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## MAGMATIC TRITIUM

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### Abstract

This is the final report of a three-year, Laboratory-Directed Research and Development (LDRD) project at the Los Alamos National Laboratory. Detailed geochemical sampling of high-temperature fumaroles, background water, and fresh magmatic products from 14 active volcanoes reveal that they do not produce measurable amounts of tritium ( $^3\text{H}$ ) of deep origin ( $<0.1$  T.U. or  $<0.32$  pCi/kg  $\text{H}_2\text{O}$ ). On the other hand, all volcanoes produce mixtures of meteoric and magmatic fluids that contain measurable  $^3\text{H}$  from the meteoric end-member. The results show that cold fusion is probably not a significant deep earth process but the samples and data have wide application to a host of other volcanological topics.

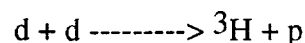
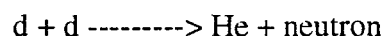
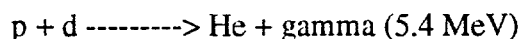
### 1. Background and Research Objectives

Jones et al. (1989) published a provocative paper in Nature proposing that "cold fusion" of deuterons inside the earth is partly responsible for the Earth's interior heat flow and (possibly) for  $^3\text{He}$  anomalies observed in volcanic emissions. These authors have also suggested that d-d fusion in the Earth might cause tritium ( $^3\text{H}$ ) anomalies detectable in volcanic emissions. Although there has been much talk and research on the physics and chemistry of "cold fusion" since 1989, very little volcanological research has been

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conducted on the topic, other than speculation (McHargue et al., 1991; Palmer, 1991; Sheely, 1991). Theoretically,  $^3\text{H}$  and  $^3\text{He}$  could be produced in the following reactions:



In the last fusion reaction,  $^3\text{H}$  ( $t^{1/2}=12.4$  y) would decay to  $^3\text{He}$ . Thus, if  $^3\text{H}$  can be found in magmatic water as opposed to meteoric waters that may contain  $^3\text{H}$  from H-bomb tests and natural atmospheric reactions, this would provide possible evidence for natural fusion in the Earth. Any primordial  $^3\text{H}$  would have decayed away by now.

Water is the dominant gas in most volcanic eruptions (Giggenbach, 1992; Tedesco, 1995), thus determination of stable isotope ( $\delta\text{D}/\delta^{18}\text{O}$ ) and  $^3\text{H}$  content of magmatic water is critical to our understanding of magmatic processes and conditions by which magma is generated in the crust or mantle. These isotopes are necessary to evaluate interactions among magmatic waters, groundwaters, and precipitation. However, no systematic experiments except ours (described below) have been conducted on the  $^3\text{H}$  content of magmatic water because of the short half-life, the large H-bomb excess in the atmosphere, and the large samples ( $\geq 300$  ml) required to accurately analyze low-level  $^3\text{H}$  ( $\leq 5$  T.U. where 1 T.U. = 3.193 pCi/kg  $\text{H}_2\text{O}$ ). The few measurements previously made on the  $^3\text{H}$  content of steam from volcanic fumaroles are poorly constrained by other chemical and isotopic data and display large analytical variations (Mazor et al., 1988). As a result, any  $^3\text{H}$  found in volcanic steam has been explained as meteoric contamination (Quick et al., 1991) and the prevailing wisdom has assumed that  $^3\text{H}$  (magma) is essentially zero.

Our objectives are to determine if anomalous  $^3\text{H}$  is present in magmatic water from active volcanoes, independent of magma composition or tectonic setting, and to determine the source of  $^3\text{H}$ . Because "cold fusion" is a highly controversial subject, we have been told repeatedly by many outside critics that a variety of volcanoes must be investigated to demonstrate whether a universal phenomena exists. As a result, we have analyzed  $\delta\text{D}$ ,  $\delta^{18}\text{O}$ ,  $^3\text{H}$ ,  $^3\text{He}$ , and other chemical/isotopic constituents in sample suites from fumarole condensates, thermal/nonthermal groundwaters, precipitation, and youngest volcanic products at 14 active volcanoes of vastly different magma type and tectonic setting

(Table 1). We began by looking at a variety of subduction zone and hot spot volcanoes of basaltic to rhyolitic composition but, with time, acquired data from volcanoes of more unusual composition or setting (i.e., Vulcano, Italy).

