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Excess ^{180}W in IIAB iron meteorites: Identification of cosmogenic, radiogenic, and nucleosynthetic components

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Abstract

The origin of ^{180}W excesses in iron meteorites has been a recently debated topic. Here, a suite of IIAB iron meteorites was studied in order to accurately determine the contribution from galactic cosmic rays (GCR) and from potential decay of ^{184}Os to measured excesses in the minor isotope ^{180}W . In addition to W isotopes, trace element concentrations (Re, Os, Ir, Pt, W) were determined on the same samples, as well as their cosmic ray exposure ages, using ^{36}Cl - ^{36}Ar systematics. These data were used in combination with an improved model of GCR effects on W isotopes to correct effects resulting from neutron capture and spallation reactions. After these corrections, the residual ^{180}W excesses correlate with Os/W ratios and indicate a clear contribution from ^{184}Os decay. A newly derived decay constant is equivalent to a half-life for ^{184}Os of $(3.38 \pm 2.13) \times 10^{13}$ a. Furthermore, when the data are plotted on an Os–W isochron diagram, the intercept ($\epsilon^{180}\text{W}_i = 0.63 \pm 0.35$) reveals that the IIAB parent body was characterized by a small initial nucleosynthetic excess in ^{180}W upon which radiogenic and GCR effects were superimposed. This is the first cogent evidence for *p*-process variability in W isotopes in early Solar System material.

Keywords

iron meteorites; tungsten isotopes; galactic cosmic rays; nucleosynthesis; ^{184}Os - ^{180}W systematics

1. Introduction

The nebular material (gas and dust) from which the Solar System formed was contributed by multiple types of stellar sources (e.g., red giants, supernovae). Measuring the isotopic composition of various Solar System objects can help to determine the degree of mixing of the starting materials, which can elucidate the physical process that operated in the protoplanetary disk. In addition, isotopic studies may help to identify specific stellar sources that contributed material to the solar nebula, and perhaps, the birth environment of the Solar

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System (e.g., Boss, 2017; Cameron and Truran, 1977; Dwarkadas et al., 2017). Isotopes produced by the p -process are among the rarest in the Solar System (e.g., Arnould and Goriely, 2003; Rauscher et al., 2013). As such, p -isotopes could provide a sensitive tracer of mixing processes in the solar nebula. In particular, the rare isotope ^{180}W (relative abundance 0.12%; Völkening et al., 1991a) may provide a powerful tool to constrain the degree of mixing and/or stellar sources that fed the solar nebula.

Compared to terrestrial material, excesses of ^{180}W occur in members from many groups of magmatic iron meteorites, but their origin is debated. Excess ^{180}W may represent p -process heterogeneity in the solar nebula (Schulz et al., 2013), radiogenic ingrowth on the parent body from ^{184}Os decay (Peters et al., 2014), or spallogenic production due to galactic cosmic ray (GCR) interactions while the material was in outer space (Cook et al., 2014).

Effects from GCRs may increase or decrease ^{180}W concentrations due to the complex interplay between neutron capture and spallation reactions. The final induced effect depends on several variables, including sample depth below the surface, elemental composition, and exposure duration (e.g., Cook et al., 2014; Leya and Masarik, 2013). Without an accurate correction for GCR effects, the true isotopic composition cannot be determined.

Correlations between ^{180}W and Os/W ratios suggest that decay from ^{184}Os may be a contributing factor to ^{180}W excesses (Cook et al., 2014; Peters et al., 2014). However, the W isotopic data of Peters et al. (2014) lacked corrections for GCR effects, and in some cases, also for nucleosynthetic variations on effects on other W isotopes that can cause apparent ^{180}W excesses (e.g., in IVB irons). In Cook et al. (2014), corrections were made for both effects, yet many samples had to be omitted from the resulting isochron because the required parameters needed to properly correct for GCR effects were unknown. Furthermore, in both studies, isochrons were constructed using mixtures of irons from several magmatic groups, which originated on separate parent bodies and with different crystallization histories and ages. The subsequent values for the ^{184}Os decay constant ($\lambda^{184}\text{Os}$) determined in these two studies differ by a factor of ≈ 2 . Hence, the need exists to better constrain the $\lambda^{184}\text{Os}$ value and to properly determine the magnitude of any ^{184}Os decay related to observed ^{180}W excesses. In addition, the application of ^{184}Os - ^{180}W systematics may be a useful approach to determine the initial $e^{180}\text{W}$ in Solar System materials.

A suite of five group IIAB irons were chosen to decipher the exact nature of ^{180}W excesses. Group IIAB irons have several characteristics that make them ideally suited for this: 1) their formation via extensive fractional crystallization (Scott, 1972; Wasson et al., 2007) permits the sampling of a wide range in Os/W ratios required to construct an isochron; 2) their metal-silicate segregation and crystallization ages are independently known from ^{182}Hf - ^{182}W and ^{187}Re - ^{187}Os systematics, respectively (Cook et al., 2004; Kruijer et al., 2014); 3) their cosmic ray exposure (CRE) ages tend to be shorter than the CRE ages of other magmatic groups (e.g., Herzog, 2004), minimizing potential GCR effects; and 4) they lack a resolvable deficit in s -process W isotopes that is observed in some other iron groups (Cook et al., 2014; Kruijer et al., 2014; Qin et al., 2008), eliminating the need to correct for collateral s -deficit effects on ^{180}W . Here, an isochron based on GCR-corrected W isotopic

ratios is used to re-assess the $\lambda^{184}\text{Os}$ value and to determine the initial $^{180}\text{W}/^{184}\text{W}$ ratio of the IIAB iron meteorite parent body.

