Volatile Evolution and Loss

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Relative to bulk solar system composition, most meteorites and the terrestrial planets are depleted in volatile elements. This abundance pattern likely arose in the solar nebula rather than during planet formation. The solar system record of volatile element abundances is reviewed and constraints on its origin from isotopic compositions and abundances of presolar grains in primitive meteorites are explored. Major models for the origin of volatility fractionations are described.

1. INTRODUCTION

The CI chondrites are generally regarded as being representative of the bulk elemental composition of the solar system, with the exception of the highly volatile elements hydrogen, carbon, nitrogen, oxygen, and the noble gases (Anders and Grevesse, 1989; Lodders, 2003; Palme and Jones, 2003). Relative to CI chondrites, all other groups of meteorites, as well as bulk terrestrial planets, are depleted in volatile elements. These depletions differ from group to group, but for the most part have the common characteristic of being reasonably smooth functions of volatility (expressed as 50% condensation temperatures). The volatile element patterns of the terrestrial planets and achondrite parent bodies are less smooth, because igneous differentiation has modified trace-element abundances. However, some incompatible elements are not significantly decoupled by igneous differentiation and ratios of volatile to refractory incompatible elements can be used to gauge the level of volatile elements in the bulk planets and parent bodies. The depletion patterns of meteorites have fascinated cosmochemists for decades, vet remain incompletely understood. In this chapter, the volatility patterns of meteorites will be reviewed and their implications for early solar system processes explored.

The subject of volatile elements in meteorites was reviewed in the first *Meteorites and the Early Solar System* volume (*MESS I*) (*Palme et al.*, 1988). Since that time, there has been considerable progress in the field. Although little has changed concerning the basic elemental abundance patterns that are to be explained, the new tools of isotopic mass fractionation and presolar grain abundances allow new constraints to be placed on the mechanism of volatile-element fractionation.

2. THE RECORD

2.1. Elemental Abundances

The CI chondrites are believed to be representative of the bulk elemental composition of the solar system, because abundances of all but the most volatile elements in them match those in the solar photosphere (*Anders and Grevesse*, 1989; *Lodders*, 2003; *Palme and Jones*, 2003). Thus, it is customary to normalize elemental abundances in other solar system objects to those in CI chondrites. In comparing elemental abundances in meteorites and planets with CI chondrites, it is useful to divide the elements into different groups based on their physical and chemical properties (*Larimer*, 1988). Since chondrites and planets have varying quantities of water and organic matter, it is also common to normalize to the abundant moderately volatile element silicon. Magnesium is sometimes used in place of silicon, particularly in sets of analyses collected by neutron activation analysis (silicon cannot be determined by this method).

The primary cosmochemical classification of the elements is based on volatility, with the elements divided into those that are refractory (having equilibrium condensation temperatures higher than the most abundant rock-forming elements, magnesium, silicon, and iron), moderately volatile (having condensation temperatures lower than those of the refractory elements, but higher than that of FeS), and highly volatile (having condensation temperatures below that of FeS). For a gas of solar composition at a total pressure of 10-4 atm, refractory elements have 50% condensation temperatures above 1335 K, moderately volatiles between 1335 and 665 K, and volatiles below 665 K [condensation temperatures from Lodders (2003)]. All these elements have the same relative abundances as the solar photosphere, within uncertainties (Anders and Grevesse, 1989; Lodders, 2003; Palme and Jones, 2003). A fourth group, the atmophile elements (hydrogen, carbon, nitrogen, oxygen, and the noble gases, having condensation temperatures of 180 K or below), are clearly depleted in CI chondrites relative to the Sun. The other important classification of elements is based on what type of phases elements concentrate in: siderophile elements concentrate into iron-nickel metal; chalcophile elements into troilite, the most abundant sulfide in meteorites; and lithophile elements into oxides and silicates. A cosmochemical periodic table of the elements is given in Fig. 1, in which the 50% condensation temperatures for a gas of solar composition at 10⁻⁴ atm total pressure (Lodders, 2003) are given.

The highly volatile elements can be lost during parentbody processes such as shock heating and redistributed dur-



Fig. 1. A cosmochemical periodic table of the elements. Only stable or very long-lived unstable elements (Th and U) are shown. Under each element is given the equilibrium 50% condensation temperature calculated by *Lodders* (2003) for a gas of solar composition at 10^{-4} atm total pressure. The elements are shaded by cosmochemical classification, with siderophile/chalcophile elements indicated by a small triangle at the upper right. Iron is given a half-triangle, because it has lithophile, siderophile, and chalcophile tendencies.

ing parent-body thermal metamorphism (e.g., *Lipschutz and Woolum*, 1988; *Friedrich et al.*, 2003, 2004). It is important not to confuse these effects with nebular fractionations, so data from shocked and/or equilibrated chondrites must be treated with caution.

