

The Collection and Analysis of Volcanic Gases from Mount Etna [and Discussion]

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Phil. Trans. R. Soc. Lond. A 1973 **274**, 119-128

doi: 10.1098/rsta.1973.0031

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The collection and analysis of volcanic gases from Mount Etna

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A new method of collecting and analysing volcanic gases is described. Analyses of gases collected from Mt Etna during the restricted activity of 1970 and during the eruption of 1971 are presented. It is concluded that the most meaningful way of comparing analyses is in the atomic form as this eliminates secondary equilibration effects. The gases of 1970 and 1971 are distinguished by graphical presentation of the analyses, the main difference being their sulphur content.

INTRODUCTION

The study of volcanic gases has a moderately long history. Workers such as Shepherd (1921) at Hawaii, Allen & Zies (1923) at Katmai, Nemoto, Hayakawa, Takahashi & Oana (1957) at Showa-Shinzan, Basharina (1958) at Kamchatka, have provided a valuable starting-point for systematic and more extensive work. Despite this history of study, relatively little is known of the role of the volatile phase in the volcanic process. On its own, a single analysis has little value in understanding these processes. Ideally the gas collection and analysis should be related to temperature, pressure and amount of both the gas phase and the magma involved, their variation in time, and the overall eruptive state of the volcano. Recent work on Etna (Tazieff, Boulay, Garand, Maulard 1968; Elskens, Tazieff & Tonani 1969; and Chaigneau 1962) has attempted to determine some of these relations. Etna in particular, apart from the recent work mentioned above, has been little studied in respect to the gas phase. The most recent compilation of volcanic gas analyses (White & Waring 1963) contains only three analyses, and the highest recorded temperature of the collected gas is 305 °C, at least 700 °C below the magma temperature. To some extent this lack of data reflects the unique problems of collecting volcanic gases; being at the right spot at the right time and working close to active volcanicity.

The data presented here are results from the collections made on Etna in 1970 during a phase of quiet eruption in the summit region and in 1971 during the extensively reported flank eruption.

METHOD OF COLLECTION

A major problem, both in collection and analysis of gases, is minimizing contamination by air. It is desirable therefore to collect the gases as near as possible to the active lava from which they are emitted. A good guide to the fulfilment of these conditions is that the temperature of the gas should be close to that of the lava. On Etna gas temperatures should be of the order of 1000 °C, otherwise contamination of the sample by air is likely to have occurred.

The apparatus used for the present collections employs a metal tube which is inserted into the gas exit, such that the gases pass along it and through a valve venting to atmosphere. When the tube has reached thermal and chemical equilibrium with the gases (several minutes), the venting valve is closed and the gases admitted via a precision valve to the collecting bottle. The bottle has a capacity of 100 cm³, constructed of Dural, and filled with silica gel to absorb the gases and prevent reaction between the gases and the container. The

bottle and silica gel are degassed before collection at 400 °C and 0.1 Pa (10^{-3} Torr), sealed with a precision valve and vacuum wax. At many gas-exit points on Etna the gas can be seen at night to be burning with a pale blue flame, the tube allows one to collect below this combustion zone.

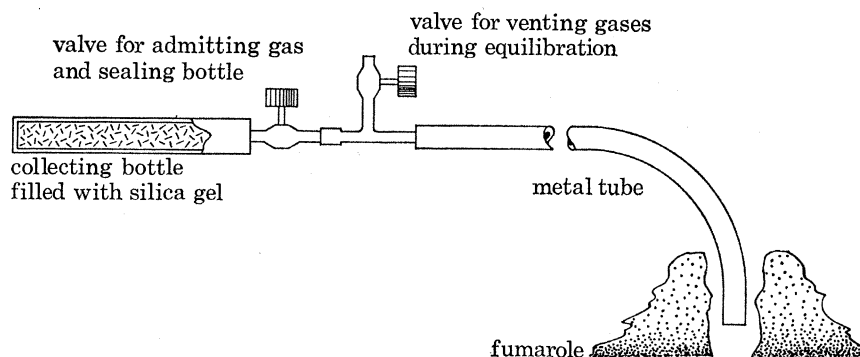


FIGURE 1. Diagram of volcanic gas collecting apparatus.

