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TITLE: # IRIDIUM AND OTHER TRACE METAL ENRICHMENTS FROM HAWAIIAN VOLCANOES

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Iridium and Other Trace Metal Enrichments from Hawaiian Volcanoes

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ABSTRACT

Particle and gas samples were collected at Mauna Loa volcano during and after its eruption in March and April, 1984 and at Kilauea volcano in 1983, 1984, and 1985 during various phases of its ongoing activity. In the last two Kilauea sampling missions, samples were collected during eruptive activity. The samples were analyzed by INAA for over 40 elements. We have found Ir in samples collected at Kilauea and Mauna Loa during fountaining activity as well as after eruptive activity. Os was also seen in the Mauna Loa samples. Enrichment factors for Ir in the volcanic fumes range from 10^4 to 10^5 relative to BHVO.

Flux calculations for Ir at Mauna Loa and Kilauea had ranges of 80 to 3000 and 10 to 315 g/d respectively. The percentage of Ir released from the magma into the fumes ranged from 1% to 12% for both volcanoes. Calculations assuming the Deccan as the source of Ir for the K/T boundary layer show that the concentration of Ir left in the basalts may be too low to account for all of the K/T Ir. It would

require a very high fraction (> 30%) of the Ir to be purged from the basalt to account for all the Ir, which cannot be supported by the Hawaiian data.

The implications of Ir from Hawaiian volcanoes for the K/T Ir anomaly are still unclear. Since Ir has not been found in volcanic fumes other than those at Hawaii, further investigations are needed at other hot spot volcanoes, especially Reunion volcano, to ascertain whether Ir enrichments are present in their fumes.

INTRODUCTION

The discovery of the Ir anomaly and the theory of a bolide impact causing the extinction at the K/T boundary (Alvarez et al., 1980) have set off a large research effort in the area of the K/T extinction. Since then several competing theories have also tried to explain the phenomena found at the K/T boundary (for example, see Officer and Drake, 1983 and 1985).

The finding of Ir in fumes from Kilauea volcano by Zoller et al. (1983) and the corroborative evidence of Olmez et al. (1986) have added fuel to the debate as to the cause of the extinctions at the K/T boundary. The finding of Ir highly enriched in volcanic fumes allows for the possibility that the anomalously high Ir concentrations in the K/T boundary layer came from volcanic emissions and not necessarily from meteoritic material following a bolide impact. Also recent evidence that the Deccan Trap flood basalts occurred at around the same time as the K/T extinction have provided the proponents of the volcanic theory with an event large enough to have theoretically produced all of the excess Ir found in the K/T boundary (Olmez et al., 1986). However, up to this time very low Ir concentrations have been found at the Deccan Traps which adds doubt to its being the source of Ir in the K/T boundary layer (Rocchia et al. 1988).

In this paper, we will discuss our data which have been collected at Mauna Loa and Kilauea volcanoes in Hawaii the past several years and try to point out the implications our data have for the K/T Ir anomaly.

SAMPLE COLLECTION AND ANALYSIS

The samples discussed within this paper were all collected at Mauna Loa and Kilauea volcanoes using the same sampling system. The sampling system consists of a filterpack, a vacuum pump, and a battery. The filterpack holds a Teflon particle filter followed by a series of 4 base-treated filters. The particle filter efficiently collects all particles greater than .01 microns in diameter. The base-treated filters are impregnated with 1 or 3 M ^{7}LiOH in order to collect acidic gases from the fumes, and a series of 4 is used to check for quantitative collection of the gases. The fumes from the volcano are allowed to mix with the atmosphere and cool before samples are collected, as our sampling system is not able to withstand high temperatures. Therefore, the concentrations of elements in the gases will vary from sample to sample depending upon how far from the vent it is collected. This makes absolute elemental concentrations on the filters of limited value. Also only particles and acidic gases are collected. None of the major gases, such as H_2O or CO_2 are retained in our samples. For a more complete description of the sampling system see Finnegan et al. (1989).