The primary source of data on elemental abundances in carbonaceous and ordinary chondrites is a series of papers by G. W. Kallemeyn, J. T. Wasson, and coworkers (Kallemeyn and Wasson, 1981, 1982, 1985; Kallemeyn et al., 1989, 1991, 1994, 1996). These workers analyzed relatively large samples, generally two 300-mg replicates, by instrumental neutron activation analysis (sometimes supplemented by radiochemical neutron activation analysis) and reported concentrations of 25-30 elements in each meteorite. More recently, the technique of inductively coupled plasma mass spectrometry has been applied to primitive carbonaceous chondrites, allowing determination of over 50 elements (Friedrich et al., 2002). Elemental abundance patterns in bulk chondritic meteorites are shown in Fig. 2, plotted as a function of volatility [where the 50% condensation temperatures at 10⁻⁴ atm in a gas of solar composition (Lodders, 2003) are used as a measure of volatility]. Data are normalized to CI chondrites and to magnesium to remove the effects of dilution by water and organic matter in CI and CM chondrites. It can be seen that among the carbonaceous chondrite groups, abundances are a smooth function of volatility, with the more volatile elements being more depleted. The carbonaceous chondrite groups, with the exception of CR chondrites, also show some enrichment in refractory elements, whereas ordinary and enstatite chondrites are not enriched in refractory elements (EL chondrites are slightly depleted in refractories). The abundance patterns in carbonaceous chondrites are smooth functions of volatility, regardless of geochemical (lithophile or siderophile) character. In contrast, the patterns in ordinary and enstatite chondrites show effects of metal-silicate fractionation. Among the ordinary chondrites, the volatile lithophile elements lie along straight lines on Fig. 2 that pass through the magnesium point at 1327 K. In contrast, the siderophile elements have enrichment factors of ~1 down to ~800 K, indicating that volatile-element fractionation of lithophile and siderophile elements occurred under different conditions. The EH chondrites are also unusual in that they are enriched in moderately volatile elements and only slightly depleted in highly volatile elements.

Elemental abundances in individual samples of planets and differentiated meteorites are modified by separation of melt and crystalline material during differentiation. In order



Fig. 2. When plotted vs. 50% condensation temperatures (*Lodders*, 2003), the CI-normalized elemental enrichment factors of the volatile elements lie along linear arrays on semilog plots. The data plotted are largely from the compilation of *Wasson and Kallemeyn* (1988), supplemented with more recent data on the groups CM (*Friedrich et al.*, 2002), CK (*Kallemeyn et al.*, 1991), CR (*Kallemeyn et al.*, 1994), CH (*Wasson and Kallemeyn*, 1990; *Bischoff et al.*, 1993), and R (*Schulze et al.*, 1994).

to see through these effects, it is common practice to plot ratios of incompatible elements that are not strongly fractionated from one another during differentiation. Shown in Fig. 3 are CI-normalized Rb/Sr ratios vs. K/U ratios in planets and bulk meteorites. Each of these ratios is of a volatile to a refractory element. It can be seen that in planets and differentiated meteorites, the extent of volatility fractionation greatly exceeds that seen in chondritic meteorites, with the most extremely fractionated body, the angrite parent body, depleted by 3 orders of magnitude in Rb/Sr relative to CI chondrites. All the planets and differentiated meteorite parent bodies are more fractionated than the most extremely fractionated chondrites, the CVs.

Large volatility fractionations are also apparent in iron meteorites. Shown in Fig. 4 are CI-normalized Ge/Ni vs. Ga/Ni ratios in iron and metal from stony-iron meteorites. Gallium and germanium are moderately volatile elements and nickel is slightly more refractory than iron. The partition



Fig. 3. CI-normalized Rb/Sr vs. K/U ratios in bulk meteorites and planets. Data for chondrites are the same as in Fig. 2; data for HED are from *Kitts and Lodders* (1998); data for Mars are from the SNC meteorite compilation of *Lodders* (1998); data for the Moon are from *Anders* (1977a); data for the bulk silicate Earth (BSE) are from *McDonough and Sun* (1995); data for angrites are from *Lugmair and Galer* (1992) and *Mittlefehldt* (2003). The line has a slope of 1 and shows that Earth, the HED meteorites, and angrites have increasingly fractionated Rb/K ratios compared to the bulk solar system.