COLLECTIONS MADE IN 1970

In 1970, Etna was exhibiting its characteristic, mild, restricted activity. Deep explosions in the central crater, explosions at the summit of the northeast cone accompanied by pyroclastics, and an area of lava flows 500 m north of the NE cone (figure 2). Through the crust of these lavas, gases were escaping and had built up small spatter cones or hornitos. The presence of magma below was demonstrated when the crust was penetrated by lava on two occasions during

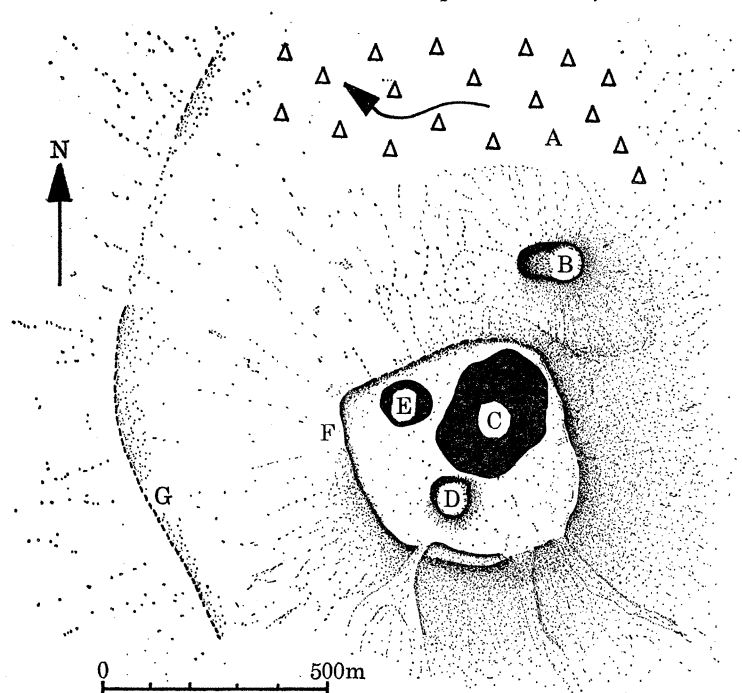


FIGURE 2. Sketch map showing detail of the summit area of Mt Etna in 1970 (based on a map by Dr J. E. Guest). A, the collecting area known as the 'hornitos area', the open triangles show the extent of the active lava; B, summit crater of the NE cone; C, the main crater known as the chasm or 'La Voragine'; D, a small cone built during the eruption of 1964; E, the crater known as the 'Bocca Nuova'; F, summit ring fracture; G, ancient caldera scarp known as 'Cratere Ellitico'.

collection. Collecting conditions at these hornitos (for location see figure 3) were good. The gases were issuing at high temperature, about 1000 °C and with sufficient pressure to throw pieces of lava, weighing about 30 g several metres into the air. The gases could be seen at night to be burning with pale blue flames about 1 m long. Sampling was carried out at three of these

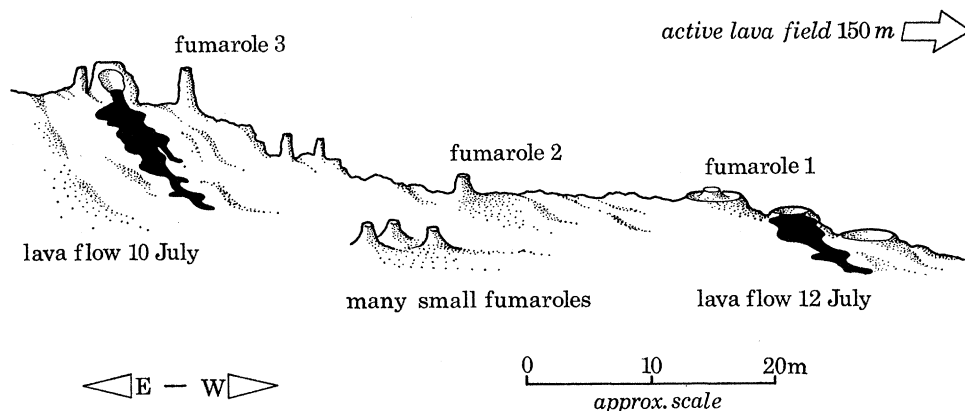


FIGURE 3. Profile view of the hornitos area.

hornitos first to evaluate the collecting apparatus and secondly to examine the variation of the gas composition in space and time. Intervals between collecting were varied from a few seconds to several hours, during which time the activity of the volcano remained substantially the same.

