GEOPHYSICAL RESEARCH LETTERS, VOL. 18, NO. 2, PAGES 207-210, FEBRUARY 1991

SOLAR-WIND KRYPTON AND SOLID/GAS FRACTIONATION IN THE EARLY SOLAR NEBULA

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Abstract. Krypton is the best candidate for determining limits on solid/gas fractionation in the early sun because of the smoothness of the odd-mass abundance curve in its mass region, which permits relatively precise interpolations of its abundance assuming no fractionation. Here we calculate the solar-system Kr abundance from solar-wind noble-gas ratios, determined previously by low-temperature oxidations of lunar ilmenite grains, normalized to Si by spacecraft solar-wind measurements. The estimated ^{83}Kr abundance of 4.1 ± 1.5 per 10^6 Si atoms is within uncertainty of estimates assuming no fractionation, determined from CI-chondrite abundances of surrounding elements. This is significant because it is the first such constraint on solid/gas fractionation, though the large uncertainty only confines it to somewhat less than a factor of two.

Chemical Fractionation in the Proto-Sun

Based on astrophysical observations it is widely accepted that the solar system formed from an interstellar cloud that was initially a multi-phase system consisting of gas plus various interstellar grain components. Until temperatures reached the order of 2 x 10^3 K, this multiphase nature must have been present throughout the formation of the sun and the planetary objects. *A priori* it may be unreasonable to assume that the formation of the sun and the solar nebula incorporated these phases in exactly the same relative proportions as in the initial interstellar cloud. The cloud itself was undoubtedly inhomogeneous on small scales, but the degree of inhomogeneity on large scales, of the order of AU, is totally unknown. Differential forces (e.g., magnetic) on different types of grains could have been important, but because of the major role they have played in shaping the solar system, gas/solid separations would seem the most likely fractionation mechanisms.

Details of the physical processes by which these separations could have occurred are complex and difficult to calculate quantitatively, but broad, qualitative outlines can be sketched. For example, if there was a stage in the gravitational collapse in which the sun was essentially totally accreted, but relatively cool, and the planetary material was widely dispersed, the Poynting-Robertson effect could have produced a relatively large flow of solid particles to the sun. Understanding the efficiency of this is complicated by the effects of gas drag which, in turn, would depend on whether or not solid/gas fractionation had already occurred in the nebula. In the absence of nebular gas (H_2, He) the grain flow could have been significant. The surface temperature of the sun is also critical. In the present-day solar system it is possible that in-falling particles are partially or totally vaporized and the products swept out either in the solar wind or by radiation pressure as a component of the so-called beta meteoroids [Grün *et al.*, 1985]. However, even if the present sun cannot accrete mass from particles, preferential accretion of small grains could have been important in the early stages of solar evolution when surface temperatures were lower. Alternately, accretion of planetesimals in comet-like orbits could conceivably have enhanced the solid/gas ratio in the final solar composition [Joss, 1974]. The converse of this is also possible, e.g., if planetesimal formation in stable orbits was very rapid and extensive in the outer solar system, then the sun could have preferentially accreted gas rather than solids.

It is also conceivable that separation was caused by plasma effects (e.g., from intense bipolar jets) in regions where low first-ionization-potential (FIP) elements were preferentially ionized. This would impose on the whole sun the type of FIP fractionations that are now well documented between the photosphere and the solar wind [Geiss and Bochsler, 1985; Bochsler and Geiss, 1989] and solar energetic particles [Breneman and Stone, 1985]. A more speculative alternative to these scenarios would be depletion as a property of the initial interstellar cloud, reflecting long-term galactic gas/solid fractionation, e.g., due to large-scale comet formation in the galaxy [Tinsley and Cameron, 1974].

For solar system depletions the amount of dilution of late in-falling material with bulk solar matter is also an issue. If we assume that prior to H burning the sun was totally convecting (as opposed to the present solar state with only a surface convection zone), then the amount of excess solid material required to produce an observable solid/gas fractionation effect is much larger than for the case of preferential accretion to the sun in its present state, in which case matter would only be added to the surface convection zone. At present the mass of rock-forming elements in the convection zone is about 20 Earth masses, which sets the order of magnitude of the mass requirement for preferential gain (or loss) of solids.

