The Nature of Pristine Noble Gases in Mantle Plumes

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High-precision noble gas data show that the Hawaiian and Icelandic mantle plume sources contain uniquely primitive neon that is composed of moderately nucleogenic neon-21 and a primordial component indistinguishable from the meteoritic occurrence of solar neon. This suggests that Earth's solar-type rare gas inventory was acquired during accretion from small planetesimals previously irradiated by solar wind from the early sun. However, nonradiogenic argon, krypton, and xenon isotopes derived from the mantle display nonsolar compositions and indicate an atmosphere-like fingerprint that is not due to recent subduction.

Noble gases are important tracers that help decipher differentiation processes on Earth. They provide evidence for the presence of a severely degassed shallow mantle and a less degassed deep reservoir associated with mantle plumes (*1–4*). Both contain juvenile helium and solar-type neon that cannot be recycled from the atmosphere or crust (*5–7*). They acquired different excesses of radiogenic, nucleogenic, and fissiogenic isotopes (^{4}He, ^{21}Ne, ^{40}Ar, ^{129}Xe, and ^{131,132,134,136}Xe) that formed after an early period of massive mantle degassing. The elemental and isotopic composition of the shallow mantle, the mid-ocean ridge basalt (MORB) source, is relatively well established (*8*, *9*), but noble gas characteristics of deep mantle plumes have remained a matter of debate (*10*). Basalts from the Hawaiian (*1*, *5*, *7*, *11*) and the Icelandic (*12*) mantle plume contain highly primitive (*13*) helium and neon. However, the isotopic composition of the heavier noble gases in these primitive mantle plumes remains uncertain because previously analyzed samples were severely contaminated by atmospheric rare gases (*10*). To elucidate this long-standing problem, we performed stepwise in vacuo crushing experiments on volatile-rich dunite xenoliths, KK27-9 and -12 (*14*, *15*), from Loihi seamount, Hawaii, and subglacially erupted basalt glasses, Dice 10 and 11, from the Reykjanes Peninsula, Iceland (*16*).

Loihi dunites have ^{3}He/^{4}He ratios with a mean value of 24.5 ± 0.5 times the atmospheric ratio R_A (Table 1), which is typical for the Hawaiian mantle source (*1*, *5*, *7*, *11*). The ^{3}He/^{4}He ratios of the Iceland glasses are plume-like (16.8 to 18.8 R_A) but are significantly lower than the maximum values (37 ± 2 R_A) found for the Iceland plume (*12*). The Ne isotopic compositions of KK27 dunites and Dice glasses (Fig. 1A) are indistinguishable from each other and are consistent (*17*) with the Loihi trend defined by analyses of submarine basaltic glass (*7*). They contrast with the MORB correlation line (*6*) and high-precision analyses of popping rock 2πD43 (*8*, *18*). The linear trends result from recent mixing of the mantle endmember, characterized by solar-like ^{20}Ne/^{22}Ne ratios, with atmospheric contaminants. The steeper slope of the Loihi trend can be interpreted as occurring because of the lower degree of degassing of this plume reservoir and the implicit higher contribution of primordial ^{22}Ne.

Loihi dunite KK27-9, Icelandic glasses Dice 10 and 11, and mid-Atlantic popping rock 2πD43 yield indistinguishable maximum ^{20}Ne/^{22}Ne ratios with a mean of 12.49 ± 0.06 in the advanced crushing steps (Table 1) (*8*, *18*), indicating that this value represents the original endmember of the mantle source preserved in the most retentive vesicles of the rocks. This ratio is different from the solar ratio of 13.80 ± 0.10 represented by present-day solar wind (*19*) but indistinguishable from the meteoritic occurrence of solar neon (Ne-B) ^{20}Ne/^{22}Ne = 12.52 ± 0.18 (*20*, *21*). The reproducibility of this Ne-B value in samples from a variety of localities with contrasting tectonic settings and inherently different mantle structures indicates that Ne-B represents the initial solar Ne component within Earth. This conclusion agrees with previously published Ne data from mantle-derived samples, taking into account the 2σ uncertainties (*22*).