## **2. Importance to LANL's Science and Technology Base and National R&D Needs**

If direct evidence of anomalous  $^3\text{H}$  can be detected in the magmatic water of several volcanoes of contrasting magma type and tectonic setting, it would lend strong support to the theory of natural fusion in the Earth and would require revisions in concepts on primordial  $^3\text{He}$ , mantle heat flow, and plate tectonics (McHargue et al., 1991). However, if we can show that magmatic  $^3\text{H}$  is essentially zero, we can eliminate "cold fusion" as a significant deep earth process.

## **3. Scientific Approach and Accomplishments**

Our approach requires direct sampling of active, high-temperature volcanic fumaroles and fresh lavas, followed by analysis of a variety of chemical/isotopic parameters on the samples. Condensates from fumaroles are collected through Ti and/or pure  $\text{SiO}_2$  glass tubes and a condenser submerged in coolant. Our approach is unique because we collect large samples of condensed magmatic water from many fumaroles for  $^3\text{H}$  analysis (never done before) and because we use three isotopes of water to eliminate effects of meteoric contamination or contamination by near-surface groundwaters (Fig. 1). We also collect extensive background samples of fluids and rocks to compare with magmatic samples.

The approach is demonstrated by our data set for Galeras volcano (Fig. 1). High-T fumaroles ( $\leq 400^\circ\text{C}$ ) occur at the summit of the active cone. The fumarole condensates have  $\text{pH} < 1$  and contain significant amounts of Cl, F, sulfur compounds, and trace metals (Goff et al., 1994). Clearly these condensates are primarily magmatic fluids. The  $\delta\text{D}/\delta^{18}\text{O}$  relations (Fig. 2) show that high-T condensates are isotopically enriched compared to all thermal/nonthermal groundwaters or rain. However, it can be seen that some meteoric water is mixed with magmatic water in even the highest temperature fumaroles, a characteristic common to most volcanic fumaroles (Shevenell and Goff, 1993). The  $\delta\text{D}$  of magmatic water ( $-35\text{‰}$ ) is determined by extrapolating the mixing line to the  $\delta^{18}\text{O}$  value of magma. The latter value is obtained from analyses of fresh lava bombs exploded out of the

crater. Note that fresh lava samples display relatively constant  $\delta^{18}\text{O}$  (+7.37‰) but variable  $\delta\text{D}$  due to open-system magmatic degassing (Taylor et al., 1983).

The same type of mixing relation between magmatic and meteoric waters is revealed in a plot of  $^3\text{H}$  vs.  $\delta^{18}\text{O}$  (Fig. 3). High-T fumarole condensates have  $^3\text{H}/\delta^{18}\text{O}$  values different from all thermal/nonthermal groundwaters or rain. Local meteoric waters (cold springs, streams, and wells) show relatively constant  $\delta^{18}\text{O}$  but variable  $^3\text{H}$  due to presence of anthropogenic  $^3\text{H}$ . Extrapolation of the mixing trend to the  $\delta^{18}\text{O}$  value of magma yields a (statistical)  $^3\text{H}$  content for magmatic water of  $0.00 \pm 0.03$  T.U.

Some of our other data sets show anomalous  $^3\text{H}$  values as high as 3 T.U. (Table 1). We resampled Mount St. Helens in FY94 to see if the previous value would remain constant but the magmatic  $^3\text{H}$  value decreased significantly. The second data set had a better distribution of  $^3\text{H}$  with respect to  $\delta^{18}\text{O}$  values. Some later data sets, such as the one for Vulcano, actually extrapolate to negative  $^3\text{H}$  values. After examining the errors associated with the various analyses and methods, we conclude that extrapolated magmatic  $^3\text{H}$  values greater than 0.0 T.U. shown in Table 1 are caused by slope errors (clustered data) or end-point errors ( $\delta^{18}\text{O}$  of magma varying or not well constrained) during extrapolation.