coefficients for gallium and germanium between solid and liquid metal are close to 1, so fractional crystallization of the cores of iron meteorites does not significantly fractionate Ga/Ni and Ge/Ni ratios. For example, the group IIAB, IIIAB, and IVA irons are each thought to represent fractionally crystallized cores, but it can be seen that each group covers only a narrow range of Ga/Ni and Ge/Ni ratios. These ratios are dominated by volatility fractionation and reach extremes beyond that seen even in angrites, with the group IVB irons having Ge/Ni ratios 4 orders of magnitude below those of CI chondrites. The only group that shows a significant spread is the IAB irons. Wasson and Kallemeyn (2002) reclassified this group and subdivided group IAB into a main group, which shows a relatively small spread in Fig. 4, and a number of subgroups. The subgroups cover a wide range of Ga/Ni and Ge/Ni ratios and it is unclear what their relationship is to the main group IAB irons. Wasson and Kallemeyn (2002) suggest transport of volatile elements during

Fig. 4. CI-normalized Ge/Ni vs. Ga/Ni ratios in iron meteorites and metal of stony iron meteorites. These ratios are mostly controlled by volatility: Fractional crystallization of metal does not significantly fractionate these ratios. Data of *Davis* (1977), *Kracher et al.* (1980), *Malvin et al.* (1984), *Rasmussen et al.* (1984), *Schaudy et al.* (1972), *Scott and Wasson* (1976), *Scott et al.* (1973), *Wasson* (1967, 1969, 1970), *Wasson and Choi* (2003), *Wasson and Kallemeyn* (2002), *Wasson and Kimberlin* (1967), *Wasson and Richardson* (2001), *Wasson and Schaudy* (1971), and *Wasson et al.* (1989, 1998).

impacts on the parent body, as fractional crystallization and partial melting models do not produce such a spread in Ga/ Ni and Ge/Ni.

2.2. Isotopic Fractionation

It is clear from experiments done in vacuum and in hydrogen at plausible nebular pressures (*Davis et al.*, 1990; *Wang et al.*, 1999, 2001; *Grossman et al.*, 2000; *Richter et al.*, 2002), as well as from theoretical considerations (*Grossman et al.*, 2000; *Richter et al.*, 2002; *Richter*, 2004), that high-temperature evaporation can lead to substantial isotopic mass fractionation effects, due to the kinetic isotope effect. It has been clear since detailed isotopic studies began on CAIs in the 1970s that while CAIs can be fractionated in the heavy isotopes of magnesium and silicon by a few permil per amu (*Clayton et al.*, 1988), bulk meteorites of all kinds do not show significant fractionation in these elements. meteorites and planets of Humayun and Clayton (1995). They studied a wide variety of solar system materials, including chondrites, achondrites, Earth, the Moon, and Mars (SNC meteorites), objects with depletions ranging from undepleted CI chondrites to the highly potassium depleted Moon and eucrites. They found no measurable isotopic fractionation effects within $\pm 0.2-0.5\%$ /amu and concluded that condensation, not evaporation, must have been responsible for variable depletion in potassium. Alexander et al. (2000) used an ion microprobe to measure potassium isotopic compositions within chondrules, which can have potassium depletions even larger than those in some achondrites, and found no isotopic fractionation within ±1.5%/amu. Since Yu et al. (2003) showed that evaporative loss of potassium in vacuum and low-pressure hydrogen fractionates potassium isotopes, Alexander et al. (2000) suggested gas-solid exchange during evaporation of chondrules.

Sulfur is the most volatile element for which searches for isotopic fractionation effects have been searched. *Gao and Thiemens* (1993a,b) reported that bulk C, O, and E chondrites have mass fractionation effects of <0.5%/amu. *Tachibana and Huss* (2005) report that troilite in chondrules has sulfur mass fractionation effects of <1%/amu. Although evaporation experiments have not proven that evaporation of sulfur produces isotopic mass fractionation, it is hard to believe that it would not. Thus, sulfur isotopic data in chondrites and chondrules also points to gas-solid exchange during evaporative loss of sulfur.

Searches have been made for isotopic fractionation effects in iron, magnesium, and silicon, but effects are limited to less than 1%/amu in chondrules. Bulk meteorites and planets are perhaps less likely than chondrules to show mass fractionation effects and no effects have been reported. These elements are not as volatile as potassium, but significant losses of all these elements are expected during hightemperature evaporation. Improvements in analytical techniques, particularly the development of multicollector inductively coupled mass spectrometry, have allowed searches for mass fractionation effects at the 0.1%/amu precision level in recent years. Poitrasson et al. (2004) reported that Mars and Vesta are lighter than Earth by 0.03%/amu and the Moon is 0.07%/amu heavier than Earth in iron isotopes. They interpreted the difference between Earth and the Moon as being caused by isotopic fractionation due to 1% iron loss during Moon formation.

