## THE ORIGIN AND EVOLUTION OF THE TERRESTRIAL ALKALI ELEMENT BUDGET

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### ABSTRACT

The origin and evolution of the terrestrial alkali element inventory is investigated in the framework of the accretion and differentiation history of the Earth. We predict that a significant percentage of the Earth's bulk alkali element inventory is in the core (30% for Na, 52% for K, 74% for Rb, and 92% for Cs). These predictions agree with independent estimates from nebular volatility trends and (for K) from terrestrial heat flow data. Vaporization and thermal escape during planetary accretion are unlikely to produce the observed alkali element depletion pattern. However, loss during the putative giant impact which formed the Moon cannot be ruled out.

#### INTRODUCTION

The depletion of Na, K, Rb, and Cs in the Earth's upper mantle and crust relative to their abundances in primitive, undifferentiated meteorites (chondrites) is a long standing problem in geochemistry<sup>1,2</sup>. The cosmochemical classification of the elements based on condensation temperatures in the solar nebula divides the alkali elements into moderately volatile (Na) and volatile (K, Rb, Cs) elements with increasing volatility from Na to Cs, while elements such as Al, Sr, and U are classified as refractory elements<sup>3</sup>. Elemental abundance ratios of moderately volatile and volatile elements to refractory elements (e.g., Na/Al, K/U, Rb/Sr) in chondrites reflect volatility related fractionations if these ratios are smaller than those found in CI-chondrites (which have primordial, solar composition except for the atmophile elements H, O, C, N and noble gases). Geochemically, the alkali elements are normally regarded as lithophile elements and therefore it is commonly assumed that the observed alkali element abundances in the Earth's mantle and crust represent the entire alkali element budget of the Earth. As can be seen from Table I, the Na/Al, K/U, and Rb/Sr ratios of the bulk silicate Earth are generally much lower than the respective chondritic ratios, which indicates that the silicate Earth underwent a massive depletion of alkali elements.

One requirement for modeling the alkali depletion process is that this process should integrate into the framework of accretion and differentiation of the Earth. In the next sections we will argue that two processes are responsible for producing the observed depletions, namely, core formation and vaporization during the hypothesized giant impact which formed the moon.

### ACCRETION AND DIFFERENTIATION OF THE EARTH

We adopt a heterogeneous accretion scenario for the Earth that is similar to models proposed by Wänke<sup>4</sup> and Ringwood<sup>5</sup>. An important feature of these models is the absence of core-mantle equilibrium. The model used here and the models of Wänke and Ringwood involve two stages: (a) initial accretion of highly reduced matter and core formation; and (b) accretion of more oxidized matter and a moon-forming impact to the Earth after core formation.

The underlying concept in this study is that chondritic matter is representative of the material which formed larger planetary objects. This is the difference between our model and the models of Wänke and Ringwood, who postulate that the initially accreting component is devoid of elements more volatile than Na. As illustrated in Table 1, none of the known chondrite groups match the hypothetical component A of Wänke and Ringwood. We discuss further below why we use EH-chondrites as the initially accreting component.

