OMSIUM ISOTPIC ANOMALIES OF INSOLUBLE ORGANIC MATTER IN CHONDRITES. T. Yokoyama[1, 2] C. M. O'D. Alexander[3] and R. J. Walker[1], [1]Dept. of Geology, Univ. of Maryland, College Park, MD 20742, USA (yokoyama@geol.umd.edu), [2]Dept. of Earth & Planetary Sciences, Tokyo Institute of Technology, Tokyo 152-8551, Japan, [3]Dept. of Terrestrial Magnetism, Carnegie Institution of Washington, DC 20015, USA.

Introduction: A variety of isotopic anomalies have been found in unequilibrated chondrites by means of ultra high-precision mass spectrometry analyses (e.g., [1-5]). Some of these anomalies are evidently due to the presence of variable proportions of presolar grains in the primitive chondrites, although heterogeneous isotope distribution in the early solar nebula is an alternative explanation for such observations. We have previously conducted high precision Os isotopic analysis of bulk chondrites plus their acid resistant fractions, and demonstrated that acid residues of some carbonaceous chondrites are enriched in Os isotopes produced by s-process nucleosynthesis, whereas bulk chondrites have uniform $^{186}Os/^{189}Os$, $^{188}Os/^{189}Os$ and $^{190}Os/^{189}Os$ ratios [1]. These acid residues were prepared from bulk chondrites via repeated treatments with HF+HCl, and mainly consist of macromolecular, insoluble organic matter (IOM). The IOM in chondrites is thought to have originated either from the interstellar medium or other presolar processes [6]. The presolar origin for the IOM is suggested based on the relatively constant ratio between the IOM and presolar grains in chondrites [7]. In fact, the separated IOM fractions are enriched in presolar circumstellar grains such as nanodiamonds and silicon carbides, which are presumably the sources of the Os isotope anomalies observed in [1-3]. However, the genetic linkage between the IOM and presolar grains is not clear, nor is the main carrier of the s-process Os. Here we present evidence for Os isotopic anomalies in the IOM fractions separated from eight chondrites, all of which are well characterized regarding their C, H, O and N contents and isotopes [6]. This approach can potentially provide detailed information about presolar nucleosynthesis as well as subsequent processes in the interstellar medium.

Experimental: The IOM fractions were prepared from seven carbonaceous chondrites (Orgueil, Ivuna, EET92042, Tagish Lake, Murchison, Allende and Leoville), and one ordinary chondrite (QUE97008) using a CsF-HF technique [8-9]. The IOM was put into a quartz Carius tube and combusted at 1000°C for 24 hours. The top of the tube was broken, followed by the addition of aqua regia. The tube was re-sealed and heated at 220°C overnight, then Os was extracted by CCl_4 and purified by a microdistillation technique.

All the Os isotope analyses were carried out by a TIMS (Thermo-Fisher TRITON) at the University of Maryland. Replicate analyses of a standard material (Johnson Matthey Os) agree well with our previous standard runs using the TRITON at the Carnegie Institution of Washington [1]. Osmium isotopes in bulk meteorites of three carbonaceous (Ivuna, Murchison and Allende) and one enstatite (Indarch) chondrites decomposed by an alkaline fusion method were also measured by the UMD TRITON. The bulk analyses yielded uniform Os isotope ratios which are identical to the previously determined 'solar values' [1]. All isotope data were normalized to ^{189}Os and corrected for mass fractionation using $^{192}Os/^{189}Os = 2.527411$. The $^{186}Os/^{189}Os$ ratio was time-corrected for ^{190}Pt decay over 4.56 Ga. The Os isotope ratios are reported in εOs units ($\varepsilon^{186}Os^{i}$, $\varepsilon^{188}Os$ and $\varepsilon^{190}Os$) which represent relative deviation from the presumed solar values.

Results and Discussion: Osmium isotope data for the IOM fractions are plotted on $\varepsilon^{186}Os^{i}$–$\varepsilon^{188}Os$ and $\varepsilon^{190}Os$–$\varepsilon^{188}Os$ diagrams (**Fig. 1**). All but one sample are characterized by positive Os isotope anomalies which are resolvable from the solar. The largest positive anomalies are found in the IOM from CI1 chondrites (Ivuna and Orgueil), of which εOs values are more than twice as large as C2 chondrites (Tagish Lake, Murchison and EET92042). The IOM from an oxidized CV3 (Allende) and an ordinary chondrite (QUE97008: L3.05) have marginal positive anomalies, whereas that from a reduced CV3 (Leoville) has negative anomalies. A noteworthy feature is that all the data are plotted on the chondrite regression lines defined in [1], the lines which represent mixing between the solar component and presumed s-process component. This indicates a common origin for s-process-enriched component in all types of chondrites.