The samples were analysed at Reading on an A.E.I., M.S. 10 mass spectrometer as follows. The sealed bottle was attached to the heated (150 °C) sampling line and a furnace placed around the bottle. The bottle was then heated to 300 °C and the desorbed gases released into the sampling line, and subsequently into the heated (115 °C) tube unit of the mass spectrometer to be

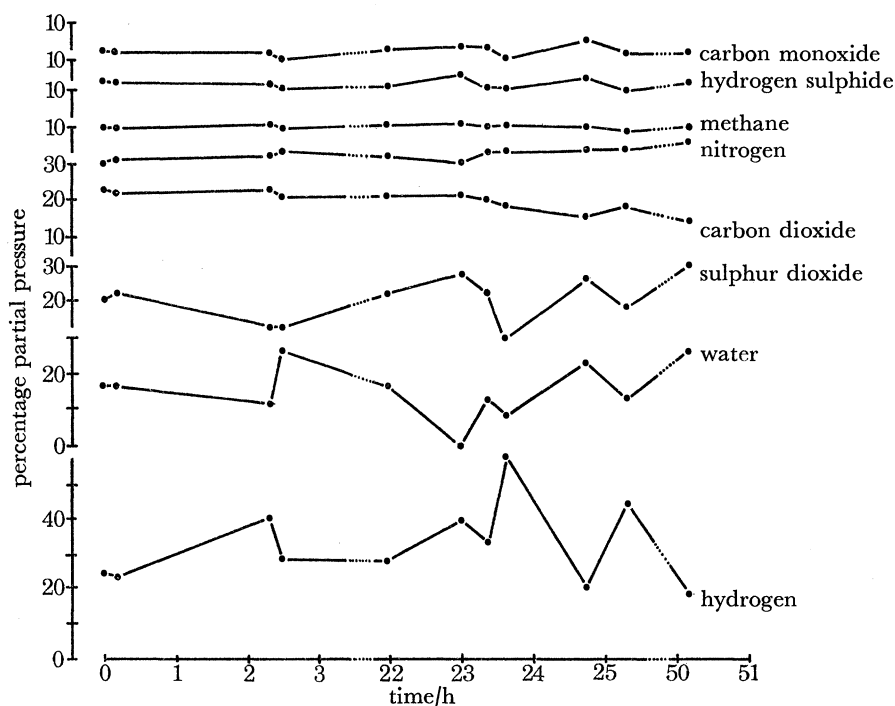


FIGURE 4. Variations in molecular composition, as determined from analysis, of gas emitted from fumarole 2, hornitos area, Etna 1970.

TABLE 1. ANALYSES OF VOLCANIC GAS COLLECTED AT THE HORNITOS AREA, ETNA 1970

Results are expressed in percentage partial pressure at 1.3×10^{-4} Pa (10^{-6} Torr) and 150°C . The temperature of collection was approximately 1000°C in all cases.

analysis number	1		2		3		4		5		6		7		8		9		10		11		12		13		14		15		16		17		
	collection site	—	3	2	2	3	3	3	2	2	2	2	2	1	1	1	1	2	2	2	1	2	2	1	2	1	2	2	2	2	1	1	2		
collection date (July)	—	13	13	13	13	13	13	13	13	13	13	13	13	13	13	13	13	13	13	12	12	12	12	12	14	12	14	12	14	12	12	12			
collection time	—	11.00	13.20	15.30	12.00	12.00	10.40	13.05	11.45	11.30	14.30	12.40	11.32	15.10	06.13	15.05	15.45	13.20	12.50																
hydrogen	19.9	57.8	44.4	34.6	28.5	34.5	39.1	27.3	20.6	25.8	3.8	30.2	18.6	41.9	19.0	45.1	24.6																		
methane	0.4	0.4	—	1.0	1.5	0.7	1.3	1.3	0.4	1.0	1.7	0.6	1.5	1.5	0.9	—	0.7																		
water	27.2	8.9	13.5	9.7	17.2	14.2	—	16.1	23.6	18.2	16.5	28.4	18.8	13.5	26.6	19.3	18.9																		
nitrogen	4.0	3.5	—	8.0	2.6	—	0.2	—	4.1	1.0	9.6	—	7.1	—	—	—	2.0																		
carbon monoxide	1.7	—	1.1	2.9	3.0	3.0	3.5	5.0	3.0	3.0	3.0	0.6	1.8	2.0	2.3	0.4	2.7																		
hydrogen sulphide	1.1	1.5	—	2.3	2.7	1.9	5.1	5.0	4.2	4.3	3.5	2.1	5.1	2.2	3.3	23.8	4.0																		
argon	—	—	1.0	—	—	0.2	—	—	—	—	—	0.3	—	—	—	—	—																		
carbon dioxide	31.4	17.8	19.6	29.4	21.0	21.0	21.4	23.1	14.8	23.9	17.7	22.0	19.5	23.0	15.0	19.4	22.4																		
sulphur dioxide	14.3	10.1	20.4	12.1	23.5	24.5	29.4	26.6	27.3	22.8	44.2	15.8	27.6	15.7	32.8	—	24.7																		

