NEUTRON-POOR Ni ISOTOPE ANOMALIES IN BULK METEORITES AND THEIR NUCLEOSYNTHETIC SIGNIFICANCE.
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Introduction: The Solar System shows input from many different nucleosynthetic environments [1]. These environments can be investigated by study of the isotopic composition of primitive meteorites and their components, which are the only samples from the beginning of the Solar System (e.g. [2]). Isotopic anomalies, or deviations from mass-dependent fractionation, on the neutron rich isotopes of Ca and Ti [3, 4] in calcium, aluminium rich inclusions were taken as strong evidence of differential input to the early Solar System of highly neutron enriched material. The source of this neutron-rich material has been hypothesized to be type Ia supernova (SN Ia) (e.g. [5]), type II supernova (SN II) (e.g. [6]) or wind from an asymptotic giant branch star (AGB) (e.g. [7]). Subsequently, smaller anomalies were found in bulk meteorites on the neutron rich isotopes of other elements (e.g. Cr [8], Ti [9]) and were taken as supporting evidence of the hypothesis of input from a neutron rich environment. However, many of these studies have employed internal normalisation to a pair of lighter isotopes to remove mass-dependent fractionation and increase precision. These data are normally expressed as part per ten thousand (‱) difference relative to a terrestrial reference and in this study the normalising isotopes are noted in a subscript, e.g. $\varepsilon^{62}Ni_{\frac{58}{61}}$. This normalisation means that for some data the finding of neutron-rich isotope anomalies is a point of interpretation as the anomalies may reside on the neutron-poor normalising isotopes.

Nickel presents an interesting opportunity to study the nucleosynthetic origins of the Solar System. Due to its moderately siderophile and moderately refractory behavior Ni is of high abundance in most meteorites. Also Ni has five stable isotopes (^{58}Ni, ^{60}Ni, ^{61}Ni, ^{62}Ni, ^{64}Ni), of which the two heaviest are highly neutron-rich and have been hypothesised as being produced in the highly neutron-rich SN Ia [5, 6] and as such measurements of Ni isotopes may provide a good test of input from this environment. This is of particular interest because an SN Ia is an explosion of a small old star that is not thought to be associated with star forming regions.

Analysis: Nickel was separated with five stage ion exchange chromatography which utilized the highly Ni specific dimethylglyoxime [10, 11]. Isotope data were obtained on clean Ni fractions using two Thermo Finnigan Neptune multiple-collector inductively-coupled plasma mass spectrometers, one for double spike and one for internally normalized data. Full analytical details are reported in [11] for mass-independent and [12] for double spike analysis.

Discussion and Results: Mass-independent Ni isotope data from a suite of chondritic and iron meteorites give a total range of 0.3 ‱ and 0.9 ‱ in $\varepsilon^{62}Ni_{\frac{58}{61}}$ and $\varepsilon^{64}Ni_{\frac{58}{61}}$ respectively. There is a strong positive correlation between $\varepsilon^{62}Ni_{\frac{58}{61}}$ and $\varepsilon^{64}Ni_{\frac{58}{61}}$ with a slope of 2.98 ±0.21 (MSWD = 0.95, n = 215 repeats of 31 samples). As with other isotope systems (e.g. $\varepsilon^{50}Ti_{\frac{49}{47}}$ and $\varepsilon^{54}Cr_{\frac{52}{40}}$) carbonaceous chondrites have the highest ratios, ordinary chondrites have the lowest and enstatite chondrites have ratios around zero. Interestingly, only EH enstatite chondrites plot within error of terrestrial peridotites which are the best estimate of the bulk silicate Earth.