2.3. Presolar Grain Abundances

Presolar grains were discovered in carbonaceous chondrites in 1987 (*Lewis et al.*, 1987), barely missing publication in *MESS I*. The abundances of the major types of presolar grains — diamond, silicon carbide, and graphite — in meteorites can be inferred from a fairly simple chemical separation followed by step-heating vacuum pyrolysis for noble gas abundances and isotopic compositions (*Huss and Lewis*, 1994a,b, 1995; *Huss et al.*, 1996). These abundances are quite diagnostic of nebular and parent-body thermal events, since the different kinds of noble gas carriers (presolar grains) are destroyed at different temperatures. A recent extensive survey of carbonaceous chondrites (*Huss et al.*, 2003; *Huss*, 2004) has shown that presolar grain abundances and bulk meteorite elemental abundance patterns are related: After correction for parent-body metamorphic effects, abundance variations among carbonaceous chondrites remain and appear to be related to volatile element depletion patterns.

For the purpose of studying thermal processing in the solar system, it is enough to know that there are several noble gas components with a range of sensitivity to thermal effects. However, it is useful to discuss what these components are and the identity of their carriers. Ne-E is a component highly enriched in ²²Ne relative to ²⁰Ne and ²¹Ne. It comes in two flavors, Ne-E(L), which is nearly pure ²²Ne, and Ne-E(H), which is not quite as strongly enriched in ²²Ne. Ne-E(L) is thought to be the product of decay of 22 Na $(T_{1/2} = 2.6 \text{ yr})$, which is produced in supernova explosions. Ne-E(H) is thought to be material from the helium shell of an asymptotic giant branch star, implanted into grains after the star lost its hydrogen-rich envelope. Xenon has a number of isotopic components that are released at various temperatures from various host phases. Xenon has nine stable isotopes that are produced by a variety of nucleosynthesis mechanisms, which explains in part why so many components have been identified. Perhaps the most famous is Xe-HL, in which the isotope enrichment pattern relative to the solar system is V-shaped, with enrichments in both light and heavy xenon isotopes. This component is associated with presolar diamonds and is released at high temperature (<1000°C), which led to their discovery (Lewis et al., 1987). There are several components containing approximately normal "planetary" isotopic compositions of noble gases (Huss and Lewis, 1994a). Xe-P1 is sited in an unknown carbon-rich carrier "Q" that releases noble gas under fairly mild heating conditions. Xe-P3 and Xe-P6 are released from diamond at fairly low (~500°C) and high (>1000°C) temperatures, respectively. Noble gas components in meteorites are reviewed by Podosek (2003).

Ordered from most susceptible to most resistant to thermal metamorphism, the noble gas components are (1) Ne-E(H) in excess of that in silicon carbide in an unknown carrier; (2) graphite, inferred from Ne-E(L); (3) Xe-P3 in diamond; (4) silicon carbide, inferred from Ne-E(H) and *s*-process Xe; (5) Xe-P1, an easily oxidized, but unknown major carrier of planetary noble gases; (6) diamond, inferred from Xe-HL; and (7) Xe-P6 in diamond. The patterns for these components in several kinds of unequilibrated chondrites are shown in Fig. 5. Relative to CI chondrites and CM chondrite matrices, CR chondrites seem to have experienced simple thermal processing and CM, CO, CV, and CH chondrites contain both highly processed and unprocessed ma-



Fig. 5. Abundances of presolar noble gas carriers in chondritic meteorites. Data of Huss et al. (2003) and Huss (2004).

terials. *Huss et al.* (2003) and *Huss* (2004) suggest that the variations in presolar grain abundances and their correlations with volatile element depletions represent varying degrees of processing of presolar molecular cloud material in the solar nebula.

3. ELEMENTAL AND ISOTOPIC FRACTIONATION DURING EVAPORATION AND CONDENSATION

Isotopic mass fractionation during evaporation and condensation and its relationship to elemental fractionation provides powerful clues about formation conditions. Condensation and evaporation are terms widely used to describe volatility fractionation processes, but they are incomplete. It is important to distinguish between equilibrium and kinetically controlled processes. Condensation is usually used as shorthand for equilibrium fractionation with falling temperature, and evaporation is often taken to mean a kinetically controlled fractionation with increasing temperature, but kinetically controlled condensation and near-equilibrium evaporation can also occur. At this point it is useful to briefly review evaporation and condensation theory. For more extensive treatment of these subjects, see Wood and Hashimoto (1993), Yoneda and Grossman (1995), Petaev and Wood (1998), Ebel and Grossman (2000), Grossman et al. (2000), Nagahara and Ozawa (2000), Ozawa and Nagahara (2001), Richter et al. (2002), Davis and Richter (2003), Richter (2004), and Davis et al. (2005).