2. Methods

2.1. Samples and preparation

The five IIAB irons were chosen to sample widely different portions of the crystallization sequence for this group (Fig. 1). Separate metal pieces were cut from the same sample mass for the various analytical objectives required for each sample; adjacent pieces were used for the analyses of W isotopes and for siderophile trace element concentrations. Samples were cut with a slow-speed saw, polished with SiC paper, and rinsed with ethanol. Pieces of ≈ 500 mg were prepared for W isotopic analyses; two pieces (≈ 100 mg each) were cut for analyses of radionuclides (e.g., ^{36}Cl , ^{41}Ca) and noble gases (e.g., Ar), respectively; pieces ranging from ≈ 330 to ≈ 460 mg were prepared for trace element determinations. In the case of Braunau, Cincinnati, and Forsyth County, the same iron meteorite chunks were sampled as in the previous study by Cook et al. (2014). For Forsyth County, insufficient material remained to make a new analysis of W isotopes; thus, the W isotopic data from Cook et al. (2014) are used here.

2.2. Tungsten separation and isotopic analyses

The separation of W from the matrix and the subsequent isotopic analyses follow the methods in Cook and Schönbächler (2016) and are briefly summarized here. Samples were digested in 15 ml of concentrated $\text{HNO}_3\text{:HCl}$ (2:1) at 130 °C. They were dried, fluxed in concentrated HCl, dried again, and dissolved in 0.5 M HCl–0.5 M HF. Tungsten was separated using a two-step anion exchange column procedure using HCl–HF media. Purified W was dried in a mixture of $\text{HNO}_3\text{:H}_2\text{O}_2\text{:HF}$ (1:1:0.5) and then dissolved in 0.5 M HNO_3 –0.2 M HF for isotopic analysis. Isotopic measurements were made using a Neptune Plus MC-ICPMS at ETH–Zürich. All samples, meteoritic and terrestrial, were measured once, bracketed by measurements of the NIST SRM 3163 W solution. Instrumental mass bias was corrected with the exponential law with either $^{186}\text{W}/^{183}\text{W} = 1.98594$ or $^{186}\text{W}/^{184}\text{W} = 0.927672$ (Völkening et al., 1991a). All measurements were corrected for nuclear field shift (NFS) effects (Cook and Schönbächler, 2016). Five replicates of the NIST Fe–Ni steel SRM 129c were chemically processed and analyzed during the same session as the IIAB irons to validate the accuracy of the measurements; this material also served as an external standard to define the long-term reproducibility of the entire analytical method. The external reproducibility (2SD), based on a total of 20 replicate analyses of the NIST Fe–Ni steel SRM 129c, on $\epsilon^i\text{W}$ values are ± 0.05 for $\epsilon^{184}\text{W}$ (6/3), ± 0.49 for $\epsilon^{180}\text{W}$ (6/4), ± 0.08 and for $\epsilon^{182}\text{W}$ (6/4) (see Cook and Schönbächler, 2016; Table 2). The notation “(6/3)” and “(6/4)” indicates the ratio used for the mass bias correction (i.e., $^{186}\text{W}/^{183}\text{W}$ or $^{186}\text{W}/^{184}\text{W}$).

2.3. Trace element analyses

Trace element concentrations (Re, Os, Ir, Pt, W) were determined at the University of Maryland (College Park) by isotope dilution. Separation of Re, Os, Ir, and Pt followed the methods in Walker et al. (2008). Samples were digested with 5 ml of concentrated HNO_3 and 2.5 ml of concentrated HCl in Pyrex[®] Carius tubes and equilibrated with a combined

platinum-group element spike (^{191}Ir and ^{194}Pt) and a Re–Os spike (^{185}Re and ^{190}Os) for 24 h at 220 °C (Shirey and Walker, 1995). Osmium was separated by solvent-extraction (Cohen and Waters, 1996), further purified using micro-distillation (Birck et al., 1997), and then analyzed by N-TIMS (ThermoScientific Triton). A natural Os in-house standard was also measured to correct for instrumental mass fractionation, which was negligible for these measurements ($\ll 0.1\%$). The remaining Re, Ir and Pt were purified using an anion exchange column procedure (Rehkämper et al., 1997). Purified and separated Re, Ir, and Pt were evaporated to dryness and dissolved in 0.8 M HNO_3 for analysis by MC-ICPMS (ThermoScientific Neptune Plus), using natural Re, Ir, and Pt standards to correct for instrumental mass fractionation. The separation of W is based on the methods from Touboul and Walker (2012). Samples were digested in 5 ml of 8 M HCl and equilibrated with a ^{182}W spike at 130 °C. Tungsten was purified using an anion exchange column and dissolved in 0.8 M HNO_3 for analysis by ICPMS, using a natural W standard to correct for instrumental mass fractionation. The total blanks for Os, Re, Ir, Pt, and W were 1, 0.4, 0.5, 4, and 30 pg, respectively, which are negligible for all samples. The uncertainties (2SE) on Os, Re, Ir, Pt, and W concentrations are 0.1%, 0.1–0.2%, 0.1–0.2%, 0.1–0.2%, and 0.1%, respectively.