Comparison of the abundance of Kr relative to nearby

Paper number 91GL00213
0094-8534/91/91GL-00213$03.00

non-volatile elements in the solar wind with that estimated by interpolation from CI chondrite data (relative to Si) provides an observational test of preferential accretion, assuming that grain/planetesimal formation occurs under sufficiently high T that Kr was not quantitatively trapped as a solid. The CI interpolation is possible because of the smoothness of CI heavy-element abundances for odd-A nuclei [Suess, 1947; Burnett *et al.*, 1989]. In the absence of fractionation during the formation of the sun, smoothness should permit an accurate average solar-system abundance for Kr to be obtained by interpolation between CI abundances of Br and Rb. There are complications with the CI Br abundance [e.g., Burnett *et al.*, 1989], but these interpolations are possible without Br (e.g., using Se). The probable presence of FIP depletions [e.g. Bochsler and Geiss, 1989; Steiger and Geiss, 1989] in the solar-wind Kr abundance is also a complication, but it does not prevent testing for preferential accretion. For example, let Kr^* be the interpolated abundance. If the solar-wind Kr/Kr^* were significantly lower than the solar-wind/photospheric fractionation factors for C,N,and O, this would be evidence for preferential accretion of solids.

Elemental abundances below Fe ($Z < 26$) are not especially smooth, so it is not possible to make the gas/solid comparison using available solar-wind Ne or Ar data with any degree of confidence. An equivalent analysis for Xe to that given here for Kr is more difficult because Xe corresponds to a local maximum in the solar-system abundance curve, making interpolation difficult. In addition, it is possible that the solar-wind xenon abundance relative to the other noble gases has fluctuated significantly over the age of the solar system [Kerridge, 1980; Becker and Pepin, 1989].

Solar-Wind Gases in Lunar Ilmenites

Direct solar-wind measurements in the range of krypton are not available. The lunar regolith and gas-rich meteorites are sources of solar-wind-implanted material. However, in general solar-wind gases from these materials are fractionated relative to the solar wind itself, even in ilmenite [Eberhardt *et al.*, 1970; 1972; Hübner *et al.*, 1975], which appears to be the most gas-retentive phase [e.g., Frick *et al.*, 1975]. To release potentially unfractionated solar-wind gases, it is necessary to attack the grain surfaces, where solar-wind ions are implanted, without disturbing deeper regions, where a fractionated gas reservoir is found. Frick *et al.* [1988] suggested that this deeply-sited fractionated component derives from inward diffusion of gases previously implanted in grain surfaces, and that gases currently in the surface reservoir were implanted during the most recent exposure to the wind and thus are relatively unaltered by diffusion. They showed that low-temperature oxidation of ilmenite from lunar soil 71501 yielded relative Ne, Ar, Kr, and Xe abundances within 10% of "solar", as inferred by Cameron [1982], and with no evidence of systematic fractionation. An ilmenite separate from soil breccia 79035, considered to contain solar-wind gases of greater antiquity [e.g. Kerridge, 1980], was also subjected to low temperature oxidation. Results fell within the same range with the exception of xenon, which, at ~2 times higher relative to Ne, Kr, and Ar, was suggested to indicate a secular decrease with time in the relative solar-wind xenon flux [Becker and Pepin, 1989] as suggested previously [Kerridge, 1980]. A puzzling feature of these gas releases is the nitrogen abundance, which was consistently more than an order of magnitude greater than expected for solar, and for which the authors could find no completely satisfactory explanation. One possibility [Frick *et al.*, 1988] is that nitrogen diffuses less readily than the noble gases, so that traces of N from many episodes tend to accumulate in the grain surface reservoir, whereas the noble gases do not.

In spite of the possible complications pointed out in the above discussion, we will assume for the following analysis that the relative noble-gas contents from the low-temperature oxidations of 71501 and 79035 ilmenites are representative of the noble-gas ratios in the solar wind.