Filterpack samples were collected during four separate missions to Kilauea and Mauna Loa. The first two sampling missions to Kilauea were made in November, 1983 and January, 1984, after episodes 11 and 13 respectively at Pu'u O'o vent. Fifteen filterpack samples were collected, and these samples are discussed in detail by Crowe et al. (1987). The next sampling mission was made during the March-April, 1984 eruption of Mauna Loa. Over 100 samples were collected during and after the eruption. These samples are discussed elsewhere by Finnegan et al. (1989). Concurrent with the Mauna Loa eruption, Pu'u O'o vent also erupted (episode 17) and 2 samples were collected during the eruption (Crowe et al., 1987) The last sampling mission to Hawaii was made during the summer of 1985. Samples were collected over a 30 day period before, during, and after episode 35 of the eruption at Pu'u O'o vent (Miller et al., 1989).

We collected three distinctly different types of samples at Mauna Loa and Kilauea, which is important to understand when trying to interpret the data. We call them (for reasons which will become obvious in the pursuing discussion: 1) aircraft, 2) active vent, and 3) cooling vent samples. The aircraft samples were collected by attaching 2 of our filterpacks to the wing of a small aircraft and flying through the ash laden fumes several kilometers downwind of the vent during the lava fountaining phase of an eruption. Active vent samples were collected on the ground during lava fountaining or spattering. These samples were collected as close to the vent as the fountaining activity would allow in heavy fumes usually between 10-100 meters from the vent. Cooling vent samples were collected very close to vents (< 50 meters) after fountaining and spattering activity had ceased, but glowing magma was still present in the vent and temperatures near the vent were still greater than 700 C.

The samples after they were collected, were frozen and returned to the laboratory for analysis. The samples were analyzed using instrumental neutron activation analysis for about 40 elements. The samples were irradiated in the Omega West Reactor at the Los Alamos National Laboratory at a flux of about 1 x 10^{13} n/cm^2 s and an irradiation and counting scheme for the samples is shown in Table 1. More detailed analytical procedures can be found elsewhere (Finnegan, 1984).

DISCUSSION

As mentioned above, approximately 40 elements were detected in the filterpack samples. Although the concentrations for many elements varied by an order of magnitude, typical concentrations for selected elements of interest are given in Table 2 for informational value. It must be mentioned that Os was only seen in the samples from Mauna Loa. This may be because of the extremely concentrated plume at Mauna Loa which was much denser than those encountered at Kilauea, or possibly because we were able to sample closer to the magma during active and cooling sampling periods at Mauna Loa than at Kilauea. Complete element analyses will not be given in this paper but can be found elsewhere (Crowe et al., 1987, Finnegan et al., 1989, Miller et al., 1989). As can be seen in Table 2, the concentrations range from 200 mg/m^3 for S to 3 ng/m^3 for Ir. This gives one a feel for the comparatively small amount of Ir in the volcanic fumes. However, when the fractionation of the Ir from the lava into the fumes is calculated, it can be seen that the Ir is greatly enriched in the fumes. To examine this fractionation, enrichment factors (EF) have been calculated. An EF is defined as follows:

$$EF = \frac{(X/Y)_{fumes}}{(X/Y)_{lava}}$$

where X is the element of interest and Y is the reference element. In normal usage, the reference element is a non-volatile element such as Fe, Al, or Sc. However, because we are looking at the volatilization of elements from the magma, and the ash on the filters is a sort of contamination, we have chosen Br, a volatile element as the reference element. For further discussion on this point see Crowe et al.(1987). In order to conform with the standard notion of an EF, the Br normalized EFs have been multiplied by a factor of 10^5 to bring the EFs for crustal elements to approximately 1. Figure 1 contains the EFs for Mauna Loa samples which are typical for Kilauea as well. The elements are ordered according to increasing EF for the active vent samples. The EFs for the lithophilic elements (ash components) increase from cooling vent to active vent to aircraft samples. This is caused by an increased amount of ash in the fumes for each sample type. As can be seen from the figure, the volatiles and enriched metals have EFs from 10^2 to 10^7. Of most interest in this discussion are the elements Ir, Os, and Au. Their EFs are approximately 10^4 for Au, 10^5 for Ir and 10^6 for Os (Os was not detected in the aircraft samples).

