

THE RECOGNITION OF QUENCHED MAGMATIC GASES IN FUMARoles AS A GEOTHERMAL EXPLORATION TOOL

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ABSTRACT

Magmatic and quenched magmatic gases in fumaroles can be identified by using a $H_2S-H_2-CH_4$ ternary diagram even where SO_2 , HCl and HF have been scrubbed from the gases or have not been analysed for. Standard gas geothermometry is not applicable to such gases although high temperatures at depth are implied by their presence. However, fluids at depth in the immediate vicinity of fumaroles with this chemistry are likely to be acid or produce acid condensates and are not currently exploitable — although outlying convective parts of the same system may be exploitable. The diagram is also useful for identifying neutral reservoirs where overlying fumaroles are superheated, a feature occasionally mistaken as magmatic.

Whilst most quenched magmatic gases have an “andesitic composition”, often represented by their position on $N_2-He-Ar$ plots, they are not exclusive to it. Gases from neutral reservoirs can also have “andesitic” compositions, hence the $N_2-He-Ar$ diagram should not be misused as an indicator of magmatic acidity at depth.

1.0 INTRODUCTION

In geothermal systems developed in areas of high topographic relief, associated hot springs may be condensates or be located along outflows well away from any potentially exploitable reservoir (Bogie and Lawless, 1987) and be diluted by groundwaters, contaminated by condensates, and re-equilibrated. Therefore such waters may no longer provide any useful information regarding any potentially exploitable reservoir at depth. Thus fumaroles, if they occur, may provide the only information regarding the nature of any potentially exploitable geothermal reservoir in terms of temperature, enthalpy and acidity. The potential for magmatic acidity is usually evaluated by analysing for the magmatic volatiles: SO_2 , HCl and HF . However, these highly reactive gases are not always found to be present in fumaroles associated with acid magmatic reservoirs, or are not always analysed for, particularly in historic analyses. The purpose of this paper is to provide a means of recognising fumaroles associated with reservoirs that contain acid magmatic volatiles, where these vent to the surface, and where SO_2 , HCl and HF are either not detected or not reported.

Superheated fumarolic gases containing SO_2 , HCl and HF are of clear acid-magmatic origin and are usually associated with volcanic centres with historic eruptions. These gases (and the others accompanying them) can be considered to be derived directly from a degassing magma body and imply high-temperature, vapour conditions at depth. They mark currently non-exploitable immature geothermal systems or non-exploitable parts of a larger system in terms of having a locally developed acid reservoir, high gas contents and possibly an unacceptably high level of volcanic risk. However, in the same systems there can also exist fumaroles without HCl , SO_2 and HF , but which, by association, obviously have an acid magmatic source. These fumaroles have lost the soluble, acidic gases as a result of condensation or interaction with groundwater near-surface. The existence of acid-chloride springs at slightly lower elevations can provide evidence for this. The fumaroles can be considered to have a “quenched magmatic” composition, and although they have lost their primary magmatic signature, they may still retain other distinctive chemical characteristics that distinguish them from those boiled off near-neutral reservoirs. The same type of “quenched magmatic” features may also be found without associated magmatic thermal activity, in which case recognition of their magmatic affinities is of considerable importance. It is also possible to find superheated fumaroles derived from near neutral geothermal reservoirs and these need to be clearly identified as

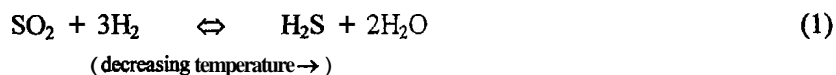
such. This paper develops a technique for determining on the basis of H₂, H₂S and CH₄ relationship, whether a fumarole without SO₂, HCl, or HF, may in fact be directly associated with an acid magmatic system.

20 DATA COLLECTION

Data on fumarole chemistries have been compiled **from** the literature and from some projects that Kingston Morrison has worked on in Iran and Indonesia. These data are collated in Table 1. In order to minimise any variations resulting from differences in tectonic environments, only analyses of gases from fumaroles related to convergent plate margins are considered here. Some of the previously unpublished data is commercially sensitive and only the general location of the area from which the analysis was obtained is given. These analyses are all from separate fields and are representative compositions of high **flow** rate fumaroles, where samples have been obtained that have insignificant or no atmospheric contamination. O₂, N₂ and Ar analyses are provided to establish this. Similarly, analyses from the literature with indications of air contamination have not been included. Analyses in bold type are those **from** systems that are clearly magmatic as shown by the presence of HCl or SO₂. Where this is the case other indications of magmatic origins such **as** volcanic activity, supercritical fumarole temperatures, or acid-cl crater lakes are also present. Despite the recognition that HF is commonly present **as** a magmatic volatile, very few analyses, particularly those with a full range of other gases, are available in the literature and HF has not **been** included in the table.

