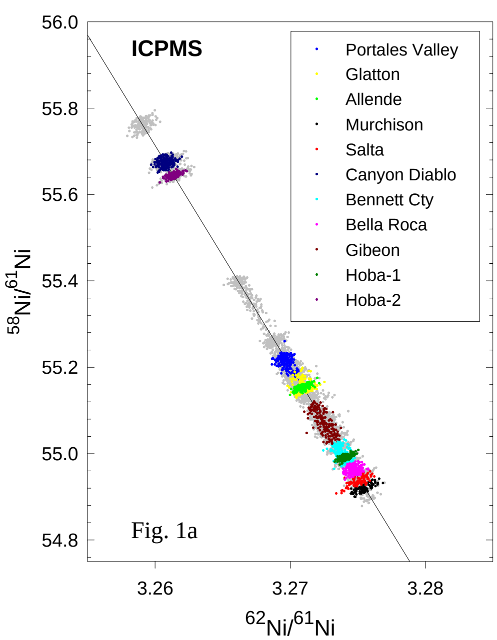
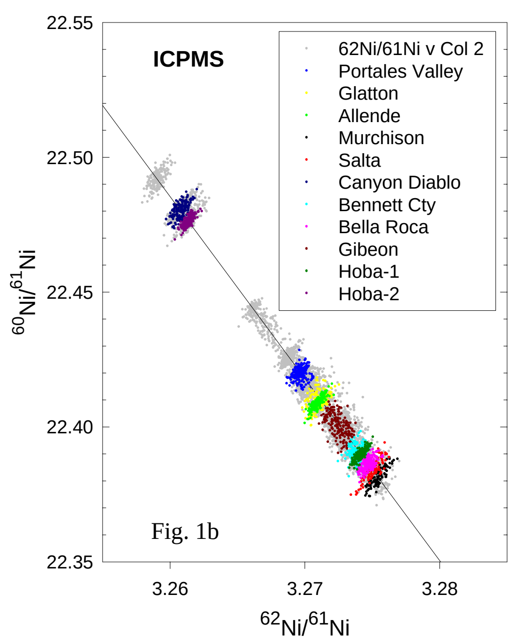
NICKEL ISOTOPE INVESTIGATION BY MC-ICP-MS AND PTIMS. J. H. Chen[1], D. A. Papanastassiou[2], Science Division, [1]M/S 183-601, [2]M/S 183-335, Jet Propulsion Laboratory, Caltech, 4800 Oak Grove Dr., Pasadena, CA 91109-8099 (James.H.Chen@jpl.nasa.gov).

Introduction: The evidence for the presence of short-lived ^{60}Fe (decaying into ^{60}Ni, with the half life of 1.5 Ma) in the early system has recently received considerable attention, both experimentally and theoretically. Evidence for ^{60}Ni excesses in eucrites, without correlation to Fe/Ni has been obtained by Shukolyukov and Lugmair [1, 2]. Recently, based on ion microprobe studies of phases with very high Fe/Ni, direct evidence for the *in situ* decay of ^{60}Fe has been obtained [3-5]. These studies indicate a significant initial abundance of $^{60}Fe/^{56}Fe$, in the solar system ($1.1\pm0.2\times10^{-7}$ and $1.7\pm0.5\times10^{-7}$ for Bishunpur and Krymka [3] and $7\pm3\times10^{-7}$ for Semarkona [4, 5]). An extensive recent review [6] of the abundances of short-lived nuclides in the early solar system and their astrophysical production sites and rates concludes that the initial $^{60}Fe/^{56}Fe$ by an AGB source would be significant, at 10^{-7} to 2×10^{-6}, for a dilution factor of $\sim4\times10^{-3}$ (the ratio of the contaminating mass to the solar parental cloud) [6]. A supernova can be the source or ^{53}Mn and possibly of ^{60}Fe [6]. Tachibana *et al.* [7] have determined evidence for ^{60}Fe in ferromagnesian chondrules from Semarkona and Bishunpur, with initial $^{60}Fe/^{56}Fe$ of $(2.2\text{-}3.7)\times10^{-7}$. In addition, early data on Ni in Allende refractory inclusions indicated the presence of general Ni isotope anomalies [8], although mass interference from Zn affected ^{64}Ni, and interferences at other Ni isotopes could not be completely eliminated.