It is also notable that the extent of s-process enrichment in the IOM is in the order of petrologic grade of the host rocks (type1> type 2> type 3). In chondrites, the IOM and presolar grains are concentrated in their matrix of which the abundance varies from ~100% in CIs down to ~13% in ordinary chondrites. The other components (chondrules + variable CAIs) are considered to have homogeneous, solar Os isotope composition. Thus, acid residues from ordinary chondrites would be more enriched in the solar component than those from CIs. Plus, chondrites of higher petrologic grade have relatively low abundances of presolar grains as a result of higher metamorphism in the parent body [10]. Combined, our observation might simply indicate a presolar origin for the s-process Os carrier in the chondrites. However, it was reported in [6] that the IOM from CR chondrites have higher δD and $\delta^{15}N$, and lower $\delta^{13}C$ than the other carbonaceous, enstatite and ordinary chondrites (excluding the anomalous CM2, Bells), presumably suggesting the preservation of primitive signatures in CR-IOM. If modification of δD, $\delta^{15}N$ and $\delta^{13}C$ values in the IOM is caused by parent-body processes which can also destroy presolar grains, it might be expected that the largest Os isotope anomalies in the IOM would be present in CR chondrites. This does not appear to be the case based on our observations. Either there is no linkage between differences in δD- $\delta^{15}N$- $\delta^{13}C$ values and Os isotope anomalies for the IOM from different chondrite groups, or the parent-body processes have selectively destroyed presolar grains enriched in r-process Os.

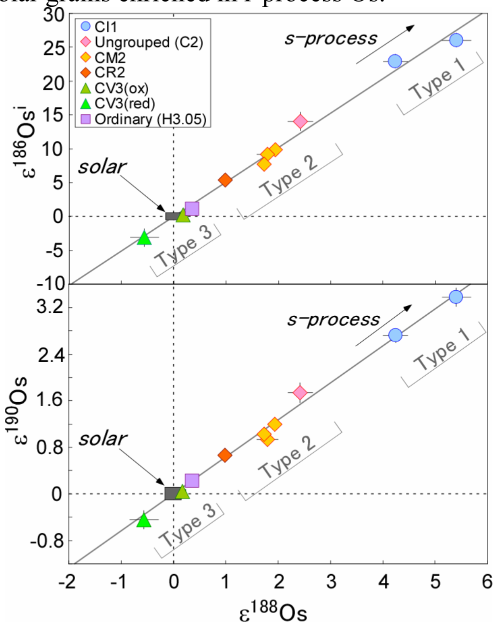

Fig. 1. $\varepsilon^{186}Os^i$-$\varepsilon^{188}Os$ and $\varepsilon^{190}Os$-$\varepsilon^{188}Os$ plots for IOM fractions from chondrites. Bold lines are the chondrite regression lines defined in [1], representing mixing lines between solar component and presumed s-process component. Gray boxes labeled solar represent the 2σ reproducibility of 13 bulk chondrites.

In order to further investigate the possible s- and r-process Os carrier in chondrites, we have carried out leaching experiments for three bulk chondrites (Ivuna, Murchison and Allende) and two IOM fractions (Murchison and Allende). The bulk rock powders were leached with 2M HCl in room temperature for several days, of which the leachate is denoted as 'CHLL'. The IOM fractions were leached with 6M HCl at ~70°C for two days. The leachate and residue after this process are called as 'HHLL' and 'HHLR', respectively. Osmium isotope data for these samples are shown in **Fig. 2**. All the leachate samples show negative Os isotope anomalies, indicating the enrichment of r-process Os components in these fractions. The magnitude of the negative anomalies are again in the order of the petrologic grade. The HHLR samples plot close to their original IOM compositions before leaching, because mass balance calculations show <10% of Os was liberated via 70°C HCl leaching. It should be noted that, although analytical errors are large, the Murchison HHLL and Ivuna CHLL depart from the chondritic regression line in the $\varepsilon^{186}Os^i$–$\varepsilon^{188}Os$ plot. The HHLL samples are known to have ^{54}Cr anomalies, an isotope that can be produced by r-process nucleosynthesis [11]. The deviance of the HHLL data from the chondrite regression line suggests the existence of an r-process Os carrier in the chondrite which has been produced via a stellar process differed from that generated most of the r-process Os in chondrites.

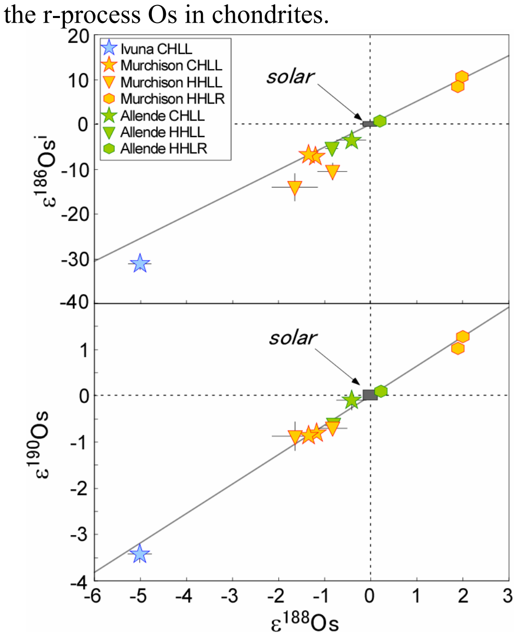

Fig. 2. $\varepsilon^{186}Os^i$-$\varepsilon^{188}Os$-$\varepsilon^{190}Os$ data for leaching experiments.

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