XENON IN THE PROTOPLANETARY DISK (PPD-Xe)

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ABSTRACT

Relationships among solar system Xe components as observed in the solar wind, in planetary atmospheres, and in meteorites are investigated using isotopic correlations. The term PPD-Xe is used for components inferred to have been present in the molecular cloud material that formed the protoplanetary disk (PPD). The evidence of the lack of simple relationships between terrestrial atmospheric Xe and solar or meteoritic components is confirmed. Xe isotopic correlations indicate a heterogeneous PPD composition with variable mixing ratios of the nucleosynthetic component Xe-HL. Solar Xe represents a bulk PPD component, and the isotopic abundances did not change from the time of incorporation into the interior of Mars through times of regolith implantations to the present.

Key words: planets and satellites: atmospheres – planets and satellites: composition – planets and satellites: formation - planets and satellites: physical evolution - solar wind - Sun: abundances

1. INTRODUCTION

In a publication termed "xenology," Reynolds (1963) reviewed information available at the time and commented on the potential of the nine-isotope Xe system for solar system research. Concurrently, Signer & Suess (1963) subdivided known solar system gas abundances into "solar" and "planetary" components. This terminology has survived to the present, although it became clear that "planetary" does not apply to planets. The term PPD-Xe (protoplanetary disk) is used here to characterize xenon in the forming solar system. The history of research on solar system noble gases is covered in reviews by Wieler (2002), Ott (2002), and Pepin & Porcelli (2002). Among Xe components carrying pre-solar phases, the Xe-HL component in presolar nanodiamonds is most abundant.

The origin of terrestrial and planetary volatiles and processes involved in their evolution are not well understood. Elemental and isotopic abundances in giant planets are generally considered to represent those prevailing in their source reservoirs at the time of formation. Spectroscopic and in situ measurements of nitrogen isotopic abundances in Jupiter and Saturn (Owen et al. 2001; Fletcher et al. 2014) agree with N in the solar wind (SW; Marty et al. 2011) and therefore agree with this assumption. However, Saturn's moon Titan has distinct N isotopic abundances (Niemann et al. 2010 and references therein). The 36 Ar/ 38 Ar and 20 Ne/ 22 Ne ratios reported for Jupiter by the Galileo Probe (Mahaffy et al. 2000) are consistent with the SW (Heber et al. 2009; Meshik et al. 2014).

Noble gases and nitrogen in Earth and Mars atmospheres are distinct from SW abundances (Heber et al. 2009) and indicate either different planetary histories or matter from different sources (Bogard et al. 2001). Xe isotopic abundances in the atmospheres of planets Earth and Mars are similar, but strongly fractionated relative to SW Xe (Marti & Mathew 1998), and in planet Mars, an interior solar (Chass-S) Xe was identified (Mathew & Marti 2001).

Albarede (2009) concluded that terrestrial volatiles were likely delivered some 100 million years after the formation of the solar system and that the depletion of volatiles on terrestrial planets are not due to loss by outgassing, as the isotopic fractionation of Zn is incompatible with volatilization. Contributions of matter from beyond the asteroidal belt were

suggested. Therefore, comparisons of planetary abundances with cometary isotopic data are important. H and N in comets reveal enrichments in the heavy isotopes (Rousselot et al. 2014). The noble gas composition of Jupiter by the Galileo Probe (Mahaffy et al. 2000) shows that the isotopic compositions of Ar, Kr, and Xe were similar to those in SW, but Xe isotopes have large uncertainties. He and Ne abundances relative to H are lower by factors of 0.8 and 0.1, respectively. Thus, no single source for planetary volatiles of Earth and Mars is known.

2. Xe ISOTOPIC CORRELATIONS

Isotopic differences in Xe have been known to exist in solar, planetary, and meteoritic matter (e.g., Pepin 2000 and references therein; Marti & Mathew 1998; Meshik et al. 2014). These isotopic signatures provide clues regarding origin and processes in the PPD (Huss et al. 1996) and regarding planetary atmospheres (Marti & Mathew 1998; Pepin 2000 and references therein; Crowther & Gilmour 2013; Meshik et al. 2014). SW Xe data from Genesis, lunar, and Pesyanoe are listed in Table 1, together with meteoritic and planetary Xe components. Abee-Xe is also included, as enstatite chondritic matter is considered to characterize matter of the inner solar system (Lee et al. 2009; Dauphas et al. 2014).