2.4. Cosmogenic nuclide analyses

2.4.1. Noble gases—Measurements of noble gas concentrations and isotopic compositions were performed at the University of Bern, following the procedures described by Ammon et al. (2008, 2011) and Smith et al. (2017). In brief, after cleaning with ethanol, the samples were loaded into an all-metal noble gas extraction line. The samples and the oven were heated in vacuum at ≈ 80 °C for ≈ 48 h to remove adsorbed atmospheric gases. Gas extraction was done in a molybdenum crucible held at 1800 °C for ≈ 30 –40 min; a boron nitride liner inside the crucible prevented the formation of MoFe alloys, which increase blanks and damage the crucible. The Ar fraction was separated from the He–Ne fraction using an activated charcoal held at the temperature of liquid nitrogen. The Ar fraction was purified using SAES[®] getters held at different temperatures and was measured in a tandem noble gas mass spectrometer. Blanks were regularly monitored in between measurements; they contributed to less than 10% for $^{36,38,40}\text{Ar}$. Calibrations were frequently performed using standard gas containing Ar with an atmospheric isotopic composition.

2.4.2. Cosmogenic radionuclides—The extraction of cosmogenic radionuclides was conducted in parallel with a project studying the cosmic ray exposure history and terrestrial age of the Twannberg iron meteorite, and details of the entire procedure can be found in Smith et al. (2017). In brief, chemical separations and cosmogenic radionuclide measurements were performed at the DREsden AMS (DREAMS) facility of the Helmholtz-Zentrum Dresden-Rossendorf (HZDR). The chemical separations were adapted following the methods described in Merchel and Herpers (1999). The samples were cleaned, weighed, and then subsequently dissolved in 2 M HNO_3 at room temperature, together with a carrier solution of natural $^{35}\text{Cl}/^{37}\text{Cl}$. Chlorine was precipitated as AgCl, and the remaining solution was mixed with a carrier solution of $^{\text{nat}}\text{Ca}$. Calcium was separated by a combination of precipitation and ion exchange chemistry. The separated Ca was converted to fluoride (CaF_2) for isotopic analysis. The chemical yields are $\approx 71\%$ for AgCl and $\approx 79\%$ for CaF_2 . All targets were pressed into Cu sample holders and measured by Accelerator Mass

Spectrometry (AMS), at the DREAMS facility. A typical full procedural blank has ratios of 1.5×10^{-14} and 3.4×10^{-14} for $^{36}\text{Cl}/^{35+37}\text{Cl}$ and $^{41}\text{Ca}/^{40}\text{Ca}$, respectively, which corresponds to corrections of $\approx 0.7\%$ and $0.2\text{--}21\%$ for ^{36}Cl and ^{41}Ca activities, respectively. Secondary in-house standards were used for normalization: “SMD-Cl-11” for $^{36}\text{Cl}/^{35+37}\text{Cl}$ and “SMD-Ca-12” for $^{41}\text{Ca}/^{40}\text{Ca}$ (e.g., Rugel et al., 2016). Here we briefly discuss the results for ^{36}Cl and ^{41}Ca ; a complete discussion of the radionuclide and noble gas data will be given elsewhere.

3. Results

3.1. Tungsten isotopes

As discussed in Cook and Schönbächler (2016), measured W isotopic ratios need to be corrected for nuclear field shift (NFS) effects, but these effects are insignificant on ratios that do not include ^{183}W . In the current study, corrections for NFS effects on measured $\epsilon^{182}\text{W}$ (6/4) values are <0.005 , and those on measured $\epsilon^{180}\text{W}$ (6/4) values are <0.002 . Three of five samples show clearly resolved excesses in measured $\epsilon^{180}\text{W}$ values relative to the NIST W solution standard SRM 3163 (Table 1). For Braunau and Cincinnati, our new data are in good agreement with those in Cook et al. (2014), which were made on samples from the same meteorite pieces. Two samples (Sikhote Alin and North Chile) show clear evidence for ^{182}W burnout due to GCR effects, as their $\epsilon^{182}\text{W}$ values are significantly lower than the pre-exposure value of -3.40 ± 0.03 for IIAB irons (Kruijer et al., 2014). The $\epsilon^{184}\text{W}$ data are consistent with previous studies (Cook et al., 2014; Kruijer et al., 2014; Qin et al., 2008). Our data, combined with the above three studies, yield a mean $\epsilon^{184}\text{W}$ value of -0.01 ± 0.02 (95% CI; $n = 22$), demonstrating that IIAB irons do not contain a resolvable deficit in s -process W isotopes.

3.2. Siderophile trace elements

In general, the IIAB irons analyzed here yield Re, Ir, Pt, and W concentrations (Table 2) that are in good agreement (within $\sim 10\%$) with those reported by Wasson et al. (2007) using the INAA technique. Concentrations of Os, Re, and Pt for North Chile agree well (within $\approx 4\%$) with those reported by Cook et al. (2004), also obtained by the isotope dilution technique. These data support the overall chemical homogeneity of individual IIAB irons as already noted in previous studies regarding their use as analytical standards (e.g., Walker et al., 2008; Wasson et al., 1989, 2007).