	Table I.	Some Compo	ositional Data i	for Chondrit	es, the Au	brite Paren	t Body (Al	PB) and the	Earth and N	Moon.
Туре	Total Fe wt%	Silicates Fe/(Fe+Mg) mole %	Fraction Fe as metal Fe <sub>met</sub> /Fe <sub>tot</sub>	metal & sulfide, wt%	δ <sup>17</sup> O <sup>a</sup> 0/ <sub>00</sub>	δ <sup>18</sup> O <sup>a</sup> 0/ <sub>00</sub>	Na/Al	K/U	Rb/Sr	Cs/Rb
Carbo	naceous C	Chondrites								
CI	19.04	45	0	3	~ 8.8	~ 16.4	0.576	68300	0.295	0.081
CM	21.00	43	0	8.7	~ 4	~ 12.2	0.347	36400	0.168	0.074
CO	24.80	33	0-0.2	~ 10.2	~ -5.1	~ -1.1	0.287	26500	0.114	0.055
CV	23.50	35	0-0.3	14.4	~ -4	~ 0	0.189	18200	0.082	0.076
Ordina	ary Chond	rites								
LL	18.50	27	0.11	9.4	3.9	4.9	0.588	60800	0.279	0.058
L	21.50	22	0.29	13.3	3.5	4.6	0.573	63500	0.279	0.090
Н	27.50	17	0.58	23.2	2.7	4.2	0.566	65000	0.290	0.041
Enstat	ite Chondi	rites								
EL	22.00	0.26	0.74	28.7	2.9	5.6	0.552	73500	0.305	0.040
EH	30.62	0.98	0.62	36.6	2.7	5.2	0.860	88900	0.439	0.068
Differe	entiated B	odies								
APB	(30.62)	0.072-0.72	0.68	38.2	2.78	5.34	$0.736^{\circ}$	68700°	0.257°	$0.053^{\circ}$
Earth	32.04	11	0.82	32.5 <sup>b</sup>	2.78	5.38	0.138°	10500°	0.028 - 0.035°	0.022 - 0.036°
Moon	10.60	19	~ 0	2.5	2.78	5.38	0.019	2500	0.009	0.045

(a) bulk isotopic composition relative to SMOW (standard mean ocean water); (b) Earth's core also contains oxygen; (c) element ratio from silicate portion only. Data sources: [6-12, and references therein].

A generally accepted and likely accretion scenario invokes condensation of chondritic matter from the nebula gas and accumulation of solids into larger planetesimals. These planetesimals accrete further to form meteorite parent bodies and also the terrestrial planets. While the primary compositional signature from condensation processes is relatively well preserved in chondrite parent bodies (al-

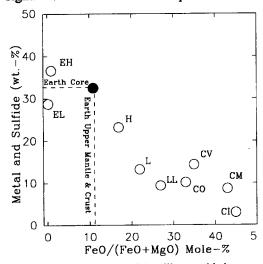


Figure 1. Comparison of the silicate oxidation state and metal-sulfide content in chondrites with corresponding data for the Earth. The terrestrial core size is easily explained if accretion started from EH-chondritic matter.

though chondrule formation and thermal metamorphism have partially erased the nebular signature), metal-silicate fractionation during core-mantle differentiation in the terrestrial planets efficiently sorted the initially more homogeneously distributed metal, sulfide, and silicate phases so that siderophile and chalcophile elements fractionated from the lithophile elements. These fractionation processes strongly depend on the oxidation state of the differentiating parent body.

We now address the question of which type of chondritic planetesimals accreted to form the Earth. From Table I we can see that chondrites display a wide range in oxidation state, as indicated by their molar FeO/(FeO+MgO) ratios in silicates, and their total metal and sulfide content. This is also shown in Figure 1. The oxidation state of terrestrial silicates falls between that of ordinary (LL, L, H) chondrites and enstatite (EH and EL) chondrites. The chondrites roughly plot along a line and the corresponding data for the Earth fall right into this trend. This plot also may indicate that there is

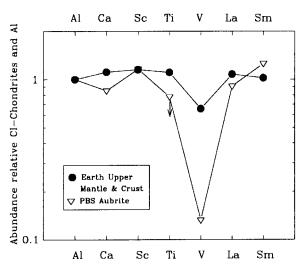


Figure 2. Refractory elemental abundances in the Earth's show that about 2/3 of the Earth accreted from upper mantle and in the Pena Blanca Spring aubrite<sup>11,13</sup>. The highly reduced matter and about 1/3 oxidized depletion of V is explained by partitioning into metal-sulfide matter.