3.0 DISCUSSION

Table 1 shows that fumaroles with supercritical temperatures (>374°C) and hence of clear magmatic origin have distinct compositions. They contain SO₂ and HCl and have high H₂ concentrations, moderate concentrations of H₂S and low concentrations of CH₄. However, included in the data set are lower temperature fumaroles ranging in temperature from local boiling point to 115°C that **also** contain SO₂ and/or HCl, they also have low CH₄ concentrations, but differ by having high H₂S and low H₂ concentrations. These relationships can be **best** illustrated by a ternary diagram for H₂S, H₂ and CH₄ with appropriate weightings to spread the plots over the diagram. This is done in Figure 1 where the sample numbers (from Table 1) are shown and Figure 2 where the measured temperatures of the fumaroles are indicated. With the exception of one point (sample No 8 **from** Papandayan), all the analyses from fumaroles with supercritical temperatures plot in the H₂ corner of the diagram. The gas compositions from lower temperature fumaroles that contain either SO₂ and/or HCl plot in the H₂S corner; as do a number of analyses where SO₂ or HCl are not reported. Some points plot along the H₂S-H₂ side of the diagram suggestive of a range **between** these two **sets** of compositions. **One** of these (8) is **from** a 400°C fumarole from Papandayan, while a 95°C fumarole from Papandayan (9) has a composition that plots in the H₂S corner of the diagram. The variation in composition **of** the magmatic gas samples, **from** H₂ rich at high temperature to H₂S rich at lower temperatures **can** be accounted for by the reaction:



However, the proportion of H₂ to SO₂ in typical magmatic gases is insufficient to convert all the SO₂ to H₂S. **As** noted by Giggenbach (1987) additional H₂ **can** be produced from oxidation of Fe²⁺ in the **rock** matrix, but this reaction is apparently not fast enough to supply the necessary H₂ to maintain equilibrium in reaction (1). **As** a result there is often considerable SO₂ in lower temperature fumaroles and this is **consumed** primarily by the precipitation of native sulphur:

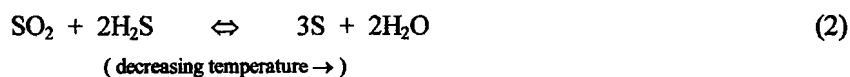


Table 1
Representative Fumarole Gas Analyses
(Clearly Magmatic Fumaroles in Bold)