TABLE 2. ANALYSES OF VOLCANIC GAS COLLECTED DURING THE ERUPTION OF 1971

Collection points are indicated in figure 6. Analyses are expressed in terms of percentage partial pressure at 1.3×10^{-3} Pa (10^{-5} Torr) and 115°C .

analysis number	1		2		3		4		5		6		7		8		9		10		11		12		13		14		15		16		17		18		19		20		21		
	collection site	—	3	3	2	3	3	3	3	3	3	3	3	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2			
collection date	—	12	12	12	7	12	12	12	9	12	9	12	7	12	7	12	7	12	7	12	7	12	7	12	7	12	7	12	7	12	7	12	7	12	7	12	7	12	7	12			
collection time	—	May	May	May	May	May	May	May	May	May	May	May	May	May	May	May	May	May	May	May	May	May	May	May	May	May	May	May	May	May	May	May	May	May	May	May	May	May	May	May			
hydrogen	24.3	7.7	9.6	11.9	10.9	12.8	3.8	8.3	6.0	4.8	5.8	0.8	7.0	0.3	1.7	0.7	0.5	0.6	0.2	0.8																							
water	61.4	47.6	65.1	52.8	58.6	38.5	46.0	65.5	38.4	57.9	35.0	81.0	51.5	84.6	77.4	57.5	54.5	54.0	65.8	62.2	66.4																						
nitrogen	—	15.3	—	—	—	—	10.6	—	—	—	13.0	—	17.2	—	2.4	1.9	—	—	—	—	—																						
hydrogen sulphide	—	—	—	—	—	—	1.6	—	31.0	—	—	—	—	0.3	0.3	—	—	—	—	—	—																						
hydrogen chloride	—	—	—	—	—	—	—	—	—	—	—	—	—	—	0.2	—	—	—	—	—	—																						
argon	2.4	—	9.5	1.3	2.4	tr.	—	5.4	tr.	3.9	tr.	0.3	—	0.7	—	3.1	10.4	5.2	1.0	7.4	11.5																						
carbon dioxide	11.9	14.1	13.9	27.2	28.1	41.0	16.4	16.6	22.7	33.4	36.1	17.2	22.2	3.7	12.7	36.1	33.8	38.5	31.3	29.9	19.3																						
sulphur dioxide	tr.	15.3	1.9	6.8	tr.	7.7	21.6	4.2	1.9	tr.	10.1	0.7	2.1	10.4	5.3	0.7	0.8	1.7	1.2	0.4	2.0																						
temperature of gas/ $^\circ\text{C}$	950	950	950	800	950	950	950	950	800	800	800	800	800	800	800	819	900	900	900	819	900	819	900	800	800	800	800	800	950	819	900	900	900	900	900	900	900	819	900	900			

analysed in the usual way. The results shown, table 1, were calibrated against a standard accurately known mixture of gases. (Composition of the standard mixture of gases determined by gas chromatography and Orsat analysis was as follows; argon 10.3 %, carbon dioxide 20.4 %, carbon monoxide 21.8 %, methane 9.90 %, hydrogen 10.1 % and nitrogen 27.5 %. The mixture and analysis supplied by Air Products Ltd. The sensitivity of the sulphur gases and oxygen were determined separately using pure samples.) These samples contained insignificant ($< 1\%$) amounts of air. It can be seen that the same major gases were present at all three fumaroles, however the sulphur dioxide/carbon dioxide ratio was different from fumarole to fumarole, and each fumarole showed a variation in molecular composition with time. This is shown for fumarole two in figure 4.