Xe isotopic correlations are used in Figures 1 and 2 to study the relationships of PPD-Xe with other components. In Figure 1,¹³⁰Xe is used for normalization, as this isotope is free of interferences from fission components. Corrections for spallation components were applied, based on known spectra and assigning¹²⁶Xe excesses to spallation Xe. Figure 1 shows a 136 Xe/ 130 Xe versus 132 Xe/ 130 Xe plot, using data from 238 Table 1. Shifts expected for fission components (244 Pu, 238 U, and neutron-induced fission of 235 U) and for addition of Xe-HL components (Huss & Lewis 1994), as well as those from mass fractionation, are indicated.

SW Xe data (Genesis, Pesy-S, lunar SW) and Chass-S Xe are clustered, but we note that the $^{136}{\rm Xe}/^{130}{\rm Xe}$ ratio in Genesis Xe (Meshik et al. 2014) plots differently from the other data. SW Xe was implanted in the different target materials at different times, while Chass-S Xe represents a Martian interior component.

 Table 1

 Isotope Abundance Ratios in Solar System Material; Spallation Corrected Literature Data (Generally Using the ¹²⁶Xe/¹³⁰Xe Ratios) Are Listed

Comp	¹³⁶ Xe/ ¹³⁰ Xe	¹³⁴ Xe/ ¹³⁰ Xe	¹³² Xe/ ¹³⁰ Xe	¹³¹ Xe/ ¹³⁰ Xe	¹²⁹ Xe/ ¹³⁰ Xe	¹²⁸ Xe/ ¹³⁰ Xe	¹²⁶ Xe/ ¹³⁰ Xe	¹²⁴ Xe/ ¹³⁰ Xe
Genesis	1.8188	2.2370	6.061	5.004	6.306	0.5103	0.02521	0.02976
(1)	± 0.0057	± 0.0069	± 0.015	± 0.014	± 0.016	± 0.0022	± 0.00055	± 0.00043
Chass-S	1.8012	2.205	6.039	4.978	6.520	0.5112	0.0255	0.0295
(2)	± 0.0091	± 0.018	± 0.027	± 0.029	± 0.034	± 0.0049	± 0.0011	± 0.0011
Pesy-S	1.8058	2.2325	6.070	4.978	6.337	0.5091	0.0262	0.0295
(3)	± 0.0055	± 0.0098	± 0.028	± 0.016	± 0.030	± 0.0040	± 0.0010	± 0.0013
SW	1.7971	2.2068	6.020	4.980	6.273	0.5102	0.0255	0.0295
(4,5)	± 0.0055	± 0.0090	± 0.033	± 0.017	± 0.042	± 0.0054	± 0.0008	± 0.0017
Abee	1.9684	2.3475	6.183	5.045	n.a.	n.a.	0.0267	0.0287
(6)	± 0.0020	± 0.0098	± 0.030	± 0.027			± 0.0022	± 0.0010
OC-Xe	1.963	2.344	6.174	5.060	6.420	0.5110	0.0255	0.0286
(7)	± 0.004	± 0.005	± 0.011	± 0.008	± 0.011	± 0.0020	± 0.0002	± 0.0003
Plan.	1.942	2.318	6.135	5.038	6.380	0.5100	0.0254	0.0289
(8)	± 0.006	± 0.007	± 0.009	± 0.007	± 0.012	± 0.0020	± 0.0002	± 0.0003
Q-Xe	1.959	2.342	6.165	5.049	6.387	0.509	0.0252	0.0283
(9)	± 0.009	± 0.009	± 0.023	± 0.018	± 0.031	± 0.002	± 0.0003	± 0.0004
Kenna-Ur	1.916	2.313	6.136	5.027	6.358	0.508	0.0254	0.0289
(10)	± 0.006	± 0.006	± 0.011	± 0.010	± 0.015	± 0.002	± 0.0002	± 0.0003
Orgueil	2.391	2.636	6.215	5.144	6.540	0.5265	0.02741	0.03276
(11)	± 0.014	± 0.012	± 0.015	± 0.017	± 0.026	± 0.0027	± 0.00017	± 0.0003
Mars Atm.	2.269	2.585	6.529	5.180	15.562	0.4728	0.0210	0.0230
(12)	± 0.016	± 0.011	± 0.021	± 0.017	± 0.041	± 0.0069	± 0.0011	± 0.0011
Earth Atm	2.17569	2.56341	6.60982	5.21551	6.48503	0.46922	0.02186	0.02337
(13)	± 0.00112	± 0.00046	± 0.00064	± 0.00050	± 0.00271	± 0.00019	± 0.00005	± 0.00008
Sol_Xe	1.8057	2.2202	6.0475	4.9850	6.3053	0.51020	0.02560	0.02957
(this work)	± 0.0094	± 0.0169	± 0.0225	± 0.0127	± 0.0320	± 0.00086	± 0.00042	± 0.00013
U-Xe	1.657	2.126	6.047	4.996	6.286	0.5083	0.02534	0.02928
(14)	± 0.0003	± 0.0004	± 0.0006	± 0.0006	± 0.0006	± 0.0006	± 0.00013	± 0.00010