No	Country	Area	Ref	S ^o	Temp °C	Xg	CO ₂	H ₂ S	NH ₃	He	H ₂	O ₂	Ar	N ₂	CH ₄	SO ₂	HCl	Total Gas wt%
											mmol/mol dry gas							
1	Cobrnbnh	El Ruiz	J	-	82	0.0454	516	38.8	-	0.0037	1.90	-	0.005	58	0.01	436	1.30	5.7
2	Cqsta Rica	Miravalles	h	-	98		983	9.0	-	0.0025	1.20	-	0.020	3.0	3.10	NR	NR	
3	Guatamala	Zunil	h	-	87		930	36.0	-	0.0045	6.44	-	0.200	25.0	1.30	NR	NR	
4	Guatamala	Tecuamburro	i	-	95	0.0440	835	130.9	-		0.02	0.000	0.150	33.6	0.09	NR	NR	9.3
5	Indonesia	Wayang Windu	c	n	93	0.0240	937	46.2	-	0.0060	1.02	-	0.014	14.7	0.65	NR	NR	5.4
6	Indonesia	Wayang Windu	c	n	94	0.0790	936	49.7	-	0.0050	0.51	-	0.014	13.4	0.19	NR	NR	15.9
7	Indonesia	Wayang Windu	c	n	91	0.0400	908	53.0	-	0.0100	0.26	-	0.100	38.5	0.05	NR	ND	8.7
8	Indonesia	Papandayan	J	y	400	0.0410	717	70.8	-		3.42	0.000	0.010	13.4	0.01	165	30.52	7.2
9	Indonesia	Papandayan	c	y	95	0.0400	594	399.0	-	0.0030	0.25	-	0.005	7.1	0.03	NR	NR	8.1
10	Indonesia	Sumatraa	e	n	98	0.0111	942	35.4	0.15	0.0031	10.6	0.000	0.011	11.8	0.09	NR	ND	2.6
11	Indonesia	Sumatra b	e	n	130	0.0021	886	83.6	3.91	0.0185	12.7	0.006	0.085	8.3	5.67	ND	ND	0.5
12	Indonesia	Sumatra c	e	n	97	0.0090	899	88.7	0.16	0.0060	3.30	0.000	0.023	8.3	0.83	NR	ND	2.1
13	Indonesia	Sumatra d	e	n	101	0.0223	964	17.0	0.03	0.0200	3.61	0.000	0.071	8.7	6.30	NR	ND	5.1
14	Indonesia	Sumatra e	e	y	105	0.0100	723	272.7	0.06	0.0085	0.014	0.006	0.024	3.9	0.28	ND	ND	2.2
15	Indonesia	Sumatra f	e	n	100	0.0093	843	141.0	0.02	0.0029	7.58	0.006	0.030	7.6	1.03	NR	NR	2.1
16	Indonesia	Sumatra g	e	n	94	0.0255	981	7.2	1.20	0.0056	1.36	0.000	0.012	6.9	2.14	NR	NR	5.8
17	Indonesia	Sumatra h	e	n	93	0.0053	941	23.8	-	0.0130	5.80	0.023	0.412	28.0	1.64	NR	NR	1.2
18	Indonesia	Darajat	e	n	118	0.0072	943	33.1	0.18	0.0293	12.3	0.000	0.068	11.2	0.35	NR	NR	1.7
19	Indonesia	Sumatra i	e	y	94	0.0160	761	234.0	0.03	0.0010	0.02	0.007	0.013	5.1	0.16	R	R	3.6
20	Indonesia	Sumatra j	e	y	112	0.0650	821	173.0	0.13	0.0050	0.01	0.017	0.008	6.1	0.03	NR	0.02	13.2
21	Indonesia	Sumatra k	e	y	95	0.0149	786	225.2	-	0.0090	0.10	0.005	0.187	7.0	1.28	NR	1.20	3.3
22	Indonesia	Merapi 78-3	f	y	820	0.0625	732	75.2	-		89.7	-	0.80	10.8	0.16	82	NR	11.0
23	Iran	Sabalan	d	n			942	48.0	-		0.01	-		10.0	0.04	NR	NR	
24	Japan	Showashinzan	j	y	336	0.0011	377	40.4	-		305	-	0.099	126	0.45	31	233.45	0.1
25	Japan	Mt Usu	j	y	690	0.0067	571	30.5	-		286	-	0.050	178	0.79	34	51.87	1.0
26	Mexico	Cerro Prieto	g	-	100		807	12.5	125		36.1	-	0.460	19.7	2.30	NR	NR	
27	New Zealand	Wairakei	h	n	104		975	7.0	-	0.0220	2.40	-	0.300	14.0	2.80	NR	NR	
28	New Zealand	Hgauruhoe	J	y	640	0.0399	402	170.3	-		66.1	-	0.053	426	0.01	255	62.60	5.2
29	New Zealand	Ketetahi	j	-	136	0.0417	906	31.2	-		244	-	0.012	12.9	25.64	0	0.00	8.8
30	New Zealand	White Is.	j	y	760	0.0720	735	34.0	-		42.0	-	0.007	2.9	0.14	113	71.98	11.8
31	New Zealand	White Is.	j	m	115	0.0250	820	150.0	-		0.60	-	0.070	10.2	8.90	5	3.20	5.5
32	Nicaragua	Momotombo	i	-			822	48.0	-		29.0	-		78.0	9.00	NR	NR	
33	Philippines	Mambucal	a	-	85	0.5370	970	7.0	-	0.0069	0.42	0.000	0.041	15.9	6.10	NR	NR	56.4
34	Philippines	Mt Apo	a	-	95	0.0055	957	15.0	-	0.0058	3.91	0.040	0.420	23.0	0.26	NR	NR	1.3
35	Philippines	Mt Pinatubo	a	-	98		898	92.0	-	0.0042	0.05	0.190	0.023	9.5	0.08	NR	NR	
36	Philippines	Billiran	b	-	102	0.0340	888	99.0	-		0.10	-		6.3	0.20	R	0.90	7.4
37	Philippines	Tongonan	k	n	95	0.0139	944	42.1	-		5.78	0.000	0.067	5.8	2.70	NR	NR	3.2
38	Philippines	Mahanagdong	k	-	98	0.0969	989	6.1	-		0.49	0.000	0.005	2.1	1.84	NR	NR	19.1
39	Philippines	Alto Peak	l	n		0.0487	941	32.0	-		20.5	-	0.026	2.5	3.97	NR	NR	10.3
40	USA	Mt St Helens	J	y	540	0.0150	607	91.1	0.00		1622	0.000	0.051	31.2	0.01	48	59.16	2.5

