing fusion crust. This was followed by more aggressive leaching first with 6 M HCl and finally with Aqua Regia. The leaching procedure removed between a quarter and a third of the sample and removed the entire preexisting surface. Following leaching, the samples were dried, weighed, and dissolved in Aqua Regia. Spiked Pb and Tl concentration splits were measured by MC-ICP-MS, using added Pt to correct for instrumental mass bias. Pb and Tl isotope compositions were measured by MC-ICP-MS. NBS 981 Pb was added to the Tl samples, and NBS 997 Tl was added to the Pb samples to correct for instrumental mass fractionation.

Results: Measured Pb concentrations vary widely—from 0.5 ppb to more than 1 ppm due to terrestrial Pb contamination, which was not entirely removed despite repeated leaching. The fraction of primordial Pb can be calculated by assuming that any deviation in the Pb isotope composition of the iron meteorites from that of primordial Pb is due to terrestrial Pb contamination. The concentration range of primordial Pb is very limited—from 0.1 ppb to 1.7 ppb. Thallium concentrations range from 2 ppt to 485 ppt, but most samples have less than 20 ppt Tl. This results in ^{204}Pb/^{203}Tl ratios for the IIABs from 0.05 to 5.8 and from 1.6 to 14 for the IIIABs, close to the chondritic ratio of 1.4 [2] and showing much less variation than the range of 0.1 to 76 seen in non-magmatic IAB irons [1].

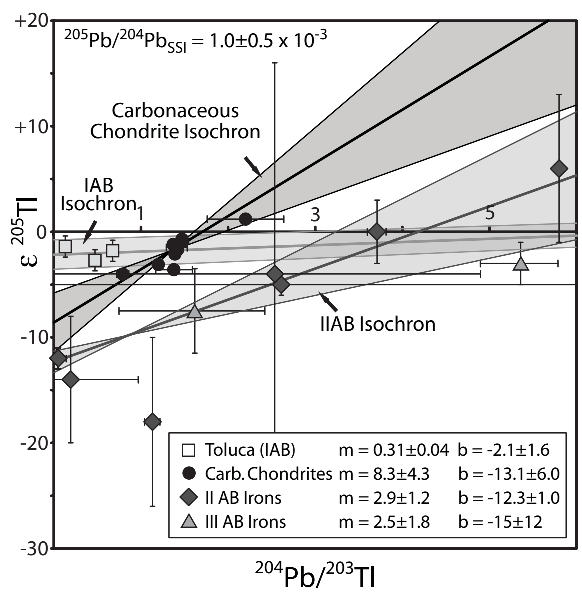

Figure 1: Pb-Tl isochrons for IAB irons [1], carbonaceous chondrites [2] and IIAB irons. Shaded areas indicate the uncertainty in the regression. Only iron meteorites with a ^{204}Pb/^{203}Tl ratio less than 6 are shown for clarity.

Values of ε^{205}Tl range from -18 to +23 and correlate with ^{204}Pb/^{203}Tl ratios (Fig. 1) suggesting that the variations in ε^{205}Tl are the result of decay of ^{205}Pb at different Pb/Tl ratios.