3.3. Cosmogenic nuclides and the exposure history to galactic cosmic rays

Concentrations of cosmogenic nuclides, along with the associated terrestrial ages and CRE ages, are given in Table 3. Measured $^{41}\text{Ca}/^{36}\text{Cl}$ ratios are independent of shielding and are used to calculate accurate terrestrial residence ages, T_{terr} (Smith et al., 2017) using the following formula:

$$T_{terr} = \frac{1}{(\lambda_{36} - \lambda_{41})} \times \ln \left(\frac{\left(\frac{{}^{41}\text{Ca}}{{}^{36}\text{Cl}} \right)_{meas}}{1.105 \pm 0.048} \right)$$

where $({}^{41}\text{Ca}/{}^{36}\text{Cl})_{meas}$ is the measured ratio, λ_{36} and λ_{41} are the decay constants for ${}^{36}\text{Cl}$ and ${}^{41}\text{Ca}$, respectively ($\lambda_{36} = 2.30 \times 10^{-6} \text{ yr}^{-1}$ (Nica et al., 2012); $\lambda_{41} = 6.66 \times 10^{-6} \text{ yr}^{-1}$ (Kutschera et al., 1992)), and the value 1.105 ± 0.048 is the production rate ratio for ${}^{41}\text{Ca}/{}^{36}\text{Cl}$ in iron meteorites from exposure to GCRs (Smith et al., 2017). The terrestrial ages are then used to correct ${}^{36}\text{Cl}$ activities back to the time of fall; this corrects for decay after arriving on Earth, which is needed to calculate ${}^{36}\text{Cl}$ - ${}^{36}\text{Ar}$ CRE ages (T_{CRE}). The production rate ratio for ${}^{36}\text{Cl}/{}^{36}\text{Ar}$ in iron meteorites is shielding-independent, making ${}^{36}\text{Cl}$ - ${}^{36}\text{Ar}$ systematics ideal for calculating CRE ages (e.g., Lavielle et al., 1999; Smith et al., 2017); The exposure ages are calculated via:

$$\begin{aligned} T_{CRE} &= \frac{1}{\lambda_{36}} \times \frac{P({}^{36}\text{Cl})}{P({}^{36}\text{Ar})} \times \frac{{}^{36}\text{Ar}_c}{{}^{36}\text{Cl}} \\ &= 511 \times (0.835 \pm 0.040) \times \frac{{}^{36}\text{Ar}_c}{{}^{36}\text{Cl}} \end{aligned}$$

where λ_{36} is the decay constant for ${}^{36}\text{Cl}$, $P({}^{36}\text{Cl})/P({}^{36}\text{Ar})$ is the production rate ratio in iron meteorites (0.835 ± 0.040 ; Lavielle et al., 1999), ${}^{36}\text{Ar}_c$ is the cosmogenic component of ${}^{36}\text{Ar}$, ${}^{36}\text{Cl}$ is the activity at the time of fall, and the value 511 corrects for the differing units in concentrations for ${}^{36}\text{Cl}$ (dpm/kg) and ${}^{36}\text{Ar}$ ($10^{-8} \text{ cm}^3\text{STP/g}$); the constant 511 also incorporates the value for λ_{36} . Terrestrial ages determined for the five IIAB irons studied here range from <12 ka (Braunau) up to 601 ka (North Chile), and CRE ages range from 12 Ma (Braunau) to 385 Ma (Sikhote-Alin). In general, the CRE ages of these samples are relatively short for iron meteorites, with four samples having ages > 200 Ma.

4. Discussion

The main goals of the current study are to determine the magnitude of the radiogenic contribution to measured ${}^{180}\text{W}$ excesses from ${}^{184}\text{Os}$ decay and to better constrain the decay constant ($\lambda^{184}\text{Os}$). This information can be obtained using an isochron approach for the Os–W system by plotting ${}^{180}\text{W}/{}^{184}\text{W}$ ratios (i.e., $e^{180}\text{W}$ values) versus the parent/daughter ratio (${}^{184}\text{Os}/{}^{184}\text{W}$). Before this can be done, the measured $e^{180}\text{W}$ values need to be corrected for effects from GCRs and for a deficit in s -process W isotopes, if present (Cook et al., 2014, 2015). As noted earlier in section 3.1, IIAB irons do not show evidence for a resolvable deficit in s -process W isotopes, so this correction to $e^{180}\text{W}$ is not required. However, the CRE ages (Table 3) show that all five studied samples have experienced exposure to GCRs; therefore, these effects on $e^{180}\text{W}$ need to be accounted for.

4.1. Galactic cosmic ray effects on ^{180}W

Cosmic ray particles can induce nuclear reactions within meteoroids that modify their initial isotopic composition. Isotopic effects from GCRs arise from both spallation reactions and neutron capture reactions. The magnitude of the effects depends on several factors, including meteoroid size, depth below the surface, elemental composition, and exposure duration (e.g., Leya et al., 2000; Masarik and Reedy, 1994; Nishiizumi et al., 1986). Furthermore, because spallation reactions dominate closest to the surface, whereas neutron capture reactions reach a peak at depth below the surface (e.g., Masarik, 1997; Reedy and Arnold, 1972), these two effects are decoupled. Cook et al. (2014) demonstrated that GCR effects on $\epsilon^{180}\text{W}$ can be significant and can lead to a net gain or loss of ^{180}W atoms within the sample. Neutron capture reactions consume ^{180}W atoms, whereas spallation reactions on Re, Os, and other W isotopes, produce ^{180}W atoms. We present an updated version of the GCR model used in Cook et al. (2014), which was based on that of Leya and Masarik (2013). The spallation component has been improved by adding Ir and Pt as target elements. Like Re and Os, these elements may be enriched relative to W in iron meteorites and their isotopes have masses greater than, but close to, 180.