Solar-Wind $^{83}Kr/Si$

This is the key ratio to compare with the interpolated CI meteorite abundances, which are normalized to Si. It is necessary to calculate the solar wind $^{83}Kr/Si$ ratio indirectly, as direct measurements are not available. Table 1 gives $^{83}Kr/^{20}Ne$ and $^{83}Kr/^{36}Ar$ ratios from the low-temperature oxidation of lunar ilmenites 71501 [Frick *et al.*, 1988] and 79035 [Becker and Pepin, 1989]. The ratios are multiplied by the solar-wind $^{20}Ne/Si$ and $^{36}Ar/Si$ ratios. To arrive at the solar-wind $^{20}Ne/Si$ ratio, spacecraft measurements of $Ne/O = 0.17 \pm .02$ [Bochsler and Geiss, 1989; Bochsler *et al.*, 1986] and $Si/O = 0.19 \pm .04$ [Bochsler, 1989] are used, along with the solar-wind neon isotopic composition of Geiss *et al.* [1972]. The greatest uncertainties are in the direct solar-wind measurements of Ne, Ar, and Si. Using the higher Si/O ratio of $0.22 \pm .07$ from Bochsler and Geiss [1989] and Bochsler [1987] would lower the estimated solar Kr abundance by 15%. For argon the only reported solar-wind measurements are from the Apollo foil noble-gas measurements [Geiss *et al.*, 1972], from which we adopt $^{36}Ar/Ne$. Thus, the $^{36}Ar/Si$ ratio in the table was obtained by:

$$(^{36}Ar/Si)_{SW} = (^{36}Ar/Ne)_{foil}(Ne/O)_{probe}/(Si/O)_{probe} \quad [1]$$

where $(^{36}Ar/Ne)_{foil} = 0.0205 \pm .0050$ [Bochsler and Geiss, 1977] and the other factors are given above. The resulting mean solar-wind ^{83}Kr abundance from Table 1 is $0.98 \pm .04$ ($Si \equiv 10^6$). Table 1 shows that ilmenite 79035, which trapped more ancient solar wind, is in close agreement with the data from 71501.

The calculated solar-wind ^{83}Kr abundance of 0.98 is distinctly lower than interpolated CI abundances, e.g. 5.2 from Anders and Grevesse [1989]. However, the simplest interpretation of this difference is that Kr in the solar wind is also subject to the type of FIP or first-ionization-time depletion already documented for C, N, O, and Ne in the solar wind [e.g. Bochsler and Geiss, 1989] and solar-flare particles [Breneman and Stone, 1985]. These investigations indicate that elements with FIPs greater than ~11 eV are depleted by a constant value relative to elements with lower FIPs [Steiger and Geiss, 1989]. Kr ionizes at 14.0 eV; hence depletion of solar-wind Kr is expected. Applying an estimated FIP correction of 4.2 ± 1.5 [Steiger

TABLE 1. Data used for the calculation of the solar wind ^{83}Kr abundance. Ilmenite data from Frick *et al.* [1988] and Becker and Pepin [1989]; solar wind $^{20}Ne/Si$ and $^{36}Ar/Si$ data from Geiss *et al.* [1972], Bochsler and Geiss [1977], Bochsler *et al.* [1986], Bochsler [1989], and Bochsler and Geiss [1989].

Sample	----- Ilmenites -----		Directly Measured S.W.		^{83}Kr
	$^{83}Kr/^{20}Ne$	$^{83}Kr/^{36}Ar$	$^{20}Ne/Si$	$^{36}Ar/Si$	(Si = 10^6)
71501	1.137 x 10^{-6}		0.83		0.94
	±.047		±.20		±.23
		5.19 x 10^{-5}		0.0183	0.95
		±.34		±.0063	±.33
79035	1.206 x 10^{-6}		0.83		1.00
	±.069		±.20		±.25
		5.59 x 10^{-5}		0.0183	1.02
		±.40		±.0063	±.36

and Geiss, 1989; Anders and Grevesse, 1989] yields an unfractionated ^{83}Kr abundance of 4.1 ± 1.5, in agreement with the CI-interpolated values in Table 2. This is shown in Figure 1 using the CI abundances of Burnett *et al.* [1989] for which the same samples were used for all elements in this mass range.

TABLE 2. Solar wind depletion-corrected ^{83}Kr abundances from the lunar ilmenite and spacecraft data given in Table 1, along with abundance estimates from near-element CI interpolations and s-process systematics.