Results: We have developed analytical techniques for the measurement of Ni isotopes by MC-ICP-MS (using the Thermo/ Finnigan Neptune, at Caltech) and by TIMS (the Thermo/Finnigan Triton, at JPL). The advantage of MC-ICP-MS rests with high ionization efficiency in the plasma source (coupled with improvements in sample transmission efficiency, based on the use of a desolvating nebulizer (ESI ApexAreo), which also reduces oxides and some molecular interferences. The disadvantages lie dominantly in potentially significant mass interferences, precisely due to the high ionization efficiency for all elements and the presence of molecular ion species. For ICP-MS, interferences at mass 58 arise from ^{58}Fe and from $^{40}Ar^{18}O$. The presence of dominant $^{40}Ar^{16}O$ does not permit the adequate monitoring of ^{56}Fe interference. To minimize the effects of molecular interferences, we obtain data under *intermediate* mass resolution and by centering the magnetic field to the narrow mass region on the peak tops, where the molecular ion beams are occluded by the collector slits [9, 10]. The Ni in the samples is chemically separated from Fe by ion exchange. The Ni solution concentrations for runs are at the level of a few ppm, which yields over 10^{-10} Amps, at intermediate mass resolution, at ~50 μL/min. We have also reduced significant mass interference from Zn (affecting ^{64}Ni), by identifying its source as the purified water, produced by distillation at Caltech. Water produced through mixed-bed ion exchange and recirculation, at JPL, is considerably more Zn-free. But, the interference on ^{64}Ni was still significant,>1‰, for both the ICP-MS and TIMS measurements. We are not reporting ^{64}Ni data. Chemical clean-up of Zn has now been achieved, but not in time for this abstract. Given





the known mass interferen-ces, it is safer for MC-ICP-MS data to be normalized for mass fractionation using $^{62}Ni/^{61}Ni$. Use of these lower abundance isotopes results in some amplification of uncertainties. We show all the data for Ni standards, obtained during the development of the techniques. The raw (uncorrected) ICP-MS ratios for the Ni standards (grey points, Fig. 1), obtained during the shorter period of the meteorite measurements, show a good correlation and a small range of instrumental fractionation of 0.5% in $^{62}Ni/^{61}Ni$. All meteorite samples (colored points, Fig. 1), analyzed during the same period of time, fall within this range. We report data on iron meteorites (Groups IAB, IIAB, IVAB), a pallasite, chondrites, and whole-rock samples of Allende and Murchison. Linear fits to the ratios for Ni standards are shown in Fig. 1. The deviations (in parts in 10^4) of the meteorite data from these lines for standards are shown in Fig. 2. The uncertainties are $2\sigma_{mean}$ (for 90-270 ratios) for each sample. For the chondrites (Portales Valley, Glatton and Murchison), pallasite (Salta) and irons (Canyon Diablo, Bennett County, Bella Roca, Gibeon and Hoba), the $\varepsilon^{58}Ni$ and $\varepsilon^{60}Ni$ values are all within about $\pm1\varepsilon$ of the normal values. However, the Allende whole rock (three analyses of the same solution) show deficits in $^{60}Ni/^{61}Ni$ (up to -2.6 ± 0.3 εu), which we consider to be a preliminary determination. Given our TIMS developments, for Ni, it will be possible to check on the ICP-MS data validity for this sample. TIMS measurements on ^{64}Ni, with improved Zn separation will also help identify the presence of further general isotope anomalies for Ni. The earlier data on Allende CAI, by Birck and Lugmair [8] do not show strong evidence for ^{60}Ni effects, when converted to our normalization convention for TIMS data, of $^{58}Ni/^{62}Ni$ (see below).