The secular  $^3\text{H}$  production by *in-situ* neutron irradiation of  $^6\text{Li}$  by U/Th decay can be calculated from the compositions of the lavas (Andrews et al., 1989; Shevenell and Goff, 1996). Contents of Li, B, F, Cl, Sm, Gd, U, and Th as well as the major components must be known to perform the calculations. Secular values of  $<0.01$  T.U. have been calculated from averaged analyses of several lava samples at most volcanoes (Table 1). Although the lavas are too young to be in secular equilibrium, the calculations show the amount of  $^3\text{H}$  that may be produced in the source regions of the magmas. These values are much less than the  $^3\text{H}$  values that we can detect at any of the volcanoes; thus fission reactions are not producing significant magmatic  $^3\text{H}$ .

In FY95, we began to look at more unusual sites such as Vulcano, Italy (microplate boundary trachy-rhyolite). Due to our sampling success, the Yucca Mountain Project funded a sampling expedition in FY95 to Parícutin, Mexico (cinder cone field andesite) because of its similarities to the Lathrop Well cone near Yucca Mountain. While in Mexico, we attempted to sample the giant andesitic stratovolcano of Popocatepetl but it was too

dangerous to sample safely. Also in FY95, we accepted samples for tritium and stable isotope work that were collected by T. Fischer, Arizona State University, at Kudriavy (Russia) and Papandayan (Indonesia).

In FY96, we combined efforts with another LDRD project (Remote Sensing-Volcanology Subtask, F. Goff, P.I.) to reduce logistical costs while taking samples and making measurements at White Island and Ruapehu volcanoes, New Zealand. Even though fumaroles were hot ( $\leq 400^{\circ}\text{C}$ ), the White Island data suffers from masking effects by a geothermal system lying above subsurface magma, an effect also noticed at Alcedo Volcano in the Galapagos Islands of Ecuador. Ruapehu was too active to sample completely and safely for this investigation, although perfect for the remote sensing study.

In conclusion, after investigating 14 active volcanoes we find that the magmatic  $^3\text{H}$  is  $< 0.1$  T.U., lower than our sampling and analytical capabilities can measure. These results imply that "cold fusion" does not occur in the deep earth to any measurable extent. We are in the process of writing up these results for journal publication.

Although we did not find magmatic tritium, this project had a series of successes with respect to other volcanic and geochemical processes. Our samples provided information on the rapid mixing of young meteoric water with degassing magmatic water in the porous cones and domes of young volcanoes (Shevenell and Goff, 1993). Our samples were used to perform secular  $^3\text{H}$  calculations on a variety of young magmas (i.e., Shevenell and Goff, 1996). Our samples show that magmatic volatiles mobilize many metals including gold due to their extreme acidity and ligand content (Fig. 1; Goff et al., 1994). Our samples offer additional insights into other degassing phenomena in magmas (Goff, 1996).

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4. Goff, F., "Vesicle Cylinders in Vapor-differentiated Basalt Flows," *Jour. Volcanol. Geotherm. Research*, **71**, 167-185 (1996).

5. Shevenell, L., and Goff, F., "In Situ Tritium Production and Fluid Mean Residence Times in Two Subsystems of the Valles Caldera Hydrothermal System, New Mexico," in (F. Goff, B. Kues, M. Roger, L. McFadden, and J. Gardner, eds.) New Mexico Geol. Soc. Guidebook, 47th Field Conference, Jemez Mountains Region, pp. 455-462 (1996).