3.1. Evaporation Theory

The evaporative flux from a solid or molten surface into a surrounding gas is given by the Hertz-Knudsen equation

$$J_{i,net} = \sum_{j=1}^{n} \frac{n_{ij} (\gamma_{ij}^{evap} P_{ij}^{sat} - \gamma_{ij}^{cond} P_{ij})}{\sqrt{2\pi m_{ij} RT}}$$
(1)

where J_{i.net} is the net flux of element or isotope i (moles per unit area per unit time), n_{ij} is the number of atoms of i in species j, γ_{ij}^{evap} and γ_{ij}^{cond} are the evaporation and condensation coefficients for the jth gas species containing i, P_{ii}^{sat} and P_{ii} are the saturation vapor pressure and the pressure at the evaporating surface of the jth gas species containing the element or isotope i, m_{ii} is the molecular weight of j, R is the gas constant, and T is the absolute temperature. The summation is over all gas species j containing i. It is usually assumed that condensation coefficients are the same as the evaporation coefficients for free evaporation, which must be true at least in the limit $P_{ij} \rightarrow P_{ij}^{sat}$ when equilibrium is approached. Except for the evaporation and condensation coefficients, the various quantities on the righthand side of equation (1) can be calculated given a suitable thermodynamic model for the condensed phase and the surrounding gas. Since there is no formalism for calculating the evaporation coefficients, these must be determined by laboratory experiments.

The gas in equilibrium with liquids of chondritic composition is dominated by species containing single atoms of silicon, magnesium, iron, and potassium (i.e., n = 1, $n_{ij} = 1$ in equation (1)), which allows us to write a simpler version of equation (1) in the form

$$J_{i,net} = \left[\frac{\gamma_i P_i^{sat}}{\sqrt{2\pi m_i RT}}\right] \left(1 - \frac{P_i}{P_i^{sat}}\right)$$
(2)

where $\gamma_i = \gamma_i^{evap} = \gamma_i^{cond}$. The quantity in square brackets is the free evaporation flux and the net evaporation rate is simply the free evaporation rate reduced by a fraction P_i/P_i^{sat} . Equation (2) shows why vacuum evaporation experiments (limit $P_{ij}/P_{ij}^{sat} \rightarrow 0$) are used to determine evaporation coefficients in that the duration of evaporation and the change in composition of the condensed phase gives a measure of J_i , while P_i^{sat} and m_i can be calculated from the thermodynamics of the system, allowing determination of the only remaining unknown, γ_i^{evap} . Equation (2) remains valid for $P_i/P_i^{sat} > 1$, in which case the condensation flux will be greater than the evaporation flux and the net effect will be condensation rather than evaporation.

Equation (2) can also be used to calculate the evaporation rate of isotopes of a given element, and thus the isotopic fractionation of evaporation residues as a function of the amount of the element evaporated. The relative rate of loss by evaporation of two isotopes of i (denoted by subscripts i,1 and i,2) as

$$\frac{J_{i,2}}{J_{i,1}} = \frac{\gamma_{i,2} P_{i,2}^{sat}}{\gamma_{i,1} P_{i,1}^{sat}} \sqrt{\frac{m_{i,1}}{m_{i,2}}} = R_{2,1} \frac{\gamma_{i,2}}{\gamma_{i,1}} \sqrt{\frac{m_{i,1}}{m_{i,2}}}$$
(3)

where $R_{2,1}$ is the atom ratio of isotope 2 to isotope 1 at the evaporating surface. In writing equation (3), it is assumed that isotopes mix ideally and that at the high temperatures of interest here equilibrium isotope fractionations between the condensed phase and the gas are negligible. Given these assumptions, the ratio of the saturation vapor pressures of the isotopically distinct gas species is the same as the isotope ratio in the condensed phase. The standard assumption until recently has been that the evaporation coefficients of isotopes of a given element are the same and therefore that the righthand side of equation (3) can be further simplified to $R_{2,1}\sqrt{m_{i,1}/m_{i,2}}$, but increasingly precise measurements of the isotopic composition of evaporation residues have shown that in all cases involving evaporation from crystalline and molten silicates this simplification is not warranted.

Equation (3) shows that the isotopic composition of the evaporation flux differs from that of the substrate by the factor $(\gamma_{i,2}/\gamma_{i,1})\sqrt{m_{i,1}/m_{i,2}}$, usually called α . If the transport processes in the evaporating material are able to keep it homogeneous and α is independent of the evolving composition of the evaporating residue, then the isotopic composition of the residue will evolve by Rayleigh fractionation given by

$$\mathbf{R}_{2,1}^{\text{res}} = \mathbf{R}_{2,1}^0 f_1^{(\alpha - 1)} \tag{4}$$

where $\mathbb{R}_{2,1}^0$ is the initial isotopic composition of the condensed phase and f_1 is the fraction of isotope 1 remaining in the residue. A good way of testing the validity of the assumption made in writing equation (4) is to plot the experimental data as $\ln(\mathbb{R}_{2,1}^{\text{res}}/\mathbb{R}_{2,1}^0)$ vs. $\ln(f_1)$, which according to equation (4) should fall along a straight line with slope α – 1. Recent experiments with high-precision isotopic analyses are very well behaved in this sense (*Richter*, 2004; *Richter et al.*, 2005).