There are additional indications that the

indeed a relationship between chondritic compositions and bulk composition of the Earth. If the Earth formed from chondritic matter, it is necessary to supply enough metal and sulfides to account for the terrestrial core size of about one third of the Earth's mass. To fulfill this requirement, we have to turn to EH-chondritic matter, which is the only type of chondritic matter which can supply enough metal and sulfide. However, the current oxidation state of the Earth's silicates is higher than that of enstatite chondrites and thus at least two components, a reduced and an oxidized one, are necessary. This heterogeneous accretion has been discussed in the models by Wänke<sup>4</sup> and Ringwood<sup>5</sup>, which show that about 2/3 of the Earth accreted from

Earth's accretion started from highly reduced enstatite chondritic matter. One is the (well known) common oxygen isotopic signature of enstatite chondrites and the Earth-Moon system (Table 1). No other chondrite group plots along the terrestrial O-isotope fractionation line<sup>10</sup>. The only other meteorites matching the isotopic signature are the differentiated enstatite-rich meteorites known as aubrites, for which a link to enstatite chondrites is established (see below).

Another connection of terrestrial accretion to enstatite meteorites comes from the depletion of refractory V in the bulk silicate Earth<sup>14,15</sup>. Figure 2 shows the abundance of refractory elements in the Earth's upper mantle and in the Pena Blanca Spring aubrite. The depletion of V in both systems is obvious, although the depletion in the Earth's mantle is smaller than that in aubrites. Under the current oxidation state of the Earth's mantle V behaves as a lithophile element. However, in a highly reduced environment V becomes chalcophile and siderophile<sup>16</sup> and can be removed from silicates by a metal-sulfide melt. This partitioning explains the depletion of V in aubrites<sup>13</sup> and it is plausible to assume that as long as the Earth accreted reduced matter, removal of V into the core also occurred.

After about 2/3 of the Earth had accreted from reduced matter, more oxidized planetesimals accreted to the Earth and oxidized the mantle to its current state<sup>4,5</sup>. If the moon were formed by the impact of a Mars-sized body with the Earth and subsequent recondensation of evaporated mantle and impactor material<sup>17</sup>, then this impact most likely occurred during or after accretion of the oxidized component to the Earth. The reason for postulating this late impact (after core formation) is due to the fact that the moon lacks a large core, implying that no metal was available in the recondensing matter. However, while it is relatively easy to identify the reduced chondritic component which initially accreted to the Earth, the task of finding the oxidized component among chondrites is more difficult. For modeling purposes, it is easiest to assume CI-composition, as done by Wänke and Ringwood in their models<sup>4,5,14,15</sup>, but in that case, the requirement that the oxygen isotopic signature of the Earth-Moon system is preserved is violated. In addition, the elemental contribution of the Mars-sized impactor is certainly not negligible, but no attempts have yet been made to estimate the composition of this supposedly differentiated object. However, we will see below that despite these obstacles, we can model the evolution of the terrestrial alkali element budget by using the accretion scenario discussed so far.

# ALKALI ELEMENT REMOVAL INTO THE EARTH'S CORE

As discussed above, the early Earth probably was highly reduced. From the mineralogy and chemistry of highly reduced meteorites (the enstatite chondrites and achondrites) we can see that we have to modify our views about the commonly accepted geochemical behavior of the elements. For example,

the high degree of reduction in enstatite meteorites resulted in the partial occurrence of Si metal alloyed with FeNi, and led to the formation of exotic sulfides such as oldhamite (CaS), niningerite ((Mg,Fe)S), alabandite ((Mn,Fe)S and Ti-bearing troilite<sup>18</sup>. Two minerals of special interest in this study are djerfisherite ( $K_3(Cu,Na)(Fe,Ni)_{12}(S,Cl)_{14}$ ) and caswellsilverite (NaCrS<sub>2</sub>), which contain about 10 wt% K and 16 wt% Na respectively. Their occurrence in enstatite meteorites underlines the chalcophile nature of the alkali elements under reducing conditions. As a consequence, some fraction of the alkali elements may have partitioned as sulfides into the Earth's core during its early differentiation.