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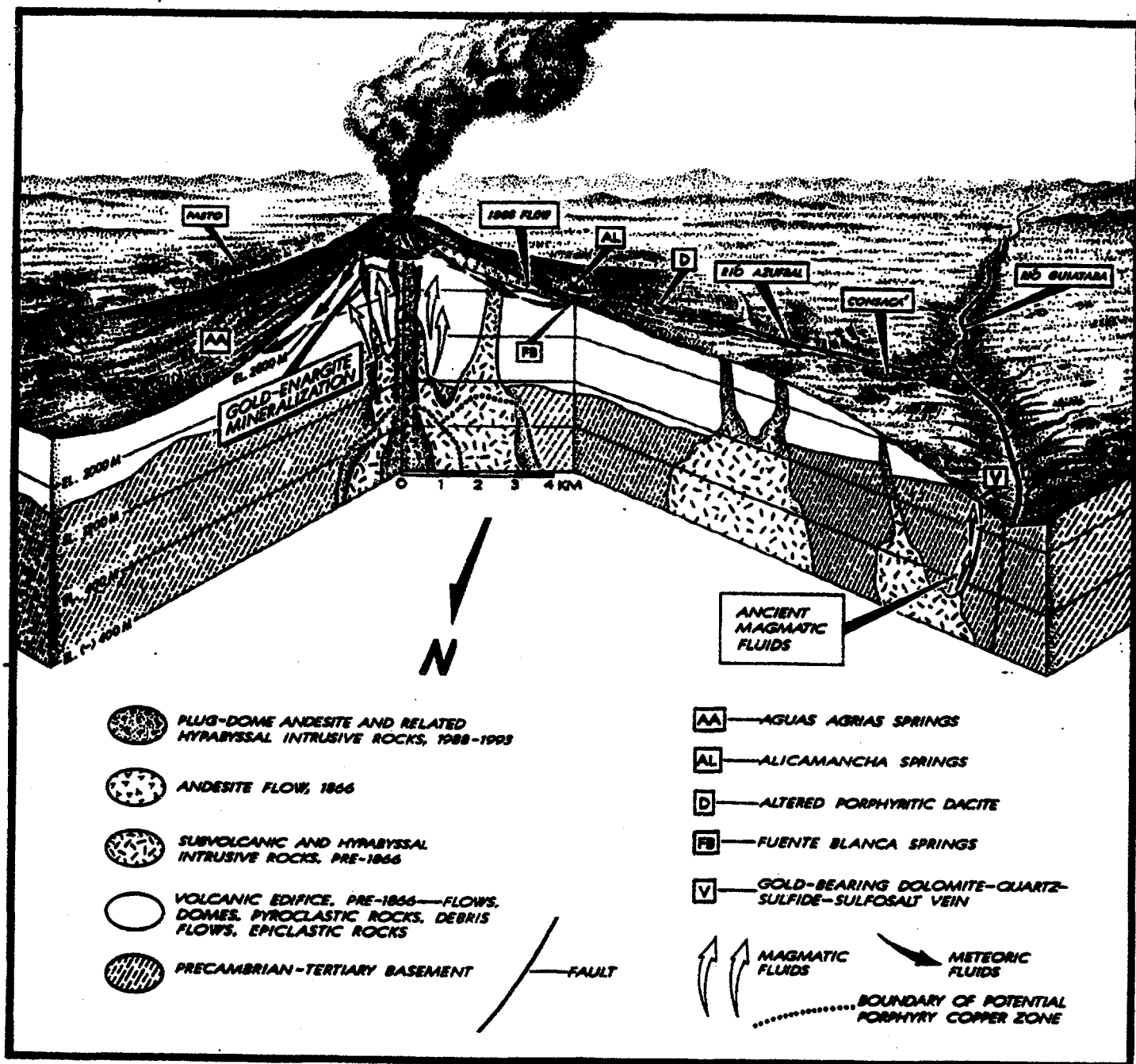


Figure 1: Modified cut-away perspective drawing of Galeras volcano (viewed from the north) showing the basic geology and configuration of shallow hydrothermal system. Magmatic volatiles mix with young near-surface groundwaters to form acid springs. Magmatic fluids discharging inside the volcano create conditions favorable for deposition of gold and copper. Magmatic fluids discharging at the summit have mixed with precipitation falling in the summit crater and cone. The resulting mixtures contain measurable tritium from the meteoric end-member but no detectable tritium from a deep source (from Goff et al., 1994).