Note. Sources of the literature data are listed below.

References.(1) Meshik et al. (2014), (2) Mathew & Marti (2001), (3) Mathew & Marti (2003), (4) Wieler (2002 and references therein), (5) Pepin (2000, 2006, 2013), (6) Lee et al. (2009), (7) Lavielle & Marti (1992), (8) Huss et al. (1996) (9) Wieler et al. (1992), (10) Wilkening & Marti (1976), (11) Frick & Moniot (1977), (12) Marti & Mathew (1998), (13) Valkiers et al. (1998) (14) Pepin (2000).

Trapped meteoritic Xe components are consistent with mixtures of mass-fractionated SW Xe plus Xe-HL (Figure 1). Figure 2 shows that fission gases can be excluded. Meteoritic components OC-Xe, Q-Xe, and Abee-Xe are consistent for all Xe isotopes. The Orgueil-residue data plot on a mixing line of a fractionated SW Xe and Xe-HL. While Mars atmospheric Xe is consistent with mass-fractionated SW, the terrestrial atmospheric Xe signature is not. No fission Xe component can produce the observed shift off the mass-fractionation line. A simple fractionation relationship of terrestrial Xe to either solar or meteoritic Xe, as was suggested (Pujol et al. 2011; Marty 2012), is not possible as pointed out earlier (Pepin 2000, 2006).

The 134 Xe/ 136 Xe versus 132 Xe/ 136 Xe for meteoritic Xe data is not consistent with mixtures of SW Xe and fission components (Figure 2). However, FVM-Xe (Marti et al. 1989) shows a shift in the direction of 235 U(n, f), neutron-induced fission. If trapped meteoritic Xe represent mixtures of mass-fractionated SW-Xe and Xe-HL, the data should fall inside the sector determined by these two lines. The isotopic systematics in Figure 2 are improved by replacing Genesis Xe by the Sol-Xe composition, where the Sol-Xe is the average of the four data sets Genesis-Xe, Chass-Xe, Pesy-Xe, and SW-Xe.

In Figure 3, Sol-Xe data are used to show the massfractionation effects and shifts due to Xe-HL. Mars atmospheric Xe is consistent with a mass-fractionated Sol-Xe component corresponding to 3.77%/amu (Marti & Mathew 1998). An additional ²⁴⁴Pu-fission component (2.5% of ¹³⁶Xe) was inferred, but Pepin (2000) suggested that a hydrodynamic fractionation is also consistent with observed abundances. Xe in Orgueil-residue (Frick & Moniot 1977) requires only the addition of a Xe-HL component, but a better fit is obtained when the Sol-Xe component is fractionated by 1 permil/amu. Meteoritic Xe components (OC-Xe, Q-Xe, and Abee-Xe) are all consistent with Sol-Xe mass fractionated by 0.9%/amu and Xe-HL components corresponding to 2.7% of the ¹³⁶Xe abundance. The Kenna ureilite shows a mass fractionation of 0.6%/amu and the Xe-HL component corresponds to 1.8% of ¹³⁶Xe.