NOTES

- a) Giggenbach and Poreda (1993)
 b) Lawless and Gonzales (1962)
 c) MandalaNusantara Ltd. (1997)
 d) Khosrawi et al. (in prep)
 e) Unpublished data.
 f) Le Guern et al. (1982)
 g) Nehring and D'Amore (1964)
 h) Giggenbach (1987)
 i) D'Amore and Panichi 1980.
 j) Giggenbach (1990)
 k) Sabonga and Auman (1997).
 l) Parilla Jr. et al. (1997)
 m) Giggenbach (1982)

ref - source of data (see reference list)

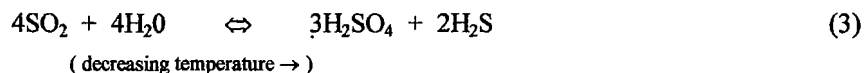
S^o - heavy sulfur depositing?Xg - molar gas ratio (moles gas / moles H₂O)

NR - Not Reported

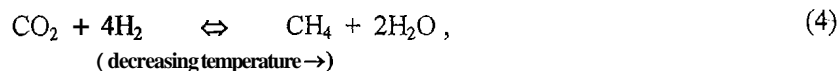
ND - Not Detected

R - Reported Qualitatively

Equation (2) explains the abundance of sulphur (often in the **form** of solfatara chimneys) around fumaroles which produce magmatic gases. Sulphur also forms at the surface through the simple oxidation of H₂S in air **but** this is more common around weaker features where poorly dispersed gas mixes with air. Evidence that reaction (2) is occurring (and that SO₂ is present close to the surface) is provided by the observation of sulphur forming **inside** high-velocity solfatara vents, where O₂ is clearly absent. Even with these apparently magmatic features, HCl can be absent and SO₂ very low if reaction 2 is close to completion (eg. sample 14). Therefore, the observation of heavy sulphur deposition (in the absence of O₂) is evidence in itself of magmatic conditions at depth. If upflowing magmatic gases are quenched by condensation in ground water, HCl will be scrubbed out and SO₂ may be consumed by the following reaction, giving rise to acid-Cl-SO₄ waters.



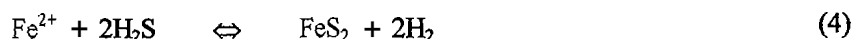
These acidic waters may appear at high elevations close to the magmatic upflow. **They** can also be transported downslope through permeable lava flows, giving rise to high-flowrate, acid-Cl springs on the lower **flanks** of the volcano, displaced from the magmatic centre of the system. This may give a false impression of the extent of acidic conditions at depth. Regional hydrology and structural/ lithological controls need to be carefully considered in assessing the distribution of acid fluids. Reaction (1) accounts for the generally low concentration of CH₄ in samples from magmatic fumarole. The concentration of CH₄ is controlled by the Fisher-Tropsch reaction:



If most of the H₂ is removed via reaction (1), as appears to be the case for most quenched magmatic samples, no new CH₄ can form from the above reaction. There are exceptions however, as shown by White Island sample (31) which plots towards the CH₄ corner of Figure 1. In this case it appears SO₂ has **been scrubbed** out on the periphery of the magmatic upflow, before equation (1) proceeds to completion, but at a sufficiently high temperature to allow generation of methane via equation (4). Similarly sample (21) also contains significant CH₄ but still has a distinctly low H₂ content and may also be being produced by this process.