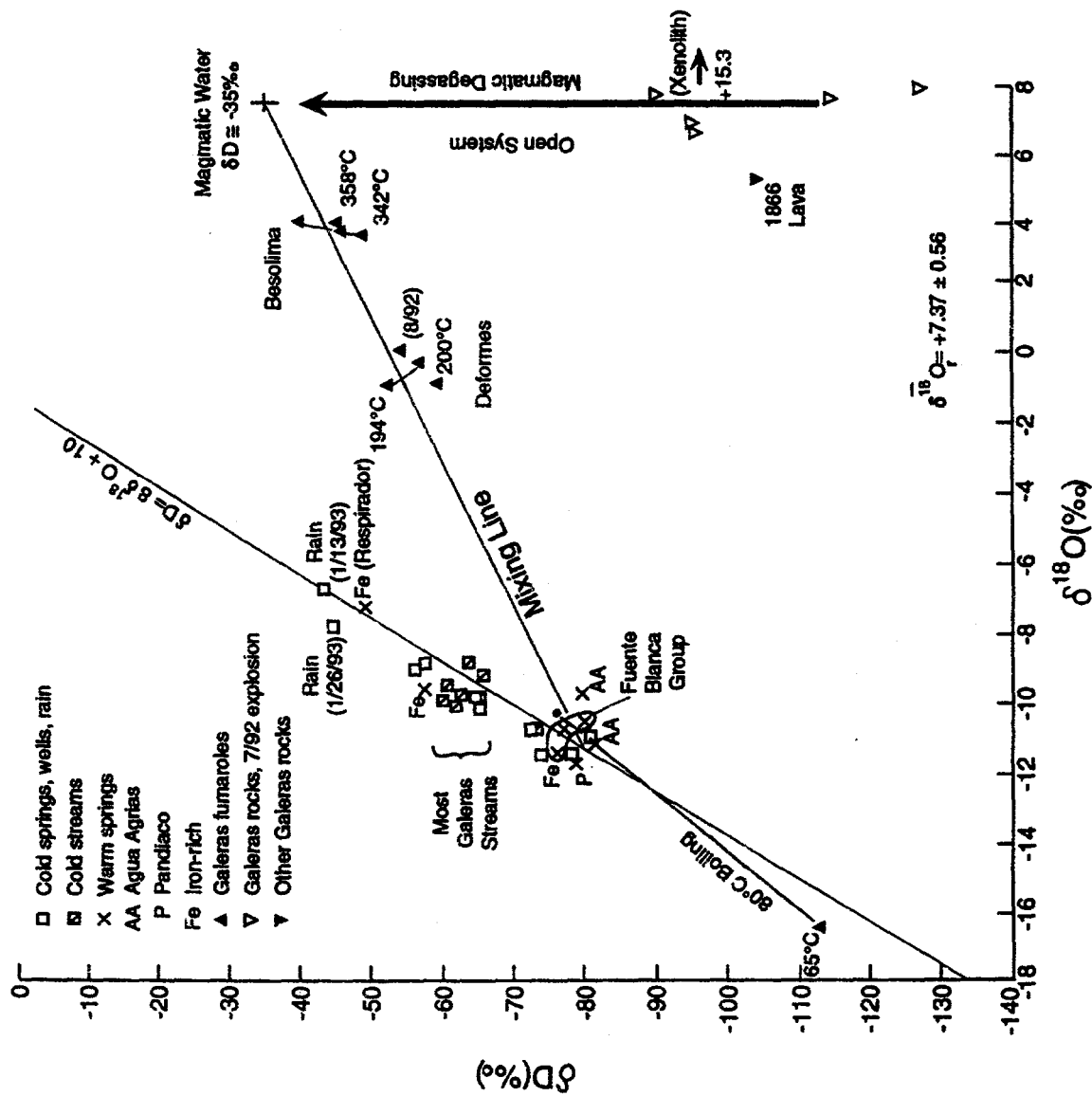


Figure 2: Plot of deuterium versus oxygen-18 for waters from Galeras volcano, Colombia. The fumarole condensates lie on a mixing line between magmatic and meteoric end-members. The extrapolated deuterium of the magmatic water is about -35‰.