Figs. 2a and b show selected model results (i.e., model results for chosen elemental ratios) that predict GCR effects on $\epsilon^{180}\text{W}$ (6/4) and $\epsilon^{182}\text{W}$ (6/4) versus depth in meteoroids with differing radii. The net effects include the combined influence of neutron capture and spallation reactions. An exposure age of 1000 Ma was chosen as a near upper limit on possible effects as most iron meteorites have CRE ages of ~1000 Ma (e.g., Herzog, 2004). Figs. 2a and b are used to illustrate potential GCR effects for two end-member cases: irons depleted in siderophile elements relative to W and irons enriched in siderophile elements relative to W. In both cases, element/W ratios were chosen to represent realistic values in iron meteorites (e.g., Cook et al., 2004; McCoy et al., 2011; Walker et al., 2008; Wasson, 1999; Wasson et al., 2007); irons that crystallize late in the sequence will be depleted in siderophile elements relative to W, whereas those that crystallize early will be enriched. Fig. 2a shows the expected effects for iron meteorites depleted in siderophile elements relative to W. In this case, neutron capture reactions dominate and lead to burnout of both ^{180}W and ^{182}W . Only slight ($\approx 0.5\epsilon$) spallation production of ^{180}W occurs near the surface. Conversely, spallation production of ^{180}W may be significant (up to $\approx 4\epsilon$) in samples enriched in siderophile elements relative to W (Fig. 2b); these effects are particularly strong near the surface and in bodies with smaller radii. Competing neutron capture effects may nonetheless lead to a decrease in $\epsilon^{180}\text{W}$ depending on radius and depth. For meteoroids enriched in siderophile elements, the inclusion of Ir and Pt abundances in the model typically increases the positive shift in $\epsilon^{180}\text{W}$ by $\approx 25\%$ and $\approx 15\%$, respectively, compared to only considering Re and Os as possible target elements (cf. Fig. 9b in Cook et al., 2014). Thus, Ir and Pt are important targets for spallation production of ^{180}W , and our new model more fully accounts for these effects. It is important to note that $\epsilon^{182}\text{W}$ (6/4) values are essentially free from spallation effects; thus, shifts in $\epsilon^{182}\text{W}$ (6/4) values in iron meteorites relative to their pre-exposure value serve as a reliable neutron dosimeter. This negates the need to use an additional system (e.g., Pt isotopes) to correct GCR effects on $\epsilon^{180}\text{W}$ (6/4).

The correlation between GCR effects on $\epsilon^{180}\text{W}$ and $\epsilon^{182}\text{W}$ was determined for each sample using our model by inputting the associated measured Re/W, Os/W, Ir/W, and Pt/W ratios, along with the CRE age. The resulting slope and intercept, based on a linear regression of the model results, were then used to calculate the magnitude of the GCR effect on $\epsilon^{180}\text{W}$ from the observed shift in $\epsilon^{182}\text{W}$ (i.e., $\epsilon^{182}\text{W} = \epsilon^{182}\text{W}_{\text{measured}} - \epsilon^{182}\text{W}_{\text{pre-exposure}}$), where, $\epsilon^{182}\text{W}_{\text{pre-exposure}} = -3.40 \pm 0.03$ for IIAB irons (Kruijer et al., 2014). To reduce uncertainties, our measured values for $\epsilon^{180}\text{W}$ and $\epsilon^{182}\text{W}$ were combined with those of Cook et al. (2014) for the irons Braunau and Cincinnati because the sample pieces are the same in both studies. The new weighted means for the measured values are $\epsilon^{180}\text{W} = 1.24 \pm 0.46$ and $\epsilon^{182}\text{W} = -3.36 \pm 0.07$ (Braunau) and $\epsilon^{180}\text{W} = 1.24 \pm 0.49$ and $\epsilon^{182}\text{W} = -3.31 \pm 0.07$ (Cincinnati). The GCR corrections on $\epsilon^{180}\text{W}$ (6/4) values for these five particular irons were relatively small (i.e., within $\pm \approx 0.40$). After correcting for GCR-induced effects (Table 1), all five samples show $\epsilon^{180}\text{W}$ excesses larger than the overall analytical resolution of ± 0.49 (2SD).

4.2. Radiogenic contribution to ^{180}W excesses

To determine the radiogenic contribution to ^{180}W excesses from ^{184}Os decay, the GCR-corrected $^{180}\text{W}/^{184}\text{W}$ ratios were plotted against the parent/daughter ratios ($^{184}\text{Os}/^{184}\text{W}$) to obtain an isochron diagram for the ^{184}Os – ^{180}W system. Abundances for W and Os isotopes were taken from Völkening et al. (1991a, 1991b) to calculate $^{184}\text{Os}/^{184}\text{W}$ ratios from measured concentrations. A regression through the data yields a slope of 0.000093 ± 0.000047 with an MSWD of 1.4; the y-axis intercept gives an $\epsilon^{180}\text{W}_i$ of 0.63 ± 35 . The relationship between ^{180}W excesses and Os/W ratios is shown in Fig. 3; the $^{180}\text{W}/^{184}\text{W}$ ratios are plotted as $\epsilon^{180}\text{W}$ to facilitate direct comparisons to earlier studies by Cook et al. (2014) and Peters et al. (2014). The spread in Os/W ratios has been extended compared to previous studies; our minimum and maximum values differ by a factor of 762, compared to a factor of 292 (Cook et al., 2014) and 183 (Peters et al., 2014). Combining our slope with the ^{187}Re – ^{187}Os crystallization age of the IIAB irons (4546 ± 46 Ma; Cook et al., 2004), gives a value of $(2.05 \pm 1.01) \times 10^{-14} \text{ a}^{-1}$ for $\lambda^{184}\text{Os}$. This value is smaller than the one proposed by Cook et al. (2014) by a factor of 1.5, although both values agree within the uncertainties. The new value is also smaller than the one reported by Peters et al. (2014) by a factor of 3.2. Thus, our new value for $\lambda^{184}\text{Os}$ is more in accord with the upper limit estimated by Sperlein and Wolke (1976) based on a counting experiment (i.e., $\lambda^{184}\text{Os} = 1.24 \times 10^{-14} \text{ a}^{-1}$). Overall, our GCR-corrected data (Fig. 3) provide strong evidence that ^{184}Os indeed undergoes α -decay with a long half-life.