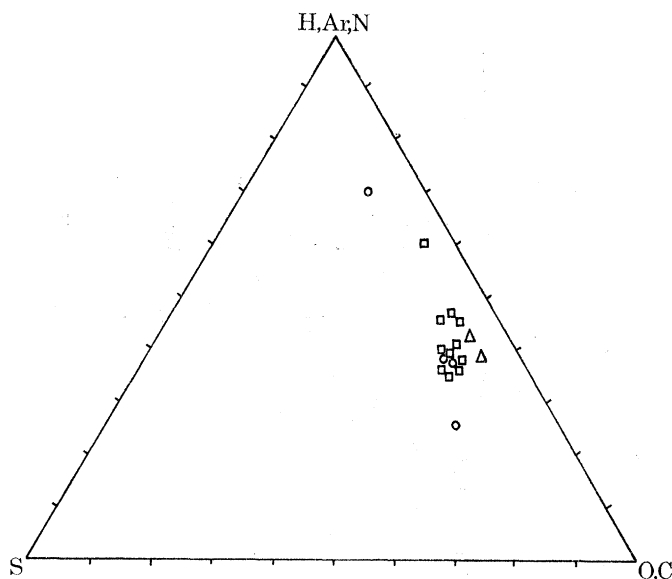


FIGURE 5. Analyses of gases collected from hornitos area 1970 plotted in terms of number of atoms. \circ , Fumarole 1; \square , fumarole 2; \triangle , fumarole 3. (Recalculating to atomic proportions eliminates equilibrium effects. The dominance of the major components, hydrogen and oxygen tends to suppress variations in the minor constituents such as carbon and sulphur when the normalized results are plotted.) The end members are chosen to give maximum separation of the gases collected in 1970 from the gases of 1971, see figure 7. A plot using the more logical end members; [oxygen and sulphur], [carbon, argon and nitrogen], and hydrogen gives a similar result in terms of grouping but not such a good separation.

Variation in concentration of gas molecular species can have three causes; change in equilibrium proportions in response to changing physical conditions, variable contamination (usually from air) and change in the atomic proportions of the gas. The last factor is most likely to be of petrogenetic significance. It should be borne in mind that the gases will tend to re-equilibrate at the lower temperature and pressure in the collecting bottle and analytical instrument and these secondary effects will be superimposed, in the analysis, on primary variations of the molecular proportions.

It could be clearly seen in the field that the pressure was different at the three hornitos and that it varied with time in a cyclic manner, furthermore, although all three fumaroles had temperatures of around $1000\text{ }^{\circ}\text{C}$, variations (maximum about $\pm 30\text{ }^{\circ}\text{C}$) were detected. In order to eliminate these effects the analyses were replotted in terms of their atomic proportions using a modification of the method of Barker (1965). When this is done, as in figure 5, the variations between analyses are reduced. Indeed considering all the possible errors in collection and

analysis the plot suggests that apart from the three apparently aberrant points the gas was the same in all cases. Similar plots using different end members, not shown here, support this conclusion. This series of analyses provided a useful reference point with which to compare analyses of gases collected during the 1971 eruption.

COLLECTIONS MADE IN 1971

During and after the eruption gases were collected as in 1970, except that the tube had a ceramic liner to reduce gas/tube reaction. The collecting sites are indicated in figure 6. Conditions for collection were not as good as in 1970, at active vents the tube frequently became

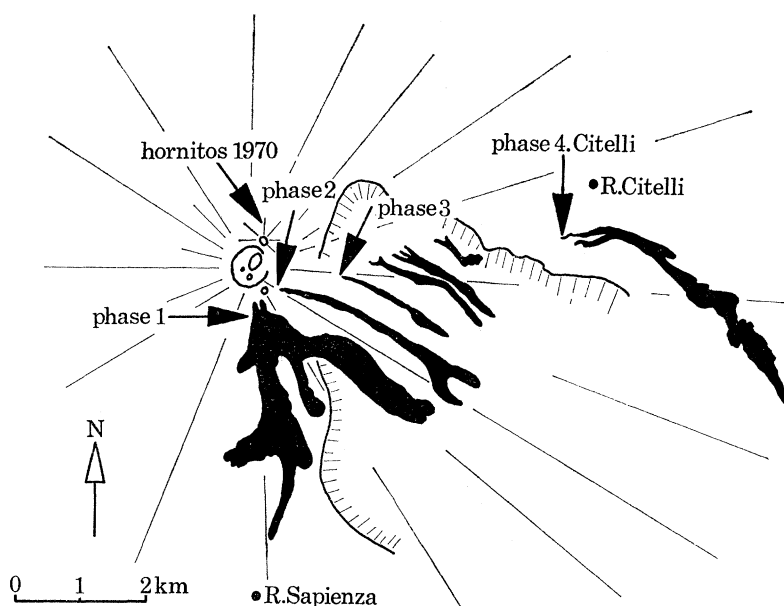


FIGURE 6. Gas collecting points in 1971. Lavas of 1971 are shown in black.

blocked with lava spatter and the vents from which the gases were collected were poorly defined. Gases were collected at the active eruptive centres of phase three, see Huntingdon (1972) for description of phases, and in the weeks that followed the eruption, at 'hot spots' of phases one, two and four. Many of these samples were found to be contaminated with air, incorporated at the time of collection. In the presented analyses the air has been removed by subtracting atmospheric proportions of nitrogen, argon and oxygen (Nordlie 1971). When this correction is applied, traces of nitrogen or argon remain in many samples, suggesting that these gases are minor components of the volcanic gas. The analyses are presented in table 2. When plotted as before, in atomic proportions (figure 7) the gases can be seen to be closely related to the gas emitted during the more quiescent conditions of 1970, but as a group they can be distinguished. The 1971 gases do not form a cluster and the elongation cannot be attributed to time of collection, eruptive centre or altitude. The best separation of the groups can be obtained by plotting atomic hydrogen against sulphur (figure 8), the 1970 samples have the higher sulphur contents, a similar relation between eruptive activity and sulphur content has been reported by Day at Hawaii (in Shepherd 1925).