There was some question in our mind as to whether we were collecting Ir quantitatively with our filterpacks. This doubt arose when Zoller et al. (1983) proposed that the Ir was released from the magma as IrF_6. If this was true then any Ir that was still IrF^6 when we sampled would pass through the filters. Up to this point we had only found Ir on our particle filters. However, Wood (1987) showed that the platinum group elements (PGE) are most volatile as chlorides and possibly oxides and also that IrF_6 emissions would not be stable under magmatic conditions. With this in mind, we placed several charcoal filters after a particle filter in a filterpack to see if any Ir could be seen on the charcoal filters. It turned out that we did see a small amount of Ir on the charcoal at Mauna Loa (Finnegan, unpublished data). The occurrence of Ir on the charcoal filters could be explained in two ways. 1) the Ir was present in the gas phase and was collected on the charcoal filters, or 2) the Ir was in the particulate phase, was collected on the particle filter, and then was subsequently revolatilized by a strong acid (sulfuric or hydrochloric) and was then collected on the charcoal. Since we are unable to determine the speciation by our analytical techniques, we cannot distinguish between the two possibilities mentioned above. Still, since Ir was found on the charcoal, the Ir values in this paper should be viewed as lower limits, as some Ir may not have been collected by our base-treated filterpacks. No Ir was ever seen on our base-treated filters.

Using the Ir data from each of our sampling missions to Kilauea and Mauna Loa, we have calculated Ir fluxes. In doing so, we have used the following assumptions: for the Mauna Loa March-April, 1984 eruption the total magma release was 220 x 10^6 m^3 and the S flux was between 10,000 and 30,000 t/d, for the Pu'u O'o eruption at Kilauea from January 1983 to July 1988, the total magma release was 1000 x 10^6 m^3 and the S flux was 5,000 to 15,000 t/d. Other assumptions were that 3 kg of S escaped from each m^3 of magma and the magma had a density of 2.9 g/cm^3. Table 3 contains the results of the Ir flux calculations. The Ir/S ratios are the average values for all of the samples included in the sample heading. All three types of samples collected are included for Mauna Loa and data for 4 separate sampling missions to Kilauea are presented. The mlo83 data were collected at the Mauna Loa Observatory during an eruptive episode of Pu'u O'o vent (Zoller et al., 1983) and only include particulate data as no treated filters were used when these samples were collected. This caused the Ir/S to be high since some of the S was no doubt still present as SO_2. Therefore, the subsequent calculations for the mlo83 data will represent upper limit values. The Ir fluxes calculated in the second column assume that the corresponding Ir/S ratio for a particular sample set is representative of the entire eruption. The range in the fluxes are calculated using the high and low estimates of the S flux. The total Ir released was calculated by multiplying the S released per m^3 of magma times the Ir/S ratio times the total amount of magma erupted. Therefore, for Kilauea, discounting the mlo83 data, the Ir flux estimates range from a low of 10 g/d to 310 g/d and the total Ir released estimates vary from 3 to 32 kg. The last column, % Ir released, looks at the fraction of the Ir removed from the total amount of magma erupted. The Ir concentrations used for Kilauea and Mauna Loa basalts are 0.29 and 0.38 ppb respectively (Orth, unpublished data). This calculation assumes that only magma which is erupted is degassed, therefore the calculated % release should be an upper limit. The air and active vent samples probably have the most representative Ir/S values and thus the fluxes are probably in the 100's of g/d and the % Ir released is most realistically in the several % or less. Similar calculations can be done for the Deccan flood basalts using some similar assumptions. We have assumed that the total volume of magma erupted from the Deccan flood basalts was 10^6 km^3 (Courtillot et al.) with a density of 2.9 g/cm^3. If the Ir concentration in the Deccan is the detection limit of 0.1 ppb of Rocchia et al. (1988) and the total Ir in the K/T boundary layer is 3 x 10^8 kg, (Alvarez et al., 1982, Officer et al., 1987) then about 50% of the total Ir would have had to be released from the magma into the fumes to account for the Ir at the K/T boundary. If the concentration is 0.2 ppb then 30% of the Ir had to be exolved from the magma. These % Ir released values are higher than those for Hawaii by a large amount and there is still the problem of spreading the Ir around the globe in fairly high concentrations.