Xe components in terrestrial well gases were studied by Caffee et al. (1999) and Lee et al. (2009), who observed atmospheric Xe, 238 U, and 244 Pu fission components and a minor component of either solar or Abee-Xe composition. Enstatite chondritic matter is considered to be present in planet Earth and also in inner solar system matter (Dauphas et al. 2014).

3. OTHER PPD-Xe COMPONENTS

As discussed, solar system components include Sol-Xe and mass-fractionated components thereof, Xe-HL, terrestrial atmospheric Xe, and FVM-Xe. Another presolar Xe-G component with excesses of ¹²⁸Xe and ¹³⁰Xe was isolated in SiC and graphite of carbonaceous chondrites. This PPD



Figure 1. ¹³⁶Xe/¹³⁰Xe vs. ¹³²Xe/¹³⁰Xe in solar system components. Expected shifts due to the addition of fission Xe and Xe-HL and due to the mass-fractionation process are indicated. The bottom panel expands the region with solar and meteoritic Xe compositions.

component has the signature of products from s-process synthesis that were probably implanted as ions, but its origin is not well understood. The s-process signature does indicate an origin from the He-burning shell of carbon-rich AGB stars (Gallino et al. 1990). Xe-G is a minor component and is not documented in our isotopic correlations, but Meshik et al. (2014) suggest that it might account in part for differences between solar and meteoritic components. Zr isotopic variations were recently reported as evidence for the heterogeneous distribution of s-process products in the solar system (Akram et al. 2015).

The replacement of Genesis Xe by Sol-Xe slightly decreased the heavy isotopes ¹³⁶Xe and ¹³⁴Xe relative to ¹³⁰Xe, while ¹³¹Xe is little affected. Similar isotopic shifts are also produced when a few % Xe with terrestrial atmospheric composition is subtracted from Genesis data. Therefore, a look at uncertainties introduced by air blank correction procedures in Genesis data reduction is useful. Genesis blanks were corrected differently in different laboratories. Meshik et al. (2014) corrected for blanks by extrapolating measured ratios to a predetermined solar value of ⁸⁴Kr/¹³²Xe. The ¹²⁹Xe/¹³²Xe in Genesis foils as reported by several laboratories shows that a 3% atmospheric blank correction increases this ratio only 2 permil. The 1.0404



Figure 2. 134 Xe/ 136 Xe vs. 132 Xe/ 136 Xe in solar system components. Expected shifts due to the addition of fission Xe and Xe-HL and due to the mass-fractionation process are indicated.

reported by Meshik et al. (2014) is similar to values of 1.045 (Crowther & Gilmour 2013), 1.05 (Heber et al. 2009), and 1.06 (Vogel et al. 2011). Meshik et al. (2014) note that there is some uncertainty in the solar ratio 84 Kr/ 132 Xe, but reported values overlapping within uncertainties.

4. EARTH'S ATMOSPHERIC Xe

The Xe isotopic abundances in the Earth's atmosphere (Table 1) show a distinct composition (Figure 1). A parent Xe reservoir, termed U-Xe (Pepin 2000, 2006), that underwent a postulated hydrodynamic fractionation process in the terrestrial environment, received much attention, but an identification of this reservoir in the solar system is lacking. The U-Xe isotopic composition (Table 1) is, with the exception of the heaviest three isotopes, similar to Sol-Xe or Genesis Xe. As no known fission Xe spectrum matches the excesses in the heavy isotopes, the presence of fractionated fission components was suggested (Igarashi 1995). We discuss a possible origin based on PPD-Xe components.