The stepwise crushing data (Table 1) yield a correlated excess of ^{20}Ne and ^{40}Ar (Fig. 1B), again reflecting a mixture of noble gases from vesicles with the mantle endmember

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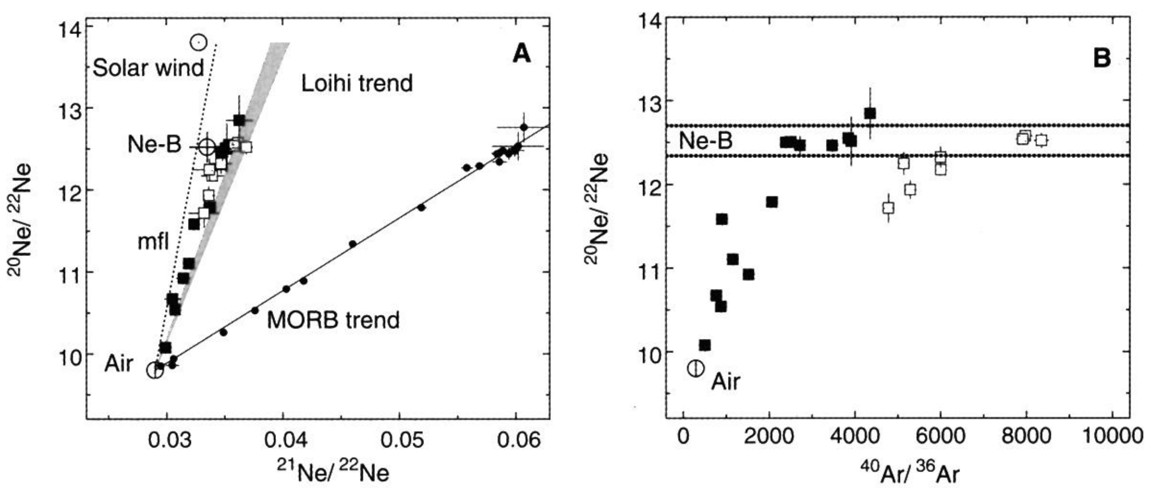

Fig. 1. (**A**) Neon three-isotope plot showing Loihi dunite (□), Dice basalt glass (■), and popping rock 2πD43 (●) (*8*, *18*) data. Clustering of data at ^{20}Ne/^{22}Ne ≈ 12.5 suggests Ne-B as the solar Ne component in Earth's mantle. mfl, mass fractionation line. (**B**) Excess of solar ^{20}Ne and radiogenic ^{40}Ar is correlated, indicating mixture of atmospheric contaminating noble gases and an inherent mantle component. Maximum ^{40}Ar/^{36}Ar ratios at Ne-B composition are constant for Loihi dunites, indicating uncontaminated mantle argon with ^{40}Ar/^{36}Ar ≈ 8000, whereas Dice samples display considerable scatter, indicating an additional atmospheric contaminant (*23*).

and from vesicles containing atmospheric contaminants. Ratios of $^{40}Ar/^{36}Ar$ at Ne-B composition are constant for Loihi dunites, indicating a value of 8070 ± 240 for the Loihi mantle source, which is somewhat higher than previously deduced from the analyses of basalt glasses (*7*). For the Dice glasses, $^{40}Ar/^{36}Ar$ ratios at Ne-B composition are variable, presumably due to another Ar-rich contaminant (*23*), which prevents precise definition of the mantle source $^{40}Ar/^{36}Ar$ ratio. We also observed small excesses of radiogenic ^{129}Xe and fissiogenic $^{131,132,134,136}Xe$ (Fig. 2 and Table 1), which indicates anomalous Xe in mantle-derived rocks with Loihi-type primitive (i.e., ^{21}Ne-poor) neon. Excess of radiogenic and fissiogenic Xe are correlated and indistinguishable from the MORB array (*9*) within uncertainties. Xenon in these rocks may be contaminated by MORB-type Xe (*24*); alternatively, it may be derived from the plume sources. In the latter case, the relative contributions of fission Xe from ^{238}U and ^{244}Pu could be different from the MORB source (*9*), which cannot be quantified here because of the analytical uncertainties of the fission isotopes $^{131,132,134,136}Xe$ (*22*). If excess of ^{129}Xe is indigenous to the Hawaiian and Icelandic plume sources, its presence would indicate early degassing more stringently than the ^{40}Ar excess.