4.3. The initial $^{180}\text{W}/^{184}\text{W}$ ratio in IIAB irons

A striking feature of Fig. 3 is that the initial $^{180}\text{W}/^{184}\text{W}$ ratio is non-zero ($\epsilon^{180}\text{W}_i = 0.63 \pm 35$), and the positive offset is resolved from the terrestrial value. This offset could potentially be explained by ^{184}Os decay prior to the crystallization of the IIAB irons. Conversely, the offset may represent an excess of p -process material in the IIAB parent body relative to Earth.

4.3.1. Radiogenic decay of ^{184}Os —The $\epsilon^{180}\text{W}_i$ for the IIAB irons represents the value at the time of core crystallization, ≈ 21 Ma after CAI formation; thus, this value will

have evolved during that time due to ^{184}Os decay. Additionally, SRM 3163 represents the modern value of the terrestrial silicate Earth, which also evolved over Solar System history. The effects on $\epsilon^{180}\text{W}$ from radiogenic ingrowth on both bodies can be estimated so that a more direct comparison can be made between Earth and the IIAB irons. Here we assume that Earth and the IIAB parent body formed from material with a chondritic Os/W ratio of 5.46 (Lodders, 2003). Osmium is highly siderophile, whereas W is moderately siderophile, and core formation under oxidizing conditions could fractionate Os from W (e.g., Campbell and Humayun, 2005). The IIAB irons have a mean Fe/Ni ratio (Wasson et al., 2007) very close to the chondritic value, indicating that the IIAB core segregated under reducing conditions. Such conditions would promote the partitioning of both Os and W into the metal phase. In the end-member case where all of the Os and W partitioned into the IIAB core, preserving the chondritic Os/W ratio, then the $\epsilon^{180}\text{W}$ value would have evolved by only +0.004 by the time of core crystallization – i.e., closure of the ^{184}Os – ^{180}W system in IIAB irons. Hence, the $\epsilon^{180}\text{W}_i$ obtained from the IIAB ^{184}Os – ^{180}W isochron effectively represents the value of the IIAB precursor material at the start of the Solar System, and radiogenic ingrowth fails to explain the observed positive offset relative to SRM 3163. On Earth, assuming core formation ≈ 100 Ma after CAIs, and using a bulk silicate Earth (BSE) Os/W ratio of 0.1 (McDonough, 2004; and references therein), the BSE $\epsilon^{180}\text{W}$ value would have evolved by +0.035 over Earth's entire history. The actual change in $\epsilon^{180}\text{W}$ may have been some-what more or less depending on which terrestrial reservoir (e.g., crust, depleted mantle, primitive mantle) was the source of the W in SRM 3163. Nevertheless, $\epsilon^{180}\text{W}_i$ for the IIAB irons should be slightly negative relative to the terrestrial value (SRM 3163) with no resolvable difference, given the current analytical precision (Fig. 4). An earlier time of core formation on Earth would shift the terrestrial evolution lines to lower values in Fig. 4; however, even terrestrial core formation as early as 50 Ma after CAIs would not produce terrestrial $\epsilon^{180}\text{W}$ values less than the initial value observed in IIAB irons. Therefore, the observed positive offset represents a true difference between the precursor materials of the IIAB irons and Earth regarding their ^{180}W abundances.

4.3.2. Heterogeneity in p -process ^{180}W —Fig. 5 shows the stellar production ratios for p -process nucleosynthesis during a type-II supernova (SNII) for the siderophile elements Mo, Ru, W, Os, and Pt (Rauscher et al., 2013). The p -isotopes of the heavy siderophile elements (W, Os, Pt) are produced in a relatively narrow temperature range that is lower and distinct from the regime in which the p -isotopes of Mo and Ru are synthesized; thus, stellar material enriched in p -process ^{180}W is not expected to be enriched in Mo or Ru p -isotopes. Although enrichments in $^{92,94}\text{Mo}$ and $^{96,98}\text{Ru}$ have been reported for IIAB irons, the variations were shown to be collateral effects caused by a deficit in s -process isotopes of those elements rather than a true p -process excess (Burkhardt et al., 2011; Chen et al., 2010; Dauphas et al., 2002; Fischer-Gödde et al., 2015). An enrichment in low-temperature p -process material may also lead to variations in ^{184}Os and ^{190}Pt in IIAB irons. Walker (2012) reported normal $\epsilon^{184}\text{Os}$ values in IIAB irons; similarly, no resolved variations were observed in $\epsilon^{190}\text{Pt}$ (Hunt et al., 2017; Peters et al., 2015). However, the external reproducibility of these studies is rather large ($\epsilon^{184}\text{Os} \pm 27$, $\epsilon^{190}\text{Pt} \pm \approx 2$) compared to the small initial $\epsilon^{180}\text{W}$ excess observed in IIAB irons. Consequently, any potential variation in ^{184}Os and ^{190}Pt may not yet be detectable.