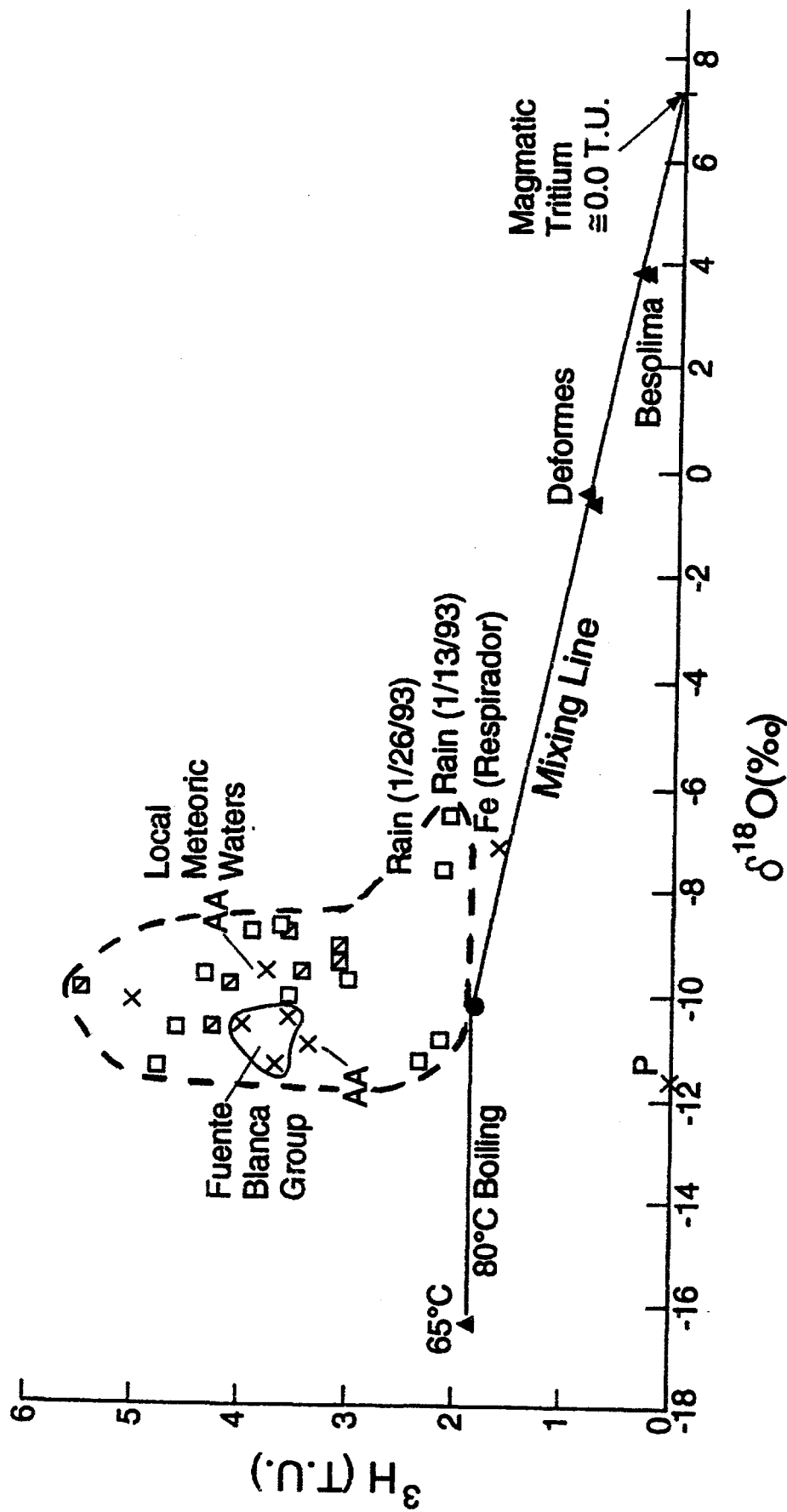


Figure 3: Plot of tritium versus oxygen-18 for waters from Galeras volcano, Colombia. When the tritium data for the fumaroles are extrapolated to the oxygen-18 value for magma, the resulting magmatic tritium is 0.0 T.U.