One way of comparing volcanic or chondritic contributions to the K/T boundary layer is to examine element ratios to Ir in each of the materials. There are obvious limitations to this procedure which include the following: 1) it does not take into account the fractionation of elements over time caused by various processes, 2) it assumes that the volcanic fume compositions from Mauna Loa and Kilauea and from the Deccan Traps are the same, 3) similarly, it is assumes that the bolide impactor had a chondritic composition, 4) volcanic fumes and the bolide may not be the only source for each of the elements in the K/T layer. Table 4 contains selected element ratios to Ir. Elements were selected in a rather arbitrary way. If the element was volatile in the volcanic fumes and there were data in the literature, the element was included in the table. The one exception is Co which was included as a non-volatile minor element enriched in chondrites. The volcanic category includes data just from aircraft samples from Hawaiian volcanoes, as these were deemed the samples most representative of fumes that would travel a reasonable distance. The ranges in the element ratios include the values obtained for all the aircraft samples. The standard Hawaiian basalt

(BHVO) was included so that the fume and basalt composition could be compared. The K/T category includes data from as many sources as could be found in a brief survey of the literature. The references are included in the table notes.

As can be seen in Table 4, there are fairly large ranges for many of the elements in the fumes and the K/T boundary. However, the ranges still give an idea within an order of magnitude of the ratios to expect in each material. There is no conclusive evidence from a comparison of the ratios which material is responsible for the K/T boundary. For the elements Zn, As, and Se, the volcanic fume ratios match up well with the K/T. For the elements Os, Au, and Co the chondrite matches us the best. For Sb and Re neither ratio matches well. The ratio which may be the most revealing however is Os/Ir which would have few other significant sources. This ratio would indicate a chondritic source. In light of the above mentioned limitations however, the ratios must obviously be viewed as very qualitative.

It is difficult to take data from one volcano and try to apply that data to another volcano. However, certain trends should be able to be carried from one to another. With the recent hypothesis that Reunion volcano could have been the source of the Deccan basalts (Courtillot et al., 1986), and that Reunion is a hot spot volcano like Hawaii, there is a good chance of there being similarities between the two. The most difficult problem for the volcanic proponents is the extremely low Ir concentrations in the Deccan basalts. With Ir concentrations less than 0.1 ppb (compared to Mauna Loa's 0.38 ppb) it is difficult to produce enough Ir from the eruption to account for the Ir in the K/T boundary layer without purging a large fraction of the Ir from the magma. Such a large fraction is not supported by the Hawaiian data. The element ratios found in chondrites, volcanic fumes and the K/T are not at all conclusive as both chondrites as well as volcanic fumes have ratios which match well with those found in the K/T boundary layer. However, possibly the most indicative ratio, Os/Ir agrees well with the chondrite composition.

A sampling trip to Reunion Island would be of great benefit. Since Ir has only been detected in the fumes of the Hawaiian volcanoes and not at others (Finnegan et al. 1984), a sampling trip would settle the fact of whether Ir is even being emitted at Reunion. Also having element ratios from the source of the Deccan flood basalts, the data would be much more reliable in doing comparisons with the K/T boundary layer. Possibly the most important data remain to be collected.