The relatively simple picture outlined above regarding the isotopic fractionation of evaporation residues becomes a bit more complicated as some of the simplifying assumptions are relaxed. If there is a finite pressure of the evaporating species at the surface of the condensed phase (i.e., $P_i/P_i^{sat} \neq 0$) then the isotopic fractionation factor $\alpha = (\gamma_i)/$ $\gamma_{i,1}$) $\sqrt{m_{i,1}/m_{i,2}}$ in equation (4) should for most practical applications be replaced by $\alpha' = 1 + (\alpha - 1)(1 - P_i/P_i^{sat})$ (*Rich*ter et al., 2002). The isotopic fractionation of a residue for a given amount of evaporation of the parent element will be reduced when the system becomes diffusion-limited to the extent that diffusion is not sufficiently fast to maintain the homogeneity of the condensed phase. Diffusion limitations are insignificant in the case of molten CAIs or chondrules (Richter, 2004), but evaporation of solids such as forsterite will be severely diffusion-limited with isotopic fractionations limited to a very thin boundary layer at the surface. Wang et al. (1999) studied the effect of diffusion on the distribution of isotopic fractionation effects within forsterite. Theoretical studies of diffusion effects as a function of forsterite grain size have been done in considerable detail (Tsuchiyama et al., 1999; Nagahara and Ozawa, 2000). Diffusion limitation of bulk isotopic fractionation can be mitigated to some extent by isotopic mass fractionation during diffusion (Richter, 2004; Richter et al., 1999, 2003).

3.2. Condensation Theory

"Condensation calculations," in which the thermodynamic equilibrium condensate assemblage is calculated as a function of temperature in a gas of fixed pressure and composition (usually solar), provide an extremely useful framework for interpreting bulk compositions of CAIs, chondrules, meteorites, and planets. The first modern calculations for the solar system were done by Grossman (1972); a comparison of the condensation sequence at 10⁻³ bar in a gas of solar composition from various workers is given in Petaev and Wood (2005). More recent calculations have explored a wider range of parameters. Wood and Hashimoto (1993) explored the effects of varying the proportions of four volatility components of solar material: refractory dust, carbonaceous matter, ice, and hydrogen gas. They found that dust enrichment (1) increases the condensation temperatures of all minerals at fixed pressure, (2) increases the iron content of mafic minerals, and (3) permits melts to be in equilibrium with gas. Yoneda and Grossman (1995) explored condensation of melts in the CaO-MgO-Al₂O₃-SiO₂ (CMAS) system using a reasonably accurate thermodynamic model for melts for the first time and found that melts can coexist with gas at pressures as low as 10^{-2} atm. *Ebel and Grossman* (2000) extended such calculations to chondritic compositions. Dust-enriched systems have two important differences from solar system composition: condensation temperatures are higher, so melts can be stabilized; and conditions can become oxidizing enough at high dust enrichments to allow condensation of a significant fraction of total iron into ferromagnesian minerals rather than metal (*Wood and Hashimoto*, 1993; *Ebel and Grossman*, 2000). *Fedkin and Grossman* (2006) explored the conditions needed to explain the high fayalite content of chondrule precursors and pointed out the difficulty in achieving sufficiently oxidizing conditions in solar nebular models.

Equilibrium condensation calculations have been applied to trace elements as well. Whereas high-quality thermodynamic data are available for major phases in condensation calculations described above, most trace elements condense in solid solution in one of the major phases. However, for most trace elements, thermodynamic data are only available for pure elements and simple compounds. Sometimes good solid solution models exist for minor and trace elements, but in many cases assumptions must be made about host phases and solution behavior, guided by microbeam trace-element analyses of individual phases in meteorites indicating which minerals concentrate different trace elements. The most up-to-date table of 50% condensation temperatures of all the elements is that of *Lodders* (2003), which has been adopted for use in Figs. 1 and 2.

Rapid, kinetically controlled high-temperature condensation can lead to isotopic fractionation (*Richter*, 2004). In this case, $P_i/P_i^{sat} > 1$ in equation (2) and isotopic fractionations can be predicted as outlined above. The only case for which isotopic fractionation during condensation can be made is for fine-grained group II calcium-, aluminum-rich inclusions (CAIs), which have REE patterns clearly indicating condensation (*Boynton*, 1975; *Davis and Grossman*, 1979) and which contain isotopically light magnesium and silicon (*Clayton et al.*, 1988). Equilibrium isotopic fractionation is minimal at high temperature, and condensation in most plausible nebular environments is expected to produce elemental fractionation without isotopic fractionation.