For TIMS we have developed improved ionization techniques for Ni and have reduced Fe ($^{58}Fe/^{58}Ni<10^{-7}$) and Zn ($^{64}Zn/^{64}Ni<1x10^{-3}$) mass interferences. For TIMS we use the preferred normalization for isotope fractionation to $^{58}Ni/^{62}Ni$, since there are no Fe and no molecular interferences for this ratio. We have obtained data on Ni normals and on Ni from the same iron meteorites, analyzed by ICP-MS. The Ni data using TIMS, shown in Fig. 3, indicate an improved precision (2σ) of $<0.5\varepsilon$ for $^{60}Ni/^{62}Ni$ and $\pm1.2\varepsilon$ for the low abundances $^{61}Ni/^{62}Ni$. Both ratios are the same as for the terrestrial normal values within uncertainties. Work on chondrites is in progress. This work shows good agreement between ICP-MS and TIMS and improved precision. The MC-ICP-MS effects in ^{60}Ni, in Allende require confirmation by TIMS.

References: [1] Shukolyukov A & Lugmair G. W. *Science 259*, (1993) 1138; [2] – *EPSL 119* (1993) 159; [3] Tachibana S. & Huss G. R. *Ap. J. 588* (2003) L41; [4] Mostefaoui S., Lugmair G. W., Hoppe P., & El Goresy A. *LPSC 34*, (2004) #1585; [5] -- *New Astron. Rev. 48* (2004) 155; [6] Wasserburg G. J., Busso M., Gallino R., & Nollett K. M. *Nucl. Phys A* (2006) in press; [7] Tachibana S., Huss G. R., Kita N. T., Shimoda G., & Morishita Y. *Ap. J. Lett.* (2006), in press; [8] Birck J.-L. & Lugmair G. W. *EPSL 90* (1988) 131; [9] Weyer S. & Schwieters J. *Int. J. Mass Spect. 226* (2003) 355; [10] Arnold G. L. et al. *Anal. Chem. 76* (2004) 322.

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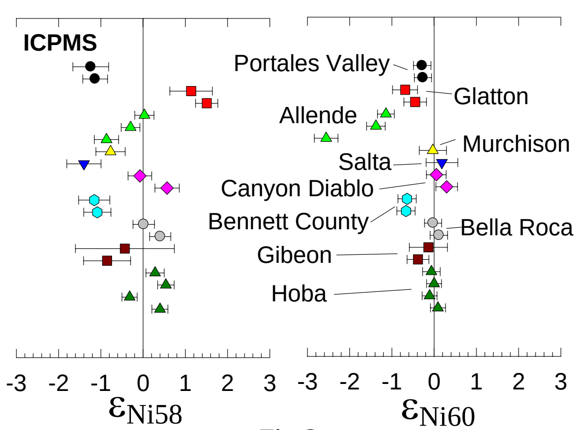

Fig. 2

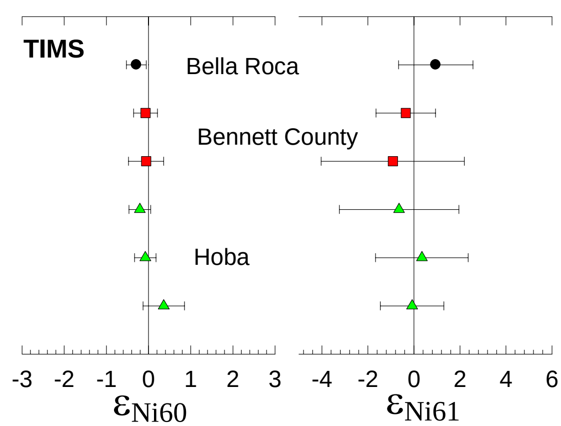

Fig. 3