5. Conclusions

We present an updated model to describe GCR effects on W isotopes and show that in addition to Re and Os (Cook et al., 2014), Ir and Pt are also important targets for the spallation production of ^{180}W . After correcting measured $\epsilon^{180}\text{W}$ values for GCR effects, the residual values show a correlation with Os/W ratios, confirming that radiogenic ingrowth from ^{184}Os decay has contributed to the observed $\epsilon^{180}\text{W}$ excesses in IIAB irons and likely extends to other groups of iron meteorites. Our revised value for $\lambda^{184}\text{Os} = (2.05 \pm 1.01) \times 10^{-14} \text{ a}^{-1}$ is in good agreement with the upper limit estimated from a counting experiment by Sperlein and Wolke (1976). The new $\lambda^{184}\text{Os}$ value corresponds to a half-life of $(3.38 \pm 2.13) \times 10^{13} \text{ a}$. Resolvable $\epsilon^{180}\text{W}$ variations in mantle rocks (either primitive or depleted mantle samples) are not expected due to the extremely long half-life of ^{184}Os and low Os/W ratios in the mantle. Thus, this decay system does not show promise as a terrestrial chronometer given the current analytical precision.

To date, only isotopic variations in ^{144}Sm (Andreasen and Sharma, 2006; Brennecka et al., 2013; Qin et al., 2011) and ^{174}Hf (Peters et al., 2017) in early Solar System materials appear to represent true variation in the p -process component of these elements. Anomalies in ^{84}Sr may also represent p -process heterogeneity but may also result from variations in s - or r -process Sr isotopes (Brennecka et al., 2013; Hans et al., 2013; Moynier et al., 2012). The intercept of the IIAB isochron yields an initial $\epsilon^{180}\text{W} = 0.63 \pm 0.35$, which is resolved from the terrestrial standard. This is the first clear evidence for p -process variability in W isotopes. Models of stellar nucleosynthesis (Rauscher et al., 2013) indicate that the IIAB irons contain an excess of a low temperature component of the p -process.

The $\epsilon^{180}\text{W}$ excesses observed in IIAB irons do not have a single origin; these excesses consist of three components which include a small initial nucleosynthetic anomaly upon which radiogenic ingrowth from ^{184}Os decay has occurred, and this combined isotopic signature has then been overprinted by GCR effects. Therefore, radiogenic effects and the initial isotopic ratio can only be determined after correcting measured $\epsilon^{180}\text{W}$ values for GCR-induced effects. Investigations of other iron meteorite groups, using the same approach, could help to elucidate if heterogeneity in p -process W was widespread in the early Solar System or if it is restricted to the IIAB parent body.

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Fig. 1.

Concentrations of Ir versus Au in IIAB iron meteorites. The five samples included in this study (Sikhote-Alin, North Chile, Braunau, Cincinnati, Forsyth County) are shown by the blue circles. Data from Wasson et al. (2007). (For interpretation of the colors in the figure(s), the reader is referred to the web version of this article.)

Fig. 2.

Model results for the combined effects of neutron capture and spallation reactions on both $\epsilon^{180}\text{W}$ (6/4) and $\epsilon^{182}\text{W}$ (6/4). The model predictions are for iron meteoroids with various radii (25, 40, 65, 85, 100 cm) and a CRE age of 1000 Ma. Panel (a) is for iron meteoroids depleted in heavy siderophile elements relative to W, and panel (b) is for iron meteoroids enriched in heavy siderophile elements relative to W; symbols are the same in both panels.

Fig. 3.

Isochron-type diagram for IIAB irons. The $\epsilon^{180}\text{W} (6/4)$ values have been corrected for GCR-induced effects. The best-fit line (black) corresponds to a value for the decay constant $\lambda^{184}\text{Os} = (2.05 \pm 1.01) \times 10^{-14} \text{ a}^{-1}$. The slopes corresponding to decay constants reported by Sperlein and Wolke (1976) and Peters et al. (2014) are shown for comparison. The gray band represents the analytical resolution ($\pm 0.49 \epsilon$; 2SD) of the current study. Uncertainties on $^{184}\text{Os}/^{184}\text{W}$ ratios are smaller than the symbol size.

Fig. 4.

Evolution of $\epsilon^{180}\text{W}$ due to radiogenic decay of ^{184}Os for the initial condition that Earth and the IIAB parent body both form from a chondritic reservoir, where Os/W equals 5.46 (Lodders, 2003). Two trajectories are shown for the evolution of $\epsilon^{180}\text{W}_i$ in IIAB iron meteorites prior to core crystallization: 1) core segregation under reducing conditions (red line), preserving the chondritic Os/W ratio in the initial liquid core; 2) core segregation under oxidizing conditions (blue line) such that the core initially has an Os/W ratio of ≈ 11 (e.g., Campbell and Humayun, 2005). Core segregation is taken at 0.7 Ma after CAIs (Kruijer et al., 2014). No further radiogenic ingrowth is shown after core crystallization is complete (i.e., ≈ 21 Ma after Solar System formation) on the IIAB parent body (red and blue horizontal lines). The evolution of $\epsilon^{180}\text{W}$ is also shown for various terrestrial reservoirs over the entirety of Solar System history: the bulk silicate Earth (“BSE”), the bulk continental crust (“crust”), and the primitive mantle (“mantle”), with Os/W ratios of 0.1, 0.00005, and 0.21, respectively (McDonough, 2004; Palme and O’Neill, 2004; Taylor and McLennan, 1995). Terrestrial core formation is assumed 100 Ma after the start of the Solar System, and crust-mantle differentiation is taken at 3 Ga ago. Neither IIAB trajectory predicts an $\epsilon^{180}\text{W}_i$ value that is positive relative to Earth, contrary to the observations (Fig. 3).