4. VOLATILITY FRACTIONATION MODELS

Palme et al. (1988) divided models to explain moderately volatile element abundance patterns into two categories, evaporation and condensation models, and discussed their merits in detail. A third class of model, in which volatile element abundances are inherited from the molecular cloud parental to the solar system, has been advocated more recently.

Anders (1964) and Larimer and Anders (1967) proposed that volatile element fractionation occurred by evaporation during chondrule formation and that the variations in volatile element abundances in bulk meteorite parent bodies and planets were simply due to variations in the mixing ratio of volatile-depleted chondrules to undepleted matrix. There was a vigorous debate (Wasson and Chou, 1974; Anders, 1975; Wai and Wasson, 1977; Anders, 1977b; Wasson, 1977) between E. Anders and J. T. Wasson over the issue of whether moderately volatile element abundances were a continuous function of volatility or "a terraced landscape where gently sloping plateaus alternate with steep declines" (Anders, 1977b). The latter description was what was predicted from volatile element loss from chondrules during melting, and Anders argued that Wasson's continuous curve was based in part on poorly known condensation temperatures or poorly determined elemental abundances. Current data (Fig. 2) with a quarter century of improvements in analytical techniques and thermodynamic equilibrium calculations favor a continuous function, especially for carbonaceous chondrites, although ordinary chondrites continue to show some scatter. Shu et al. (1996) proposed a new location for chondrule formation, close to the Sun, at the X point, where infalling material divides into matter that falls into the Sun and that which is thrown out, either out of the solar system or back out to the solar nebula. This region provides a qualitative explanation for short-lived thermal events needed to explain chondrule formation and would permit mixing of volatile-depleted chondrules with unprocessed material further out in the solar nebula. Connolly and Love (1998) and Desch and Connolly (2002) have proposed that chondrules are heated by the passage of nebular shock waves, and that the complementary volatile element patterns of chondrules and matrix arise from this process. Alexander et al. (2001) also favor a two-component explanation of volatile element abundances, with kinetically controlled volatility fractionation accompanying chondrule formation (shockwave heating is one way to do this). They postulated some equilibration with nebular gas on cooling to explain the lack of isotopic mass fractionation effects in potassium and iron in chondrules and partial loss of nebular gas to explain the relatively smooth relationship between CI-normalized abundances and condensation temperature. In summary, the lack of isotopic fractionation effects in the moderately volatile elements iron and potassium in chondrules argues against significant volatile loss during melting, at least in the simple picture of Anders, so current models of this sort appeal to back-reaction to reduce mass fractionation effects, chondrule formation in restricted regions of the solar nebula in order not to lose presolar grains from material now in carbonaceous chondrite matrix, and partial loss of nebular gas in chondrule-forming regions to produce smooth fractionation patterns (Alexander et al., 2001; Alexander, 2004).

Wasson and Chou (1974) and *Wai and Wasson* (1977) proposed that the abundances of moderately volatile elements in ordinary chondrites arose by equilibrium condensation with continuous loss of nebular gas. They argued that the abundance pattern was a smooth function of condensation temperature and that complete volatile loss during chondrule formation was unlikely because of the high cooling

rates inferred for chondrules. Although they calculated condensation temperatures, they did not present a quantitative model of exactly how the process worked. Cassen (1996, 2001) presented such a model and found that the patterns in CO and CV chondrites could be reproduced by simple disk models, but had difficulty matching CM chondrites. These models envision a hot solar nebula, with significant evaporation out to 3 AU and a high mass accretion rate, >10⁻⁷ M_{\odot} yr⁻¹, with chondrites achieving their volatility fractionation patterns in roughly their final formation locations. Wulf et al. (1995) evaporated Allende under reducing conditions in a gas-mixing furnace and showed that volatile element patterns in residues did not match those in bulk CAIs. They concluded that condensation, not evaporation, must be the controlling process in the solar nebula. The major problem with models envisioning a hot solar nebula are the presence of presolar grains in a wide variety of chondritic meteorites with minimal parent-body metamorphism and the presence of isotopic differences in titanium (Niemeyer and Lugmair, 1984; Niederer et al., 1985), chromium (Lugmair and Shukolyukov, 1998), and molybdenum (Dauphas et al., 2002; Yin et al., 2002) among different bulk meteorites.