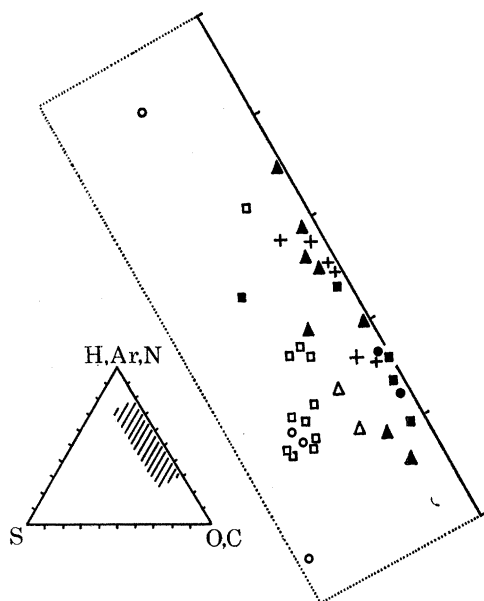


FIGURE 7. Analyses of gases collected in 1970 and 1971 plotted in terms of the percentage number of atoms. The points are plotted on a portion, indicated by hatching, of the triangular diagram shown. \circ , \square and \triangle , fumaroles 1, 2 and 3 (1970); \bullet , $+$, \blacktriangle and \blacksquare , phases 1, 2, 3 and 4 (1971).

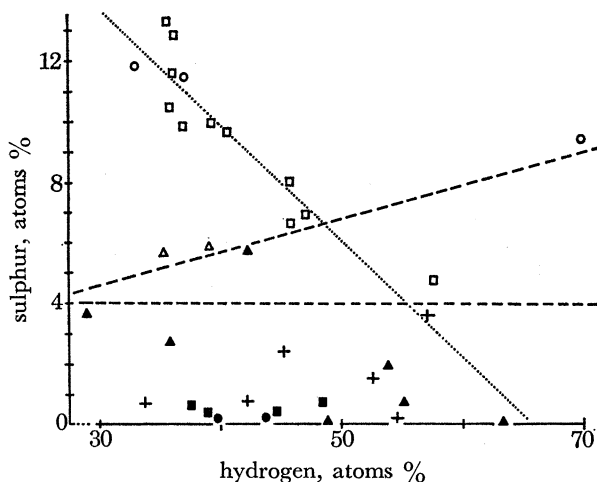


FIGURE 8. Plot of the 1970 and 1971 analyses percentage number of atoms of sulphur against percentage number of atoms of hydrogen. The symbols are the same as figure 7. The area between the dashed lines separates the 1970 gases from the 1971 gases (a 90% separation) and the dotted line is a hypothetical mixing line of sulphur dioxide and water.

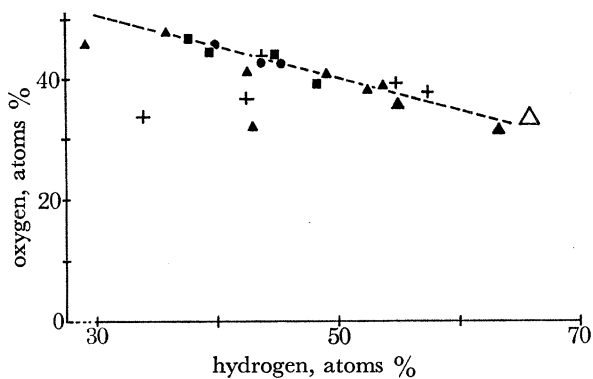


FIGURE 9. A plot of percentage number of atoms of oxygen against percentage number of atoms of hydrogen for the gases collected during the 1971 eruption. The dashed line represents a hypothetical mixing line between water and a constituent with 66% oxygen. \triangle , water; \bullet , $+$, \blacktriangle and \blacksquare , phases 1, 2, 3 and 4.