Strikingly, the slope obtained from these internally normalized data is within error of the slope expected from an anomaly located on ^{58}Ni (2.96). The likelihood of an analytical artifact on ^{58}Ni has been examined in detail and ruled out in a previous contribution [11], therefore, the possibility of anomalies on ^{58}Ni is in need of investigation. High precision mass-dependent Ni isotope ratios, obtained by double spike analysis, were combined with our mass-independent data to achieve 'absolute' Ni isotope compositions for two meteorites, Orgueil (CI) and Butsura (OC), which span the entire range in $\varepsilon^{62}Ni_{\frac{58}{61}}$ and $\varepsilon^{64}Ni_{\frac{58}{61}}$. These data show that the ^{62}Ni/^{61}Ni and ^{64}Ni/^{61}Ni absolute Ni isotope compositions of these two meteorites are within error, whereas those of ^{58}Ni/^{61}Ni show a clearly resolved difference of ~0.8 ‱, relative to an uncertainty of ~0.25 ‱. This is strong evidence that Ni isotope anomalies indeed reside on the neutron-poor isotope ^{58}Ni. This finding of neutron-poor anomalies in Ni has significant implications for the location of anomalies in other iron group elements. While it is possible that Ni uniquely samples a neutron-poor source, absolute ratios have not been determined in bulk samples for other elements which, therefore, may also exhibit neutron-poor isotope anomalies. This has implications for the source of the isotopically anomalous material sampled by bulk meteorites.

These Ni isotope data can be used to further investigate the nucleosynthetic sources of anomalous input to the Solar System by comparison with models of different nucleosynthetic environments, e.g. SN Ia [13], SN II [14] and AGB [15]. The criteria used to determine a successful model were matching the correct slope in $\varepsilon^{62}Ni_{\frac{58}{61}}$ vs. $\varepsilon^{64}Ni_{\frac{58}{61}}$ and the major anomaly being located on ^{58}Ni. We find that none of the models of bulk nucleosynthetic environments studied can provide a robust case as the source of Ni isotope anomalies in bulk meteorites.

An alternative to the bulk nucleosynthetic environments is that of individual shells an SN II as modelled by [16]. Fig. 1. shows the effects of mixing fractions each of the individual $2x10^{31}$ g shell of a 15 M⊙ modelled by [16] in to the Solar System. The Si/S zone of all masses of SN II modelled by [16] (15-40 M⊙) matches the slope in Ni isotopes by overproduction of ^{58}Ni. This suggests that the Ni isotope heterogeneity observed in the Solar System may be provided by the Si/S zone of an SN II which is of particular interest because the Si/S zone has been suggested as the source of the 'X-grain' population of pre-solar grains [17].

Conclusion: Nickel isotopes exhibit neutron-poor anomalies which has implications for the location of anomalies in other iron group elements. Comparison between the Ni isotope composition of meteorites and those modelled for different bulk nucleosynthetic environments shows none can provide a robust case as the source of Ni isotope heterogeneity in the Solar System. However, the Si/S zone from all masses of SN II matches the criteria set by measurements of bulk meteorites suggesting the Si/S zone may be the source of anomalous Ni in early Solar System materials.

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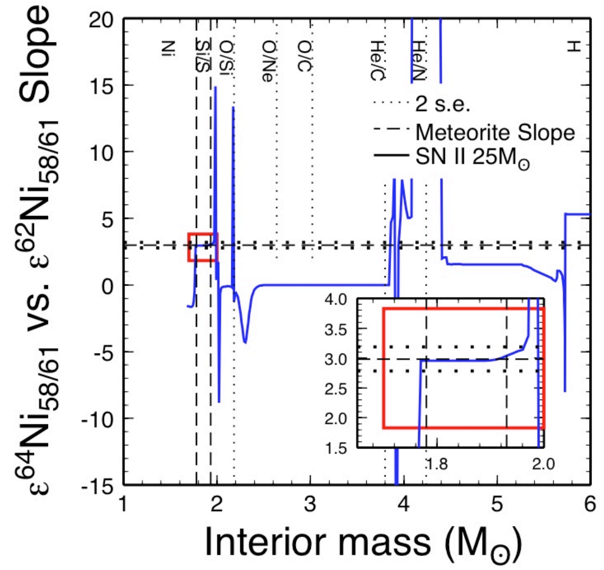

Fig. 1. Plot showing the effect of mixing fractions of individual $2x10^{31}$ g shells (modelled by [6]) into the Solar System on the slope of $\varepsilon^{62}Ni_{\frac{58}{61}}$ vs. $\varepsilon^{64}Ni_{\frac{58}{61}}$ (blue line). Successful solution are those where the blue line overlaps the observed slope of $\varepsilon^{62}Ni_{\frac{58}{61}}$ vs. $\varepsilon^{64}Ni_{\frac{58}{61}}$ for Solar System samples, namely a value of 2.98 ±0.21. Also shown are the names of the zones defined by the two most abundant elements produced (after [18])