Symonds and Gerlach (1998) modelled the quenching process by calculating the progressive mixing of a 915°C magmatic gas from the Merapi volcano with water. Although they did not include CH₄ in their calculations, they demonstrate the increase in H₂S and decrease in H₂ observed in the lower temperature fumaroles and the scrubbing out of HCl. Hence analyses with high H₂S, and very low CH₄ and H₂, but without reported SO₂ and HCl **can** also be considered to be quenched magmatic **gases**. Without this knowledge, quenched magmatic gas chemistry can present interpretative dilemmas, particularly when gas geothermometry is applied. The fumaroles may be physically impressive and located on apparently youthful volcanic centres, but the low H₂ concentrations can give anomalously low geothermometer temperatures — particularly using the H₂/Ar geothermometer of Giggenbach (1992). Another process by which a high proportion of H₂S and low H₂ and CH₄ can be obtained is by repeated boiling. Both H₂ and CH₄ are much less soluble than H₂S and are lost rapidly **on** initial boiling leaving some of the **H₂S** in solution. If such a water then moved laterally and boiled again it would produce a high proportion of **H₂S** in comparison to H₂ **or** CH₄. This is illustrated by the plots of samples 5 to 7 in Figure 1. They lie on a line indicative of progressive gas loss from solution and whilst H₂ and CH₄ concentrations are low they are not as low as that observed where there is other evidence for the **gases** being cooled magmatic gases. This is because some degree of re-equilibration to produce H₂ and CH₄ would **occur** during lateral movement which would **also** consume H₂S, **see** equation (4 and 5). Evidence of boiling and degassing is **often seen** in the low total gas contents of these features, in contrast to the relatively high gas contents in the magmatic fumaroles. Conversely if boiled-off gases are trapped in a **steam** zone, **or** the field is vapour dominated, the **gases** will re-equilibrate to the vapour phase and become H₂ rich and H₂S poor and plot towards the H₂ corner in Figure 1, **for** example sample (18) from Darajat.

Figure 1 can therefore be used to identify three groups of gases: primary magmatic gases, quenched magmatic gases and re-equilibrated gases. The fumaroles that have compositions that plot away from the H₂-H₂S and H₂S-CH₄ sides of the diagram come from fields that have been for the most part successfully exploited or by implication are viable prospects themselves, at least in terms of not containing magmatic gases with potential for producing acidity. An exception to this is Alto Peak (Reyes *et al.*, 1993), where the fumarole chemistry is indicative of a neutral reservoir and whilst a neutral reservoir was initially encountered, deeper drilling encountered acid magmatic volatiles. This is a result of the massive cold water influx from the surface being sufficient to quench the magmatic volatiles at depth, so there was re-equilibration of gases before they got to the surface, but not at sufficient depth to yield an exploitable system. Hence the absence of fumaroles with magmatic or quenched magmatic chemistry does not guarantee the absence of acid magmatic volatiles at depth, particularly where the field is in an area of high rain fall. However the main source of gases in exploitable fields will still be mainly primary magmatic, these gases have been cooled, neutralised and partially incorporated into hydrothermal mineral assemblages. Exploitable fields are those where this process is removed from the acid-magmatic centre in time or space. In terms of the species represented on Figure 1 the main process for this change in chemistry is deposition of pyrite where iron is derived from the country rock and the H₂S is derived from equations (1) and (3) according to:



The H₂ released reacts with CO₂ to produce CH₄ thereby driving plots towards the CH₄ corner of the diagram, although as pointed out by Giggenbach (1992), the Fisher-Tropsch reaction is very slow at temperatures less than about 250°C. During this process gas equilibria are established which are those calibrated for gas geothermometers (D'Amore and Panichi, 1980 and Giggenbach, 1992). Therefore gas geothermometers should not be utilised on fumaroles that do not plot in the neutralised part of the diagram.

He and Ar are only reported for some of the analyses, these can be plotted on to the N₂-He-Ar plot of Giggenbach (1987) to establish sources for the gases (Figure 3). Analyses indicated to be "magmatic" on this diagram are those that either contain SO₂ or HCl, or plot in the same areas as those that do on Figure 1. As would