Alkali element partitioning into the Earth's core was modeled by assuming that alkali element partitioning during core formation on the aubrite parent body (APB) is analogous to that on the early Earth<sup>12</sup>. Aubrites (enstatite achondrites) represent samples of the silicate portion of the highly reduced, differentiated aubrite parent body. These brecciated meteorites consist mainly of almost iron free pyroxene (enstatite)<sup>19</sup>. Other major minerals are forsterite, diopside and albite. It has been shown earlier<sup>13</sup> that the bulk APB is most likely EH-chondritic in composition and that the silicate portion of the APB (which is sampled by the aubrites) formed as a result of core-mantle differentiation under very reducing conditions. If, as seems likely<sup>12,13</sup>, the APB and the early Earth differentiated from EH-chondritic matter, then the APB provides a natural laboratory to study how core-mantle differentiation proceeds under highly reducing conditions. Because we can determine the compositions of the primitive EH-chondrites and the differentiated aubrites by measuring samples available from our meteorite collections, we can estimate the composition of the core formed by differentiation of EH-chondritic matter.

The calculated concentrations of the alkali elements in the core of the APB are obtained from the general mass-balance equation:

$$C(EH) = X(silicates) \cdot C(silicates) + X(core) \cdot C(core)$$
 (1)

where C stands for concentration by weight and X for the mass fraction of silicates (= 0.618) and core (0.382) in the APB. Details of these calculations are described elsewhere<sup>12</sup> and are only briefly repeated here. The composition of the entire APB is assumed to be EH-chondritic (Table II, column 1) and the composition of the silicate portion of the APB is computed from element correlations for analyses of aubrites (column 2). The calculated elemental abundances in the core of the aubrite parent body are listed in column 3. These results show that the APB core contains 2590 ppm Na, 550 ppm K, 3.4 ppm Rb, and 0.31 ppm Cs. Using equation (1) and the compositions of caswellsilverite and djer-fisherite in column 4, we can also calculate how much caswellsilverite and djer-fisherite are required on the entire APB to remove the alkali elements as sulfides into the core. For Na, about 0.63 wt% caswellsilverite is necessary and for K and Rb we need about 0.22 wt% djerfisherite.

	EH-Chondrites (= bulk APB)	APB silicates ( = aubrites) <sup>b</sup>	APB Core	Djerfisherite (D) & Caswellsilverite (C)	
	(1)	(2)	(3)	(4)	
Al	8200±200	13300±500	0	0	
Na	7050±60	9800±500	2590±1000	15.7±1 wt% (C)	
K	850±60	1030±120	550±260	9.5±1 wt% (D)	
Rb	3.1±0.4	2.9±1.1	3.4±2.1	660 (D)	
Cs	0.21±0.06	0.15±0.10	0.31±0.24	?	

Table II. Alkali Element Mass-Balance in the Aubrite Parent Body and Alkali Element Concentration in Sulfide Minerals.

<sup>(</sup>a) data in ppm, if not noted otherwise, for data sources see [12]; (b) concentration of APB silicate from element correlations of aubrite samples; (c) core composition calculated from data in column (1) and (2) and equation (1).

The amounts of each mineral required are fairly low and agree with the trace abundances of these minerals that are observed in enstatite meteorites<sup>20,21</sup>. In addition to the low abundances of djerfisherite and caswellsilverite, the appearance of these minerals as solidified relics of sulfide melts in aubrites makes it plausible to assume that they are indeed alkali element carriers into the core.

If we assume that elemental fractionations during core formation on the APB and the early Earth are similar, then the Earth's core could contain the same alkali element abundances as the core of the APB. Indeed the concentration of alkali elements in the Earth's core calculated here agree with independent published estimates based on nebular volatility trends<sup>11</sup> (see also discussion below). One could argue that the analogy between the APB and early Earth is only valid as long as the Earth is comparable in size to asteroidal objects like the APB, because higher pressures and temperatures prevailing in the growing Earth may have altered the partitioning behavior of alkali elements between sulfides and silicates. However, Stevenson<sup>22</sup> presented arguments that metal-sulfide/silicate equilibration most likely occurred in the top layers of a differentiating body and that the dispersed metal-sulfide bodies from an incoming undifferentiated object equilibrated with the surrounding silicates until the metal-sulfides bodies accumulated into larger blobs which sank to the core. Because the pressure regime in the upper region of a larger planet is probably not very different than that in asteroidal sized bodies, we can expect that elemental partitioning between metal, sulfide, and silicates under comparable redox conditions and temperatures will lead to similar results.