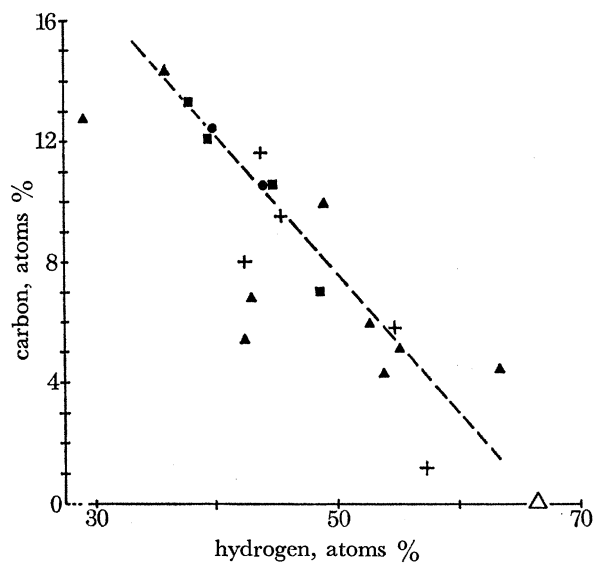


FIGURE 10. A plot of percentage number of carbon atoms against percentage number of hydrogen atoms for the gases collected during the 1971 eruption. The dashed line represents a hypothetical mixing line between a constituent with 66% hydrogen and another with 33% carbon. The symbols are the same as figure 9.

A plot of atomic hydrogen against atomic oxygen, of the 1971 gases (figure 9) suggests that the gases lie on a mixing line between water and a constituent with 66 % oxygen. A further plot of atomic hydrogen against atomic carbon (figure 10) suggests that the gases lie on a mixing line between water (from figure 9) and a constituent with 33 % carbon. Comparison with the molecular analyses shows that this second constituent is carbon dioxide. This mixing accounts for 80 to 90 % of the gas composition.

In the absence of isotopic data it is not fruitful to speculate at length on the origin of these Etna gases. Many authors believe that at least in part volcanic gases are derived from the mantle and play a major role (Bailey 1970) in initiating deep melting and magma formation. Water is generally accepted as a trace constituent of the mantle and the work of Roedder (1965) suggests an equally ubiquitous distribution of carbon dioxide; if this is the case the above results are compatible with the gases originating in such a mantle.

CONCLUSIONS

Genuine variations of volcanic gas compositions can be determined only from their atomic proportions.

Although large variations of molecular composition take place with small time periods, real variations may have time constants similar to those of major eruptive activity, i.e. weeks or years. The apparent variations of the molecular composition are more a measure of high-temperature gas kinetics than fundamental chemical composition.

If the Etna gases return to the sulphur levels of 1970 without further lateral outbreaks of activity a useful relation between gas composition and volcanic activity may be established for this volcano. Subsequent monitoring of gas composition may thus provide a valuable indicator of changes in the eruptive condition of the system.

I am indebted to the International Volcanological Team organized by Professor Tazieff with members from the Centre National de Recherche Scientifique, Paris, Commission de L'Energie Atomique, Saclay and the Instituto Internazionale di Vulcanologia, Catania, without whose help and support in the field this work would not have been possible, in particular Mr F. Le Guern with whom I have worked closely in the field. Support for travelling expenses has also been received from the Department of Geology, University of Reading, and the Royal Society, to whom I would like to express my thanks. The Department of Chemistry, University of Reading, kindly provided analytical facilities. Mr D. Hutchings and Mr S. Powell of this department helped with the design and manufacture of the collecting apparatus; Dr D. K. Bailey kindly read the manuscript and I have incorporated many of his suggested alterations.

The work was conducted during the tenure of a University of Reading Research Studentship.

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Discussion

DR M. SATO: The analyses showed very high hydrogen concentration. Has the author tried to calculate the oxygen fugacity values using Naughton, Heald & Barnes (1963) or Nordlie's method to see if the hydrogen was produced by the reaction of the sampled gas with the metallic container?

A. T. HUNTINGDON: The brief answer to Dr Sato's question is that the oxygen fugacity of the collected gases has not been calculated. However, I can perhaps indirectly throw some light on the presence of hydrogen in the analyses.