Source	^{83}Kr (Si = 10^6)
Solar wind (71501 Ilmenite)	4.0 ± 1.4
Solar wind (79035 Ilmenite)	4.2 ± 1.5
Cameron [1982]	4.7
Anders and Grevesse [1989]	5.2
Burnett *et al.* [1989]	4.75
S-process systematics*	5.2 ± 0.9

*as quoted in Anders and Grevesse [1989]; updated from Walter *et al.* [1986]

For xenon, a solar abundance estimate of ^{130}Xe = 0.24 ± .09 (Si = 10^6) is made from 71501 ilmenite data [Frick *et al.*, 1988] using the same conversion and solar-wind fractionation factors as krypton. As mentioned earlier, interpolation of Xe from CI data is more difficult. However, calculations based on s-process systematics may be quantitatively reliable for A ≥ 100 AMU. It is interesting to note that our abundance estimate is very close to the s-process ^{130}Xe value of 0.22 ± .09 [Käppeler *et al.*, 1989]. However, the xenon problem needs more study to be well understood, especially regarding its possible abundance variations with time in the solar wind.

Discussion

Within uncertainty there is no difference between our inferred solar-wind Kr abundance and that interpolated at mass 83 from CI meteorite data on adjacent elements. Thus, following arguments developed above, there is no evidence for gas/solid fractionation in the formation of the sun from the solar nebula. It is important to recognize the major assumption we have made in equating the relative noble-gas abundances of the solar wind with those from two lunar samples. Also, the problem with the high nitrogen to noble gas ratio in the ilmenite data must be resolved before our conclusion is unequivocal.

The propagated errors in the calculated $^{83}Kr/Si$ are relatively large, and the lack of preferential accretion should be regarded as little better than a factor of 2 conclusion. Nevertheless, this is significant, as there are no other observational constraints on this important issue. It is important to repeat the comparison we have made with improved solar-wind data, ideally for nearby elements (Se, Br, Rb, Sr) as well as Kr itself.

In hindsight it is perhaps surprising that the sun appears to have captured gas and dust from the solar nebula in approximately quantitative proportions. Conceivably, this is a crucial constraint on the early stages of solar contraction.

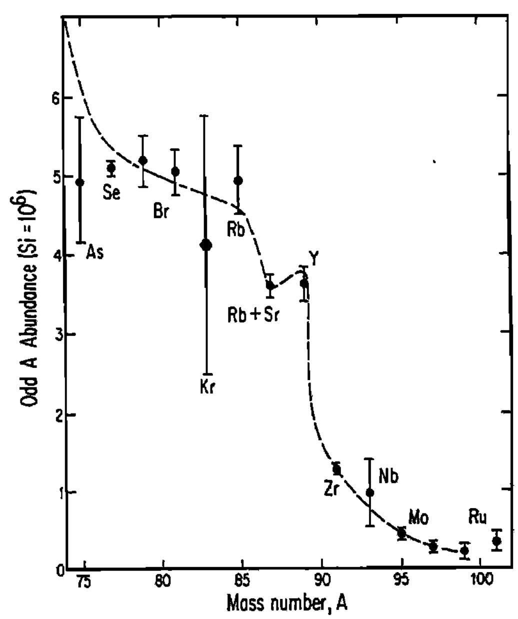

Fig. 1. Solar-system abundances of odd-mass isotopes in the range of A = 75 - 101. All data except Kr are average CI concentrations from Burnett *et al.* [1989], normalized to Si = 10^6. The Kr data point is averaged from the ilmenite results in Table 2, which have been corrected for photosphere/solar-wind fractionation using a value of 4.2 ± 1.5 relative to Si. The solar Kr abundance thus derived is within uncertainty of interpolation and s-process estimates, which assume no solid/gas fractionation.

Acknowledgements. This research was supported in part by NASA grants NAG 9-94 to D. Burnett and NAG 9-60 to R. O. Pepin. J. Kerridge and an anonymous reviewer are thanked for their assistance, and profitable discussions with D. Stevenson are acknowledged.

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(Received August 17, 1990;
revised November 19, 1990;
accepted November 21, 1990)