Fig. 5.

Stellar production of selected p -isotopes as a function of temperature in a SNII with a $20 M_{\odot}$ progenitor. The y -axis shows the ratio of the final abundance of an isotope to its initial abundance in the pre-supernova star. Net production of p -isotopes is highlighted by the gray shaded area. Data are from Rauscher et al. (2013).

Table 1

Tungsten isotopic compositions for IIAB irons.

Sample	$\epsilon^{184}\text{W} (6/3)^a$	$\epsilon^{182}\text{W} (6/4)^a$	$\epsilon^{180}\text{W} (6/4)^a$	$\epsilon^{180}\text{W}_{\text{GCR}} (6/4)^b$
Sikhote-Alin	-0.03 ± 0.04	-3.79 ± 0.10	0.30 ± 0.65	0.68 ± 0.68
North Chile	-0.04 ± 0.04	-3.56 ± 0.10	0.33 ± 0.65	0.55 ± 0.68
Braunau	-0.03 ± 0.04	-3.34 ± 0.10	1.14 ± 0.65	1.22 ± 0.48
<i>Cook et al.</i>	-0.03 ± 0.05	-3.38 ± 0.10	1.33 ± 0.64	
Cincinnati	-0.08 ± 0.04	-3.20 ± 0.10	1.27 ± 0.65	0.86 ± 0.53
<i>Cook et al.</i>	0.03 ± 0.05	-3.40 ± 0.09	1.21 ± 0.76	
Forsyth County ^c	-0.02 ± 0.04	-3.46 ± 0.09	2.62 ± 0.80	2.68 ± 0.84

All epsilon values are expressed relative to the standard SRM 3163 in parts per 10^4 . Terrestrial W isotopic ratios in SRM 3163 are calculated from abundances given in Völkering et al. (1991a). The notation “(6/3)” and “(6/4)” indicates the ratio used for the mass bias correction (i.e., $^{186}\text{W}/^{183}\text{W}$ or $^{186}\text{W}/^{184}\text{W}$). Uncertainties on measured values (2SD) are based on all measurements of the bracketing standard (SRM 3163) during the analytical session.

^aMeasured values are corrected for nuclear field shift effects (Cook and Schönbachler, 2016).

^bValues corrected for galactic cosmic ray (GCR) effects; corrections for Braunau and Cincinnati are based on weighted means of measured values, combining those in Table 1 and those from Cook et al. (2014) - see text (section 4.1) for more details.

^cMeasured values taken from Cook et al. (2014).

Table 2

Trace element concentrations (ppb) in IIAB irons.

Sample	Re ^a	Os ^b	Ir ^a	Pt ^a	W ^a
Sikhote-Alin	1.528 ± 0.003	12.14 ± 0.01	23.08 ± 0.05	5377 ± 5	735.2 ± 0.7
North Chile	208.8 ± 0.2	1068 ± 1	3585 ± 4	26590 ± 30	2463 ± 2
Braunau	772.5 ± 0.8	5450 ± 3	11020 ± 10	30080 ± 30	2946 ± 3
Cincinnati	1530 ± 2	13020 ± 7	19940 ± 20	33330 ± 30	3208 ± 3
Forsyth County	3700 ± 4	44570 ± 36	39270 ± 40	37610 ± 70	3542 ± 4

All values determined by isotope dilution. Uncertainties on individual values are 2SE.

^aMeasured by MC-ICPMS.

^bMeasured by N-TIMS.

Table 3

Cosmogenic nuclide concentrations in IIAB irons, along with terrestrial residence (T_{terr}) and cosmic ray exposure (T_{CRE}) ages.

Sample	$^{41}\text{Ca}^a$	$^{36}\text{Cl}^d$	$^{36}\text{Cl}^b$	$^{36}\text{Ar}^c$	T_{terr}^d	T_{CRE}^e
Sikhote-Alin	9.401 ± 0.634	7.300 ± 0.094	7.300 ± 0.094	6.592 ± 0.258	fall	385 ± 24
North Chile	0.134 ± 0.053	1.673 ± 0.022	6.675 ± 1.149	1.249 ± 0.049	601 ± 92	80 ± 18
Braunau	21.25 ± 1.00	20.13 ± 0.23	20.13 ± 0.90	0.564 ± 0.022	<12	12 ± 1
Cincinnati	16.94 ± 0.76	20.46 ± 0.22	23.82 ± 0.83	11.16 ± 0.44	66 ± 14	200 ± 14
Forsyth County	5.541 ± 0.416	12.03 ± 0.13	19.07 ± 1.92	1.850 ± 0.073	200 ± 20	41 ± 5

All uncertainties are 1SD.

^aMeasured activities in dpm/kg.

^bActivity at the time of fall after correction for terrestrial ages.

^cCosmogenic component ("c") of ^{36}Ar in $10^{-8} \text{ cm}^3\text{STP/g}$.

^dTerrestrial residence ages in ka, based on $^{41}\text{Ca}/^{36}\text{Cl}$ systematics (see text, section 3.3, for more details).

^eCosmic ray exposure ages in Ma, based on $^{36}\text{Cl}-^{36}\text{Ar}$ systematics (see text, section 3.3, for more details).