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FIGURE 1. Plot of EFs by element for the three sample types collected at Mauna Loa and Kilauea volcanoes. The EFs are normalized to Br using elemental values from BHVO for the source rock. The calculated values are multiplies by 105 to bring the ash EFs to approximately 1. The plotted EFs are the geometric mean for multiple samples. The EFs are ordered by increasing values for the active vent samples.

TABLE 1. IRRADIATION SCHEME

Irradiation	Length	Elements
Sh-shorts	30 sec.	F, Cl
Shorts	5 min.	Al, V, Cu, Mg, Ca, S, Ti, In, Na, Cl, K, Mn, Br, I
Longs	4 hr.	Na, K, Sc, Cr, Ni, Zn, Ga, Se, As, Br, Sr, Rb, Mo, Ag, Sn, Cd, Sb, Ba, Cs, La, Sm, Eu, Tb, Yb, Lu, Hf, Ta, W, Re, Os, Ir, Au, Hg, Th, U

TABLE 2. TYPICAL VOLCANIC FUME CONCENTRATIONS

Element	ug/m^3	Element	ug/m^3
S	200,000	As	1.2
Cl	8,000	Re	0.1
F	5,000	Sb	0.04
Se	20	Os	0.015
Zn	10	Au	0.01
Ni	6	Ir	0.003

TABLE 3. IRIDIUM FLUX CALCULATIONS FOR HAWAIIAN FUME SAMPLES

Sample Set	Ir/S (x 10^6)	Ir Flux (g/d)	Ir_{total} (kg)	% $Ir_{released}$
Mauna Loa 1984				
aircraft[*]	0.76	80-240	2.5	1
active vent[*]	4.4	500-1400	14.5	5
cooling vent[*]	9.9	1000-3000	32.7	12
Kilauea (1983-88)				
mlo83[+]	10	500-1500	150	15
cooling vent 83[§]	0.2	10-30	3.0	0.4
aircraft 84[**]	1.3	70-200	19	2
aircraft 85[++]	2.1	110-320	32	4

[*] Data are from Finnegan et al., 1989.

[+] Data are from Zoller et al., 1983.

[§] Data are from Olmez et al., 1986.

[**] Data are from Crowe et al., 1987.

[++] Data are from Miller et al., 1989.