Figure 3 shows that several Xe compositions display variable amounts of Xe-HL. If this component was carried into the PPD by individual nanodiamonds, then only a few in a million diamonds were actually required as carriers. Isotopic variations in meteorites, including refractory elements (Dauphas et al. 2014; Steele & Boehnke 2015), suggest that the PPD was not totally homogeneous. Since Xe in meteoritic minerals contains variable amounts of Xe-HL, PPD matter at different locations must have accumulated varying amounts of nanodiamonds. A location with less nanodiamonds will lack in heavy ¹³⁴Xe and ¹³⁶Xe, compared to Sol-Xe. This is what is observed in mass-fractionated terrestrial Xe. The U-Xe composition (Pepin 2000, 2006) and a primordial Xe composition reported by Takaoka (1972), based on Xe isotopic correlations, both show low ¹³⁴Xe and ¹³⁶Xe abundances. The U-Xe reservoir, after hydrodynamic fractionation, requires the addition of ²⁴⁴Pu-fission Xe to match the terrestrial atmospheric reservoir.

We calculate a pathway, starting in a PPD location with less than average nanodiamonds. In this case, the Xe-PR (PR for precursor) composition differed from Sol-Xe, lacking some Xe-HL. The deficit used here is 5% at ¹³⁰Xe, an isotope not



Figure 3. 136 Xe/ 130 Xe vs. 132 Xe/ 130 Xe in solar system components. Expected shifts due to the addition of mass fractionation and due to the addition of Xe-HL are indicated.

affected by fission Xe. This Xe-PR reservoir is then subjected to a mass-fractionation process of 3.77% per amu, as found for the atmospheres of Mars and Earth. When comparing the mass-fractionated Xe-PR reservoir to terrestrial atmospheric Xe, we calculate that deficits on the heavy Xe isotopes are consistent with the spectrum of ²⁴⁴Pu fission Xe within uncertainties. The fission ¹³⁶Xe to be added is 6.3% of the atmospheric ¹³⁶Xe abundance, exceeding the amount that can be produced from terrestrial ²³⁸U (<1%). The calculated Xe-PR data are given here, for ease of comparing with U-Xe data: ¹²⁴Xe:¹²⁶Xe:¹²⁸Xe:¹²⁹Xe:¹³⁰Xe:¹³¹Xe:¹³²Xe:¹³⁴Xe:¹³⁶Xe = 0.0283:0.0250:0.5062:6.277:1.00:4.960:6.025:2.120:1.662.

The ratio ¹²⁸Xe/¹³⁰Xe is not affected by fission components, and differs by 4 permil from U-Xe (the lighter isotopes have larger uncertainties and depend on the Xe-HL composition; a match for ¹³⁶Xe in U-Xe is obtained by subtracting 5.17% ¹³⁰Xe-HL from Sol-Xe). Igarashi (1995) reported an approach to determine "Primitive Earth Xe," using the intercept of a mass-fractionated atmospheric Xe with the Xe multi-dimensional isotopic correlations of Xe data from carbonaceous meteorites. The correlations revealed that Xe in carbonaceous chondrites has additional minor components. This "Primitive Earth Xe" data also differed from U-Xe.

Thus, an Earth-Xe precursor can be located in a PPD-Xe environment with variable Xe-HL mixing ratios. Existing data on Martian atmospheric Xe do not indicate a deficit of Xe-HL. No model is currently able to explain the nucleosynthetic origin of Xe-HL, as the heavy Xe isotopes do not match an r-process Xe component, as observed in solar Xe (Kratz et al. 2014). Possibly p- and r-process products were implanted into carriers, into very few, as only one atom of Xe-HL is observed in about one million nanodiamonds. Thus, varying mixing ratios of p- and r- products in molecular clouds or in the PPD are possible, although no variability in excesses of light and heavy Xe isotopes has been documented for Xe-HL. Additional nucleo-synthetic Xe products are present in Xe-HL, including an s-process component, and these may have been added in the parent molecular cloud.