The presence of solar-type He and Ne in Earth led to a number of discussions about the isotopic composition of the heavy noble gases (*8*, *18*, *25*, *26*). The primordial nuclides ^{38}Ar and ^{36}Ar allow us to quantify the contribution of solar-type Ar in the mantle because the atmospheric and solar-wind $^{38}Ar/^{36}Ar$ ratios differ by about 7% (*26*). Table 1 shows that the $^{38}Ar/^{36}Ar$ ratio at high $^{40}Ar/^{36}Ar$ and—implicitly—high $^{20}Ne/^{22}Ne$ (i.e., after correcting for recent local atmospheric contamination) is atmosphere-like (0.1880 ± 0.0003) or planetary (*27*), similar to Ar associated with Ne-B (0.186 ± 0.004) (*20*). The 2σ confidence bands of a line fit (*22*) restrict the contribution of solar-wind Ar ($0.1724 < {}^{38}Ar/^{36}Ar < 0.1786$) to less than ≈10% at $^{20}Ne/^{22}Ne \approx 12.5$, a similar limit as found for MORB glass 2πD43 (*18*). Nonradiogenic Kr and Xe isotopes may also serve to distinguish between solar-wind, planetary, or atmosphere-like components. Mean values of Kr isotopes (Fig. 3A) (*22*) can be reconciled much better with atmosphere-like Kr than solar Kr (*28*). Values for nonradiogenic Xe isotopes obtained by linear extrapolation to a $^{129}Xe/^{130}Xe$ value of 7.0 (*22*) do not agree with solar or planetary composition (Fig. 3B).

Solar-like He and Ne with atmosphere-like nonradiogenic Ar, Kr, and Xe isotopes are a property of the whole mantle because the mantle source of MORBs also has this characteristic (*6*, *9*, *18*). Atmosphere-like (*29*) Ar, Kr, and Xe do not necessarily

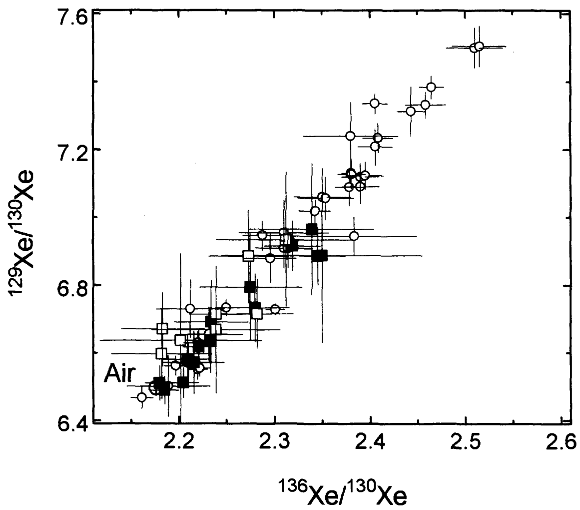

Fig. 2. Loihi dunites (□) and Icelandic glasses (■) contain correlated excess of ^{129}Xe and ^{136}Xe in similar proportions as the MORB reservoir (○, only data with $\Delta^{129}Xe/^{130}Xe < 0.1$) (*8*, *9*).

Table 1. Noble gas data of Loihi dunites KK27-9 and -12 and Icelandic subglacial glasses Dice 10 and 11. Concentrations are in cm³/g at standard temperature and pressure. Numbers in brackets are 1σ uncertainties and refer to last digit given.