Interpretation: If the relationship between ε^{205}Tl and ^{204}Pb/^{203}Tl for the IIAB iron meteorites is interpreted as an isochron, this gives an age of 4^{+16}_{-11} Ma after the carbonaceous chondrites. The IIIAB irons give a similar but less well-constrained age of 4^{+29}_{-14} Ma. These are very close to the Hf-W model ages of metal/silicate separation [3], suggesting rapid cooling of the IIAB and IIIAB irons. The IIAB and IIIAB isochrons have intercepts that are more negative than that of the carbonaceous chondrites (Fig. 1) with an $\varepsilon^{205}Tl_0$ = -12±1 for the IIAB isochron compared to $\varepsilon^{205}Tl_0$ = -7.6±2.1 for that of the carbonaceous chondrites [2]. This could imply that the metal/silicate separation of the IIAB parent body was associated with mass dependent Tl isotope fractionation of about -7 ε-units; with the silicates becoming correspondingly enriched in ^{205}Tl. However, experimental data [4] suggest that a fractionation of this magnitude is not likely. Alternatively, if the carbonaceous chondrite isochron is recalculated rejecting the 3 samples that suffer from large terrestrial Pb contamination or are desert finds, the slope of ε^{205}Tl vs. ^{204}Pb/^{203}Tl for the remaining 8 samples is significantly steeper with an $\varepsilon^{205}Tl_0$ = -13±6 which intersects the IIAB isochron (Fig. 2). This increases the age difference between the carbonaceous chondrites and the iron meteorites by about 11 My and also increases the uncertainties due to the limited spread in ^{204}Pb/^{203}Tl ratios for the carbonaceous chondrites. The IIAB and IIIAB isochrons now give ages of 15^{+20}_{-12} Ma and 14^{+32}_{-15} Ma, respectively, after the carbonaceous chondrites. Likewise the IAB isochron gives an age of 69^{+16}_{-10} Ma after the carbonaceous chondrites, younger than silicate inclusion [5] and Pd-Ag [6] ages of around 15 Ma after Allende CAI, suggesting that the Pb-Tl for the IAB iron meteorites record a later lower-temperature impact event, perhaps the same event recorded in the ~4.5 Ga Ar-Ar ages [7]. It is not known if the Pb-Tl ages of the IIAB and IIIAB magmatic irons represent core crystallization ages or whether they might represent the time of break-up of their respective parent bodies. The IIAB isochron intersects the carbonaceous chondrite isochron at ^{204}Pb/^{203}Tl of 0.15. This indicates that metal-silicate segregation took place 2^{+16}_{-10} My after the closure of the Pb-Tl system in the carbonaceous chondrites, and this, though poorly-constrained, agrees well with Hf-W ages [3]. It also indicates that the metal portion of the IIAB parent body had a very low Pb/Tl ratio, with a ^{204}Pb/^{203}Tl ratio of ~0.15. This appears to be in agreement with the suggestion that the initial metal composition of the IIAB irons was sulfur rich [8] and with existing experimental partitioning data [4] that suggest a sulfur-rich metal phase should be significantly enriched in Tl compared to Pb.

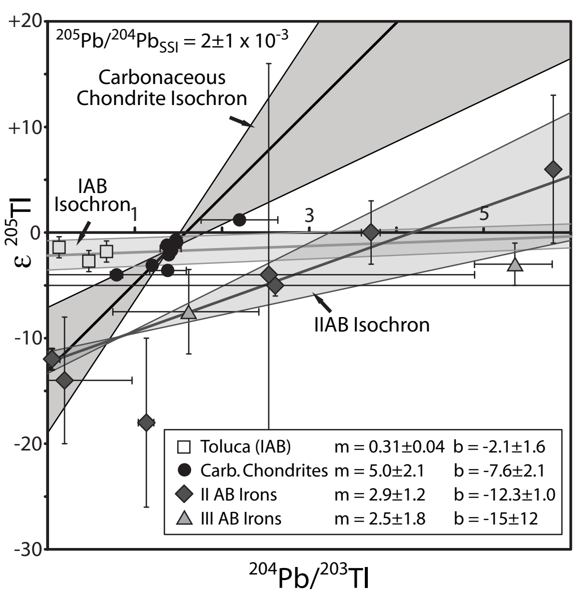

Figure 2: As figure 1. The carbonaceous chondrite isochron has been recalculated by excluding terrestrially contaminated samples.

Solar System Initial Abundance of ^{205}Pb. If the carbonaceous chondrite isochron is reinterpreted without the most contaminated samples so that it intersects the isochrons of the IIAB and IIIAB irons, the calculated Solar System initial ^{205}Pb/^{204}Pb increases from 1±0.5 x 10^{-3} [2] to 2±1 x 10^{-3}, somewhat higher than the upper estimate of 5.3 x 10^{-4} based on stellar models [9]. Further work is needed to establish whether this is reasonable, and if it is, whether it suggests (i) a higher proportion of recent AGB material was injected into the Solar Nebula, or (ii) a higher proportion of ^{205}Pb surviving in the surface layer of AGB stars than the stellar models predict.

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