As we do not have more information on the fractionation process itself and the timing of acquisitions of inner planet atmospheres, we cannot rule out the possibility that the nebular Marti & Mathew

environment has evolved. The atmospheres of planets Earth and Mars contain ²⁴⁴Pu-fission Xe and radiogenic ¹²⁹Xe components.

5. EVOLVED COMPOSITIONS

Pujol et al. (2011) reported a fractionated Xe isotopic composition as observed in old quartz samples of the Dresser Formation (Australia), intermediate between modern atmospheric and chondritic OC-Xe. Pepin (2013) offered an alternative interpretation, suggesting an admixture of a U-Xe component from the mantle. It is not clear that a U-Xe component is present in the mantle, but components of either solar Xe (Caffee et al. 1999) or Abee-Xe (Lee et al. 2009) are found in well gases and may produce similar results.

The FVM-Xe, as observed in temperature steps of chondritic metal separates (Marti et al. 1989), shown in Figure 2, is shifted in the direction of 235 U neutron fission Xe. A large PPD-Xe inhomogeneity may be indicated in these metals or in inclusions instead of large neutron fluences. Ignoring neutron-induced fission and 248 Cm fission, Forest Vale Metal Xe in Figure 2 can be explained by the addition of 244 Pu fission Xe to a PPD reservoir that is strongly deficient in Xe-HL. In this case, the Xe-PR source is lacking more than 5% nanodiamonds as Xe-HL is depleted by about 10%, relative to Sol-Xe. There are some uncertainties in the data regarding mass fractionation and spallation, but further work on these metals should be of interest.

We have not discussed mechanisms for isotopic fractionation of atmospheric Xe. In addition to the hydrodynamic evolution, processes in dusty plasmas, like low-energy ion implantation, can produce isotopic fractionations (Ponganis et al. 1997). The atmospheres of Mars and Earth reveal large fractionations, while meteoritic components show much smaller fractionations, and Xe in the Orgueil residue is only fractionated by <1%.

The Earth's noble gas elemental abundance ratios with low or "missing Xe" abundances were addressed in studies of Xe reactions with olivine in depth (Sanloup et al. 2011), in reactions with metal at temperatures and pressures of the Earth's core (Zhu et al. 2014), and in work on Xe and Ar solubilities in perovskite (Shcheka & Keppler 2012). Further, an ionization model (Hebrard & Marty 2014) was considered in the context of a photochemical evolution of the atmosphere, but also in this case there is no indication of a fractionation relationship with Sol-Xe.

6. CONCLUSIONS

The term PPD-Xe is used for components present in precursor molecular cloud material at the time of formation of the solar disk. The discussed components (Table 1), and their isotopic make-up as studied in Figures 1, 2, and 3, include SW Xe (Sol-Xe), atmospheres of Earth and Mars, meteoritic Xe, Xe-HL, and fission Xe components. As discussed, Xe-PR was observed in presolar grains but is not documented in the figures.

The Genesis SW Xe isotopic composition and SW isotopic abundances of implanted Xe (lunar and enstatite achondrite regoliths), as well as the Martian interior (Chass-S) Xe, agree within errors and determine the average composition of Sol-Xe. There are indications that uncertainties in the Sol-Xe data could be reduced if Genesis data from an extrapolated set of ratios is used, corresponding to an SW ratio 84 Kr/ 132 Xe = 9.3.

The isotopic compositions of OC-Xe, Q-Xe, and Abee-Xe agree within errors. This composition consists of Sol-Xe, mass fractionated by 0.9%/amu, plus an Xe-HL component of 2.7% of the ¹³⁶Xe abundance.

Terrestrial Xe is not related to solar or chondritic Xe by a mass-fractionation process, as implied by Pujol et al. (2011) and Marty (2012). Pepin (2000) inferred a precursor composition U-Xe for the terrestrial reservoir. We calculate that a precursor composition Xe-PR in the PPD source that is lacking 5% ¹³⁰Xe-HL (relative to Sol-Xe) is similar to U-Xe. Therefore, solar system Xe data suggest that that PPD was heterogeneous and that isotopic abundances were determined by variable mixing ratios with the presolar component Xe-HL.

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