Huss et al. (2003) and Huss (2004) noted relationships between volatile-element abundances and presolar grain abundances and suggested that initially cold interstellar cloud material was heated in the solar nebula to varying degrees. They pointed out that volatile element enrichment patterns (e.g., Fig. 2) are generally flat at high temperatures, and that elements below a certain temperature are progressively depleted relative to CI chondrites. The temperature at which this break occurs differs for different meteorite groups and appears to be correlated with the abundance patterns of presolar grains. The sequence is from CI to LL to CO to CR to CV. Since presolar grains are in the matrix and the elemental abundance patterns are controlled by high-temperature components (chondrules, metal, and CAIs), Huss argues that these patterns must have been established prior to chondrule-forming events and probably represent a process common to all chondritic parent material. The presence of presolar grains in a wide variety of chondritic meteorites makes it clear that some fraction of material in the solar nebula escaped vaporization and subsequent condensation, because such conditions would have destroyed presolar grains. Huss proposes that a fraction of material from the solar system's parental molecular cloud was partially evaporated and the evolved gas lost, producing high-temperature materials and volatile element depletion patterns in bulk meteorites. Some of the dust was thermally processed, but did not go through chondrule formation, producing the variety of presolar grain abundance patterns. Huss points out unsolved problems, such as the fact that the temperatures of thermal processing of matrix dust (<~200°C for CI and LL chondrites, <700°C for CO, ~700°C for CR, and $>700^{\circ}$ C for CV) are much lower than the temperatures needed to produce volatile element enrichment patterns. It is also not at all clear why the patterns in low-temperature and high-temperature components should be correlated.

Spectroscopic studies of absorption lines show that the gas phase of the interstellar medium (ISM) is depleted in refractory elements, with the more refractory elements more depleted (Palme and Jones, 2003). Dust in the ISM presumably has a complementary pattern and is enriched in refractory elements. Since this pattern is qualitatively similar to moderately volatile element depletion patterns in chondrites, Palme (2001) and Palme and Jones (2003) raised the question of whether the pattern in chondrites was inherited from the ISM. They dismiss this possibility on two grounds. (1) Interstellar medium dust must represent many individual stars with a wide variety of isotopic compositions (as is seen in presolar grains), yet the solar system is remarkably isotopically uniform. However, this may not be a strong argument. Presolar grains are circumstellar condensate grains that retain the isotopic signature (generally very different from solar system composition) characteristic of their parent star. But it may be that most dust is reprocessed in the ISM, where shocks are expected to efficiently sputter away circumstellar grains. Modelers of the ISM have great difficulty understanding how the presolar grains found in meteorites survived. Furthermore, the recent discovery of "abundant" presolar silicate in meteorites (Nguyen and Zinner, 2004; Nagashima et al., 2004) only corresponds to ~25 ppm of primitive meteorites. (2) In the ISM, most of the strontium is in the dust and most of the rubidium is in the gas. Palme (2001) argues that over hundreds of millions of years this should produce a significant variety in 87Sr/86Sr ratios, yet the initial solar system strontium ratio is relatively uniform.

For the past several years, Yin (2002, 2004) has advocated the idea that the solar nebula inherited its volatileelement abundance pattern from the ISM. In a recent paper, Yin (2005) goes into more depth, pointing out the complementarity of carbonaceous chondrite and ISM gas elemental abundance patterns. He points out that in dense molecular cloud material, refractory elements are in dusty cores and volatiles (apart from hydrogen and helium) are in icy mantles. He proposes that in the solar nebula, evaporation of icy mantles and loss of gases allowed solar system materials to inherit the elemental abundance pattern of the dust. He counters Palme's strontium-isotope argument by proposing that the recycling in the long-lived diffuse ISM homogenizes strontium isotopes and that the molecular cloud lived for only a relatively short time, so large heterogeneities could not develop. The ISM inheritance model surely needs more development, as many problems are left unsolved. No explanation is offered for metal-silicate fractionation, for which meteoritic evidence is quite clear. It is also not clear whether ISM fractionations are sufficiently large to explain some meteoritic patterns. For example, Ge/Ni ratios among iron meteorites vary by more than 4 orders of magnitude, but the ISM Ge/Ni ratio only varies by a factor of 100.

In summary, it is clear from the presence of presolar grains in meteorites and isotopic variations in titanium, chromium, and molybdenum among bulk meteorites that the inner solar system cannot have been completely vaporized and homogenized. It is also clear that chondrules and CAIs were exposed to high temperatures, at least for brief periods of time. What is still not clear is whether volatile element depletions in solar system objects arose from transient high-temperature events somewhere in the solar system or were inherited from the interstellar medium.

5. CONCLUSIONS

Moderately volatile elements are depleted in meteorite parent bodies relative to bulk solar system composition. The depletions are smooth functions of condensation temperature when plotted on semilog plots. The lack of isotopic massfractionation effects in planets and bulk meteorites indicates that the elemental fractionation process, be it condensation or evaporation, whether it occurred in the solar nebula or somewhere else, occurred under near-equilibrium conditions, but further tests are needed. The survival of presolar grains in primitive meteorites indicates that some material was never vaporized and recondensed in the solar system.

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