Even if we assume that metal-sulfide blobs equilibrated with surrounding silicates in deeper portions of the Earth, we expect that the partitioning of the alkali elements into sulfides still took place. The terrestrial occurrence of djerfisherite in diamond<sup>23</sup> shows that djerfisherite is stable at higher pressure in deeper regions of the mantle. In addition, sulfide/silicate partition experiments at 4-6 GPa and 1525-2585°C show that K partitions more strongly into sulfides at high pressures than at lower pressures<sup>24</sup>. However, it is important to keep in mind that alkali removal into the core will only occur as long as highly reducing conditions are present during accretion and core formation. Once accretion proceeds with more oxidized matter, the redox state of the terrestrial silicates will not allow the continued formation of djerfisherite and caswellsilverite, and the alkali elements will remain lithophile.

At this point we want to mention that enstatite meteorites also contain oldhamite (CaS) which is the main Ca and REE carrier in these meteorites. It may appear from Figure 2 that some Ca was lost from the Pena Blanca Spring sample, indicating possible removal of CaS into the core. However, this depletion is an artifact from the normalization to Al, which is about 10% lower in the Pena Blanca Spring sample than in CI chondrites. Presently, no definitive conclusion can be drawn regarding possible Ca depletions in the silicate portion of the APB and possible CaS loss to the APB core. Comparison of the Ca abundances in the silicate portion of the APB and enstatite chondrites is hampered by the inhomogeneous distribution of CaS in aubrites and the lack of any element correlation of Ca with other major elements. Thus, no estimate of the total Ca abundances in the APB silicates can be made. It is also not very likely that CaS is as efficiently removed into the core as are the alkali-bearing sulfides because of its high melting point of ~2720 K and its low density ( $\rho$ =2.5 g/cm<sup>3</sup>), which is lower than that of silicates. Formation of CaS solid-solutions with FeS, (which is likely to have occurred for the alkali element bearing sulfides and FeS) probably did not occur because the denser Fe/FeS melts (ρ> 4.5 g/cm<sup>3</sup>) are removed at lower temperatures, before a possible FeS-CaS eutectic can form. The Fe-FeS eutectic is at ~1270 K and the FeS-CaS eutectic is at 1393 K (at a composition of 80 wt.-% FeS and 20 wt.-% CaS<sup>25</sup>). Given the fact that CaS is about 5 times less abundant than FeS in EH chondrites, the temperature where a solid solution between CaS and FeS would occur are even higher. Therefore, liquid Fe-FeS melts would migrate towards the core before CaS can be incorporated into FeS, leaving CaS behind in the silicate.

# ALKALI ELEMENT REMOVAL BY THE MOON-FORMING IMPACT

After the reducing stage of accretion, the bulk silicate Earth would be similar in composition to aubrites, if the analogy between the early Earth and the APB is made. As can be seen from Table I, the elemental ratios (Na/Al, K/U, Rb/Sr) in the silicate portion of the APB (the aubrites) are significantly

higher than those presently observed in terrestrial silicates. Because continuing accretion of a more oxidized component probably added even more alkali elements to the silicate portion of the Earth, some mechanism is required to reduce the alkali element abundances to their current state.

Vaporization is one possible mechanism to remove alkali elements from the early Earth. However, evaporation from a magma and thermal escape from the Earth during accretion and differentiation is unlikely for elements as heavy as the alkalis<sup>26</sup>. To loose alkalis, temperatures must first rise high enough to evaporate alkali elements and their compounds. Second, the gaseous vapors must leave the gravitational field of the (growing) Earth. Vaporization of the alkalis can be calculated using the MAGMA code<sup>27</sup>, while thermal (Jeans) escape is governed by the ratio of the thermal velocity ( $v_{th}$ ) of the gaseous species to the planetary escape velocity ( $v_{tsc}$ ). Two cases ( $v_{tsc}/v_{th} = 1$  and 5) were considered at the Fe-FeS eutectic temperature of 1270 K where planetary differentiation begins<sup>26</sup>. The results show that alkali loss is negligible from Mars sized and larger bodies under these conditions. Thus, thermal escape of the alkali elements from the Earth after core formation can be ruled out.