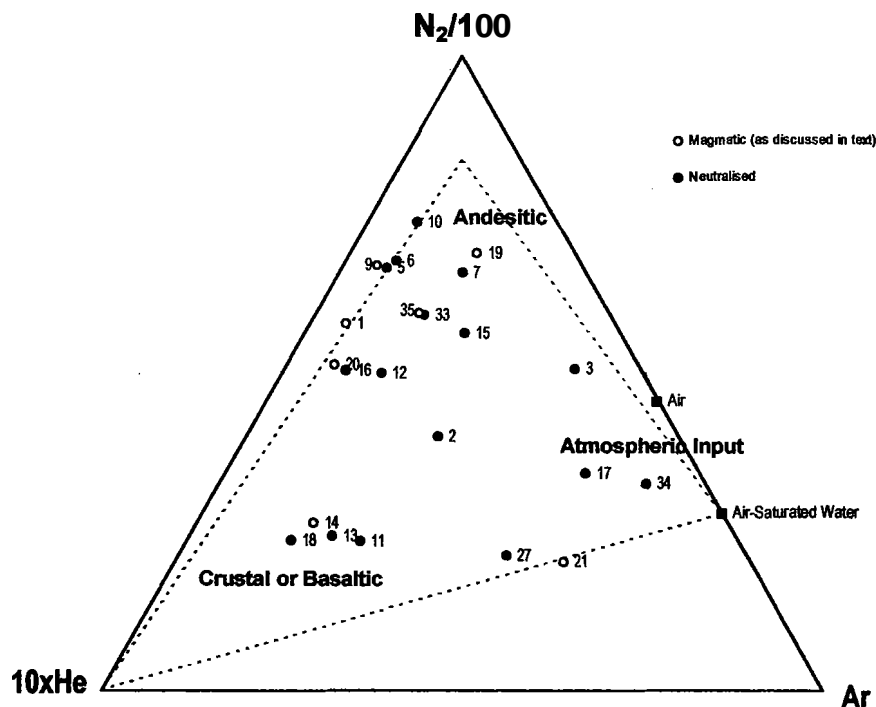


Figure 3. Relative N₂, H₂ and Ar contents of fumarole steam, with sample numbers shown (from Table 1)

be expected, the majority of analyses plot in the andesitic gas area, or on possible mixing lines with air saturated water. However, it must be remembered that N_2 , He and Ar are highly conservative gases, with proportions that survive the neutralisation process described above. Therefore, whilst this diagram indicates source it is not indicating neutralisation and can not be used to discriminate acid reservoirs, nor did the progenitor of the diagram ever advocate this (Giggenbach 1992). A second group of analyses plots towards the basaltic α crustal input area, α on possible mixing lines with air saturated water. "Magmatic" samples are found within this group and the fact that a sample plots here also indicates nothing with regard to its degree of neutralisation.

Another property of fumaroles that is measured, but not always reported, is the total gas content, given as Xg in Table 1. The range of gas contents in each of the different groups of primary magmatic gases, quenched magmatic gases and re-equilibrated gases, overlaps such that the gas content can not be used to discriminate them. Neutralised gases have the widest range and the single highest value possibly reflecting lower temperatures and greater opportunity for condensation of steam. Consequently using fumarole gas contents to estimate reservoir gas contents can at best only derive a maximum and high gas concentrations cannot be used as a predictor of acid conditions at depth. Within a single field however, it is important to look at the variation in total gas contents as this provides information about condensation and boiling processes, which in turn gives an indication of which data are most representative of the deep reservoir.

4.0 CONCLUSIONS

Magmatic fumarole steam that has been quenched and SO_2 and HCl removed (or not analysed) can be recognised by its high H_2S and low H_2 and CH_4 contents. Such a chemistry can be best recognised on a H_2S - H_2 - CH_4 ternary plot. Standard gas geothermometers are not applicable to gases of this composition, although high temperatures are implied by the acid-magmatic associations. Deep fluids, including steam condensates, may be too acid to exploit in the immediate vicinity of such fumaroles. Therefore, recognition of fumarole gases with a quenched chemistry, and where other indications of acid conditions at depth are lacking, is useful in guiding exploration programmes away from potentially acid areas at depth. Neutral hydrothermal conditions may still exist beyond the zone of acid-magmatic conditions. The above technique provides a method of identifying acid-magmatic conditions *only* where suitable fumaroles exist. Whether magmatic vapours reach the surface with their magmatic signature intact will depend to a large degree on structural/ lithological controls and shallow hydrology. So, the absence of magmatic volatiles at the surface does not guarantee finding a neutral resource. This method is useful for identifying gases from neutral reservoirs where they occur in superheated fumaroles, a feature occasionally mistakenly interpreted as magmatic. Such reservoirs contain neutralised magmatic gases the source of which can be established using Giggenbach's (1992) N_2 -He-Ar diagram, however this indication of source does not indicate the degree of neutralisation and is not a useful predictor of acid conditions at depth.

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