TABLE 1: Tritium Contents of Magmatic Fluids and Other Information for Volcanoes of This Investigation. (all volcanoes visited and samples collected by F. Goff except for Kudriav and Papandayan samples which were provided by T. Fischer, Arizona State University).

Volcano	Date	Type & <sup>1</sup> Setting	Temp <sup>2</sup> (°C)	Measured <sup>3</sup> H (T.U.)	Magmatic <sup>3</sup> <sup>3</sup> H (T.U.)	Secular <sup>4</sup> <sup>3</sup> H (T.U.)	Percent <sup>5</sup> Meteoritic	Funding <sup>6</sup> Organ.
Mount St. Helens	1988-89	Dacite/SZ	≤270	4.66-4.41	3.01±0.52	<0.01	75-50	OBES-AC
Mount St. Helens	1994	"	≤560	2.79-2.71	0.65±0.25	"	60-55	LDRD
Pu'u O'o, Kilauea	1990-91	Basalt/HS	≤275	3.03-2.40	2.97±0.15	<0.01	80-20	OBES-AC
Pacaya, Guat	1992	Basalt/SZ	≤340	1.25-0.92	0.76±0.05	<0.01	20-10	OBES-AC
Galeras, Colom	1993	Andesite/SZ	≤360	0.81-0.32	0.0	<0.01	50-20	LDRD
Satuma, Japan	1993	Rhyolite/SZ	≤890	0.42-0.04	0.0	<0.01	10-5	LDRD
Sierra Negra, Ecud*	1995	Basalt/HS	≤210	0.48-0.12	---	<0.01	>95	LDRD
Alcedo, Ecud**	1995	Basalt/HS	≤100	15.4-0.30	---	<0.01	>95	LDRD
Vulcano, Italy	1995	Trachyte/MP	≤550	0.84-0.27	0.0	<0.01	34-17	LDRD
Paricutin, Mex *	1995	Andesite/CC	≤270	---	---	---	100	YMP
Popocatepetl, Mex	1995	Andesite/SZ	---	Too Dangerous To Sample!	---	---	---	YMP-LDRD
Kudriav, Russ	1995	Basalt/SZ	≤920	2.45-0.06	0.0	<0.01	75-8	LDRD
Papandayan, Indon	1995	Andesite/SZ	≤280	0.01	0.0	---	20	LDRD
White Island, NZ**	1996	Andesite/SZ	≤400	0.24-0.04	0.0?	---	80?	LDRD
Ruapehu, NZ	1996	Andesite/SZ	≤250	Too Dangerous To Sample Completely!	---	---	---	LDRD

<sup>1</sup>Type=magma type; Setting: SZ=subduction zone; HS= hot spot; MP=microplate boundary; CC=cinder cone field.

<sup>2</sup>Temperature of hottest fumaroles that could be safely sampled.

<sup>3</sup>Magmatic value determined by extrapolation of <sup>3</sup>H vs. <sup>18</sup>O plot as per Fig. 1b

<sup>4</sup>Calculated from major element analysis of fresh rock and B, Gd, Li, Sm, Th, and U contents (Andrews et al., 1989; Shevenell and Goff, 1996).

<sup>5</sup>Meteoritic water calculated from mixing proportions as per Fig. 1a.

<sup>6</sup>OBES-AC=Office of Basic Energy Sciences, Advanced Concepts; YMP=Yucca Mountain Waste Repository Project.

\*No magmatic water in fumaroles; essentially all meteoritic water.

\*\*Magmatic water masked by interaction with geothermal system between magma body and surface fumaroles. At White Island this is probably modified seawater.