Crush no.	^{4}He × 10^{-8}	^{22}Ne × 10^{-12}	^{36}Ar × 10^{-12}	^{84}Kr × 10^{-12}	^{130}Xe × 10^{-14}	$^{3}He/^{4}He$ (R_A)	$^{20}Ne/^{22}Ne$	$^{21}Ne/^{22}Ne$	$^{38}Ar/^{36}Ar$	$^{40}Ar/^{36}Ar$	$^{129}Xe/^{130}Xe$	$^{136}Xe/^{130}Xe$
						KK27-9 (1.699 g)						
40×	8.3	2.1	17.6	0.64	0.97	24.6 (7)	11.93 (11)	0.0336 (4)	0.1875 (3)	5284 (103)	6.67 (19)	2.24 (7)
150×	14.7	3.4	27.1	1.11	1.95	25.0 (7)	12.17 (6)	0.0339 (9)	0.1876 (3)	5998 (117)	6.67 (11)	2.18 (4)
200×	15.5	3.2	21.8	0.91	1.56	24.5 (7)	12.57 (6)	0.0362 (5)	0.1881 (3)	7964 (155)	6.88 (14)	2.27 (4)
300×	16.5	3.6	24.4	1.11	2.26	24.8 (7)	12.54 (6)	0.0359 (5)	0.1879 (3)	7908 (154)	6.71 (10)	2.28 (4)
600×	27.6	6.6	40.3	2.18	4.87	24.5 (6)	12.52 (8)	0.0369 (5)	0.1884 (3)	8340 (162)	6.71 (8)	2.24 (3)
						KK27-12 (1.003 g)						
100×	4.3	3.6	30.0	0.94	1.28	25.4 (10)	11.72 (17)	0.0332 (13)	0.1871 (4)	4779 (95)	6.93 (20)	2.31 (7)
200×	4.5	2.6	26.9	0.74	1.37	23.8 (9)	12.25 (13)	0.0337 (15)	0.1879 (4)	5135 (102)	6.64 (26)	2.20 (9)
500×	5.3	3.7	34.3	1.21	2.72	24.1 (8)	12.32 (12)	0.0347 (9)	0.1877 (3)	5994 (118)	6.60 (13)	2.18 (5)
						Dice 10 (1.841 g)						
10×	37.9	7.1	279.6	10.27	8.48	17.2 (5)	11.58 (4)	0.0324 (3)	0.1883 (2)	903 (18)	6.58 (8)	2.21 (2)
25×	87.2	28.2	755.7	20.70	21.21	17.6 (5)	10.67 (6)	0.0305 (7)	0.1879 (3)	768 (15)	6.51 (5)	2.21 (2)
50×	79.7	11.6	166.5	11.27	13.04	16.8 (5)	11.79 (2)	0.0338 (2)	0.1880 (3)	2066 (40)	6.51 (5)	2.18 (2)
100×	117.0	12.4	122.4	4.65	3.79	17.0 (5)	12.55 (2)	0.0354 (2)	0.1880 (3)	3843 (75)	6.89 (9)	2.34 (3)
200×	94.1	9.4	147.0	4.99	3.97	17.5 (5)	12.50 (2)	0.0351 (3)	0.1881 (3)	2387 (47)	6.73 (10)	2.28 (4)
250×	55.1	5.0	77.7	2.54	3.08	17.1 (4)	12.50 (4)	0.0351 (4)	0.1880 (3)	2508 (49)	6.64 (10)	2.23 (3)
450×	58.3	4.9	48.5	1.82	2.46	17.2 (5)	12.46 (7)	0.0347 (5)	0.1887 (4)	3467 (68)	6.69 (12)	2.23 (4)
						Dice 11 (1.877g)						
10×	83.1	74.7	1503.8	34.60	21.92	17.1 (5)	10.08 (2)	0.0299 (2)	0.1879 (2)	505 (10)	6.49 (4)	2.19 (1)
25×	45.2	8.1	173.7	5.83	4.37	17.3 (5)	11.10 (7)	0.0319 (3)	0.1877 (3)	1154 (23)	6.57 (9)	2.22 (3)
50×	170.0	73.9	1179.9	27.45	16.99	17.5 (5)	10.54 (2)	0.0307 (2)	0.1880 (2)	878 (17)	6.62 (5)	2.22 (2)
100×	105.7	26.8	318.5	8.56	5.86	16.8 (5)	10.92 (4)	0.0315 (6)	0.1885 (2)	1524 (30)	6.92 (8)	2.32 (3)
200×	53.7	5.1	70.9	1.62	1.54	17.1 (5)	12.46 (11)	0.0348 (5)	0.1862 (3)	2716 (53)	6.79 (16)	2.28 (5)
250×	21.5	1.9	15.6	0.52	0.49	18.8 (8)	12.51 (29)	0.0352 (11)	0.1877 (4)	3912 (82)	6.89 (26)	2.35 (11)
500×	22.4	1.7	13.5	0.54	0.88	17.1 (11)	12.85 (31)	0.0363(11)	0.1877 (4)	4350 (93)	6.97 (20)	2.34 (7)

require subduction of atmospheric nuclides. A recent study (30) suggested that a solar-atmospheric noble gas hybrid in Earth's interior was acquired in precursor planetesimals by gravitational capture and accompanied fractionation of solar-type noble gas species. Our finding that Ne-B may be the solar neon component in Earth rules out such models for neon. However, it strengthens the general argument that precursor planetesimals were responsible for Earth's primordial noble gases, because Ne-B is the commonly found solar-type Ne in meteorites (20, 21). Its origin is related to solar corpuscular radiation, and the occurrence inside a large planet like Earth has more far-reaching implications. It requires mechanisms to effectively incorporate solar irradiation in Earth, e.g., an active sun at a stage when accreting planetesimals were small and the accretion disk was relatively transparent, i.e., had lost most of its gaseous or volatile component (31). A model involving early irradiation in small planetesimals was previously advocated to explain the high fraction of gas-rich carbonaceous chondrites and the irradiation features of single meteoritic grains (32, 33).