TABLE 4. SELECTED ELEMENT RATIOS TO IRIDIUM

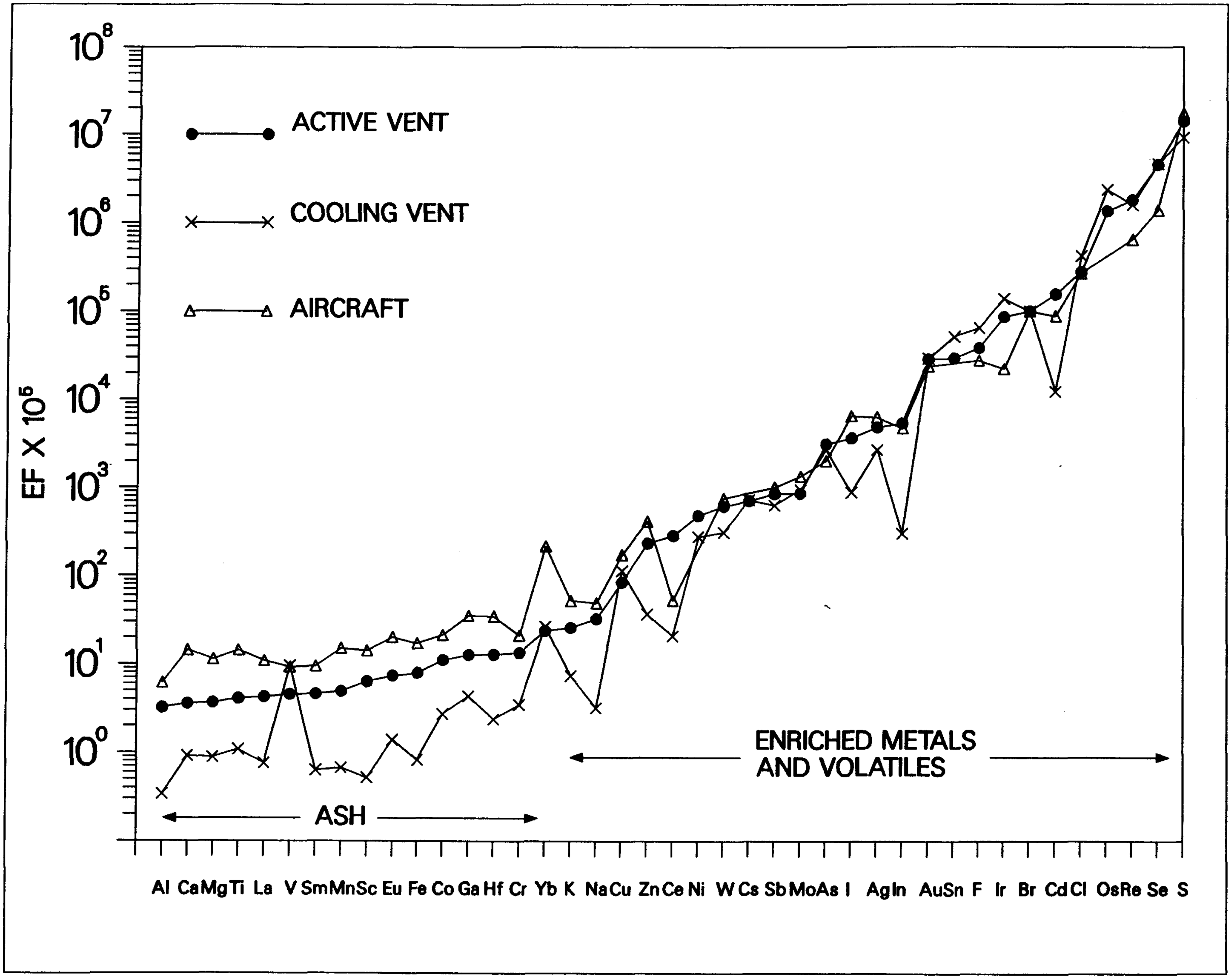
Element	Volcanic[*]	BHVO[†]	Chondrite[‡]	K/T Layer[**]
Co/Ir	30-70	102,000	1050	700-11,000
Zn/Ir	2600,5000	232,000	650	5000-53,000
As/Ir	300-9000	3400	4	400-13,000
Se/Ir	6000-111,000	250	39	200-5000
Sb/Ir	10-20	390	0.3	60-4000
Re/Ir	10-50	1.1	0.8	0.2-0.9
Os/Ir	3-7	0.95	1.1	0.6-1.2
Au/Ir	3-17	3.4	0.29	0.1-1.1

[*] Data are from Finnegan et al., 1989 and Crowe et al., 1987.

[†] BHVO is Basalt, Hawaiian Volcanic Observatory. Data are from Gladney and Goode, 1981.

[‡] Data are from Anders and Ebihara, 1982.

[**] Data are from Alvarez et al., 1980, Bohor et al., 1986, Brooks et al., 1984, Crocket et al., 1988, Ganapathy, 1980, Gilmore et al., 1984, Kyte et al., 1980, Lerbekmo and St. Louis, 1986, Millard, 1987, Orth et al., 1987, and Smit and Hertogen, 1980.


EF X 10⁵
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ACTIVE VENT
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AIRCRAFT
ENRICHED METALS AND VOLATILES
ASH
Al Ca Mg Ti La V Sm Mn Sc Eu Fe Co Ga Hf Cr Yb K Na Cu Zn Ce Ni W Cs Sb Mo As I Ag In Au Sn F Ir Br Cd Cl Os Re Se S