The average molecular hydrogen concentration in the 1970 analysed gases is 28.1 % and in the 1971 gases 5.0 % (considerably higher values have been reported from different volcanoes as will be mentioned later). If the averages expressed in atomic form are compared with the much quoted average of the Hawaiian gases (collected by Shepherd (1925)) expressed in atomic form by plotting them on a similar diagram to figure 7 in the text, the Hawaiian gas plots within the elongate cluster of the 1970 analyses, suggesting that they will have similar oxygen partial pressures. Nordlie (1971) has calculated the oxygen partial pressures of the Hawaiian gases and they fall in the magnetite buffer zone. Downes (1973, this volume) has indicated from his experimental work that the Etna lavas crystallized in an atmosphere with a partial pressure of oxygen compatible with the presence of magnetite in the lavas. Hence it is considered that the presented analyses are capable of having the oxygen partial pressure suggested by the lavas, particularly when the effects of temperature and pressure on the homogeneous equilibrium (and the oxygen partial pressure) are considered.

The possibility that the hydrogen indicated by the molecular analyses has been generated by collection or post collection reactions has been investigated in the following ways:

(1) The reaction between the collecting tube and the volcanic gas. The reaction would be oxidation of the metal tube, producing a metal oxide and hydrogen, by steam contained in the volcanic gas. At temperatures of 1000 °C the production of a 'thick' oxide layer (10 nm) would take place within 1 or 2 s. Once this layer is formed the direct reaction would be inhibited and would proceed by the 'parabolic law mechanism', governed by the diffusion rates of the reacting gas and metal through the oxide layer, providing it was mechanically stable. The equilibrating period referred to in the description of the sampling method would ensure that if any reaction took place it would be by the latter mechanism. From known diffusion coefficients, the area of reaction (the internal surface of the tube) and the estimated velocity of the gas within the tube the maximum level of hydrogen produced by this reaction can be calculated. This proves inadequate by several orders of magnitude to supply the levels of hydrogen indicated by the analyses.

(2) Reaction between the gases and the Dural collecting bottle and/or silica gel. This is a less likely source of the hydrogen as the gases are rapidly quenched within the bottle. In 1970 several bottles were used without silica gel and when opened for inspection, after collection and analysis, were seen to be corroded by the gases. The analysis of the reacted gases however showed

no increase in hydrogen levels, the agents of corrosion were the sulphur gases which had almost completely reacted and were in low concentrations in the residual gas. No such corrosion was found in the bottles containing silica gel (all the presented analyses were of gases contained in bottles with silica gel). Silica gel was introduced into the bottles, specifically to prevent reaction between the gases and the bottle and interaction between the gases, as suggested by Naughton *et al.* (1963). To test whether the silica gel itself, or by reaction with water, produced hydrogen, bottles with silica gel and silica gel and water were analysed using the described heating and analysing procedure, no hydrogen was detected.

Finally, gases were extracted, by a cool crushing process, from air and water quenched samples of lava collected at the same location as the 1970 gases. Analysis of the released gases gave similar results to those of the analysed fumarolic gases, i.e. similar levels of molecular gas species including hydrogen. These lava samples and extracted gases had never been in contact with the collecting apparatus.

These brief tests suggest that the sampling and analysing procedures did not introduce or produce hydrogen. There remains the possibility that hydrogen in the quantities quoted was present in the volcanic gases.

In 1970, at the hornitos area, the gases could be seen at night to be burning with pale blue flames. The combustible gas could be either hydrogen or carbon monoxide (the temperature was too high for methane to be stable). The carbon monoxide content, 2 %, is insufficient to support this combustion. During the eruption of 1971 Tazieff (1971) reported chemical explosions at the phase one cones, explosive behaviour is more characteristic of hydrogen than carbon monoxide. Rittmann (1972) has witnessed explosions at Stromboli which he believes to be due to the presence of hydrogen gas, indeed Chaigneau (1965) has reported levels of more than 30 % of hydrogen in the Stromboli gases.

Hydrogen gas in substantial concentrations has been reported (in White & Waring 1963) by several authors; Lindal at Hekla, Allen at Vesuvius, Grange at White Island, Basharina at Zavaritskii, Iwasaki at O-Shima and Oana at Showa Shinzan. It would seem that hydrogen is present in the gases of many volcanoes, particularly in those gases collected at high temperatures.

The presence of hydrogen in volcanic gases has important implications. For example, volcanic explosions are often attributed to a rapid release of gases trapped in a sealed volcanic throat, perhaps some of these explosions are chemical in origin, i.e. rapid combustion (explosion) of hydrogen. The explosion of Bandai San in 1888 (Sekiya & Kikuchi 1889) appeared to take place without the production of lava, perhaps this was a chemical explosion.

In conclusion the oxygen fugacities of the gases collected from Etna in 1970 and 1971 are comparable with those of Hawaii, i.e. within the magnetite buffer zone, that hydrogen is present in the 1970 and 1971 Etna volcanic gases in proportions indicated by the analyses and that hydrogen may have a major role in the volcanic process.

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