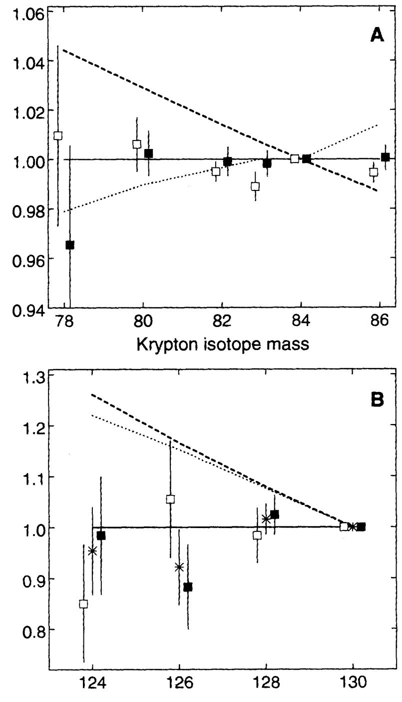

Fig. 3. **(A)** Isotopic composition of mantle krypton (□, KK27-9 all extractions; ■, Dice 10 extractions with $^{20}Ne/^{22}Ne > 12.45$). **(B)** Isotopic composition of mantle xenon (□, Loihi dunite; ■, Dice; and ∗, total data extrapolated to $^{129}Xe/^{130}Xe = 7.0$). Values (22) are best reconciled with atmosphere-like composition (solid line), excluding solar composition (dashed line), or excluding planetary composition (dotted line).

Such a scenario contrasts with models where the massive proto-Earth gravitationally captured a dense primary atmosphere within the dense solar nebula (34): After incorporation of solar-type noble gases into the mantle, partial atmospheric loss caused isotopic fractionation and established the planetary-like pattern of Earth's atmosphere (28). These models require subduction of atmospheric nuclides to explain the atmosphere-like nonradiogenic heavy noble gas nuclides in the mantle sources. However, subduction processes must have been limited if they occurred after the dying out of ^{129}I; otherwise ^{129}Xe anomalies in the mantle would have been erased. Moreover, recent subduction into a massively degassed MORB mantle and less degassed plume reservoirs should result in different $^{20}Ne_{solar}/^{36}Ar_{atmospheric}$ ratios for the two mantle domains (35). This, however, is not indicated by various data sets (7, 30), including data presented here. It follows that subduction—if it occurred—happened before the last homogenization of the two mantle reservoirs, which is currently regarded as most feasible to have occurred early in Earth's history (1, 35).

References and Notes

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13. Loihi and Icelandic mantle sources have the highest known $^{3}He/^{4}He$ ratios among the world's mantle reservoirs, i.e., the highest proportion of (primitive) primordial relative to radiogenic nuclides.
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22. Web figures 1 through 8 and Web table 1 are available at *Science* Online at www.sciencemag.org/features/data/1048312/shl.
23. Ar-rich means a higher $^{36}Ar/^{22}Ne$ ratio (as well as a higher $^{36}Ar/^{130}Xe$ ratio) compared with the mantle source (8), which could be obtained by fractionating air similar to air dissolved in deep seawater [M. Ozima and F. A. Podosek, *Noble Gas Geochemistry*, (Cambridge Univ. Press, Cambridge, 1983)].
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31. The required clearing of the accretion disk and accompanying gas and volatile loss before the larger terrestrial planets were accreted can be reconciled with the observation that K loss in the inner solar system was not accompanied by fractionation of K isotopes (as would be expected for K loss from large planetary bodies [M. Humayun and R. N. Clayton, *Geochim. Cosmochim. Acta* **59**, 2131 (1995)].
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35. This conclusion can be achieved in particular for upper mantle "steady state" models; i.e., the noble gases have a limited lifetime in the upper mantle ($\approx$1.4 Ga) and the degassing flux of primordial isotopes is compensated by transfer from the lower mantle [D. Porcelli and G. J. Wasserburg, *Geochim. Cosmochim. Acta* **59**, 4921 (1995)]. Because steady state models require the same $^{20}Ne_{solar}/^{36}Ar_{atmospheric}$ ratio of the plume and upper mantle domains, they can only accommodate subduction processes if these occurred exclusively into the lower mantle. Additional subduction into the upper mantle would lead to different $^{20}Ne_{solar}/^{36}Ar_{atmospheric}$ ratios. However, exclusive subduction into plume reservoirs seems to be quite unlikely, so early subduction is more feasible at a time when mass exchange from the upper to the lower mantle was possible.

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