However, another efficient process involving vaporization which could have removed volatile elements from the Earth may have been the putative Moon-forming impact. If there were no loss of volatiles during the impact, one would expect that lunar Na/Al and K/U ratios would be similar to those in the terrestrial silicates, because the common oxygen isotope systematics suggest that the terrestrial and lunar silicates have a common parent. However, the lunar volatile to refractory element ratios are significantly lower than those found in the Earth's silicates (Table I) indicating that alkali elements were lost from the Earth-Moon system. Details of this volatile loss mechanism have not yet been modeled and require further investigation.

### ALKALI ELEMENT BUDGET IN THE EARTH

We derive the alkali element abundances for the bulk Earth (core + mantle + crust) by combining our estimated abundances in the Earth's core with published estimates of alkali element abundances in the silicate portion of the Earth<sup>11</sup>. Terrestrial silicate abundances were derived from upper mantle and crust data<sup>11</sup>. We follow the generally made assumption that the abundances determined for the upper mantle and crust are representative for the entire silicate portion of the Earth and that no differences for alkali element abundances are found in the lower mantle.

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	Earth' Mantle & Crust 11	Core (this work)	Core11	Bulk Earth (this work)	Bulk Earth <sup>11</sup>	CI- Chondrites <sup>6</sup>	Ordinary Chondrites <sup>7</sup>	EH- Chondrites <sup>12</sup>	
Na	2932 (2330-3640)	2590 (± 1000)	1400 ?	2820	2450	5000	6400-7000	7050±610	
K	232 (175-300)	550 (±260)	210 ?	340	225	558	780-825	850 <del>±6</del> 0	
Rb	0.6 (0.48-0.73)	3.4 (±2.1)	1.1 ?	1.5	0.76	2.3	2.9-3.1	3.1±0.4	
Cs	0.013 (0.007-0.025	0.31 (±0.24)	0.14?	0.11	0.055	0.187	0.12-0.18	0.21±0.6	

Table III. Alkali Element Abundances in the Earth and in Chondrites (ppm).

Bulk Earth abundances calculated from equation (1) and assuming a core mass fraction of 0.325 for the Earth.

The data for the silicate portion, core, and the bulk Earth are given in Table III. Alkali element abundances for the bulk Earth of 2800 ppm Na, 340 ppm K, 1.5 ppm Rb, and 110 ppb Cs are obtained using the mass balance equation (1). A large fraction of the terrestrial alkali elements is hidden in the core (30% Na, 52% K, 74% Rb, and 92 % Cs). However, the calculated bulk Earth abundances of the alkalis are only 40% (Na), 43% (K), 50% (Rb) and 80% (Cs) of those found in ordinary chondrites suggesting that the process which established the alkali abundances in the terrestrial silicates was dominated by vaporization.

Our derived alkali abundance estimates for the core are consistent with independent results obtained by Kargel and Lewis<sup>11</sup>. These authors calculated the volatility trend for the Earth and compared their predictions with observed abundances in the silicate Earth. The difference between calculated bulk Earth abundances and the silicate portion was assigned to the core. Their core data are also shown in Table III and we see that their data are about 1.5-3x smaller than the core abundances determined here. However, within the uncertainties of the data both studies are consistent.

We can also compare our calculated K abundance with K abundance estimates from heat-flow data. Recently Breuer and Spohn<sup>28</sup> calculated that the present day heat flow can be matched if about 20 ppb U are in the mantle and 400-800 ppm K are in the core. Their estimated K abundance agrees well with the 550±250 ppm K derived in this study.

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