RENDICONTI Società Italiana di Mineralogia e Petrologia, 38 (1): pp. 405-412
Comunicazione presentata al Congresso SIMP di Cagliari il 16 ottobre 1981

PETROGENETIC SIGNIFICANCE OF F AND Cl DISTRIBUTION IN VOLCANIC ROCKS FROM THE AEOLIAN ISLAND ARC

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ABSTRACT. — The content in volatiles, mainly F and Cl, in magmatic rocks appeared recently of a certain interest as petrogenetic indicator and also in the study of eruptive mechanisms.

The values here obtained for lavas from the Aeolian arc (Sicily) point out a significant increase of F in shoshonitic with respect to calcalkaline rocks, for a given SiO_2 concentration; an increase in F with both K and SiO_2 is also observed.

Cl levels are instead very scattered although still a noticeably higher abundance is shown by shoshonitic series.

The relationships of F versus SiO_2, K and Li, allow to point out the residual character of this volatile, whose abundance in the more primitive among the studied rocks could depend on the degree of melting which produced the parent magmas.

The large scattering of Cl values appears an evidence for the occurrence of an additional mechanism possibly consisting in an input to the system from an external source, whose extent seems to overshadow the effect due to petrogenetic processes.

RIASSUNTO. — Il contenuto in volatili, soprattutto Cl e F, nelle rocce magmatiche è apparso recentemente di un certo interesse quale indicatore petrogenetico, ed anche nello studio di meccanismi eruttivi. I risultati qui ottenuti per lave provenienti dall'arco Eoliano (Sicilia) indicano un aumento significativo di F in rocce shoshonitiche rispetto a rocce calcalcaline, a parità di contenuto di SiO_2; si osserva anche un aumento di F all'aumentare di K e SiO_2. I valori di Cl sono invece molto dispersi, anche se la serie shoshonitica mostra ancora contenuti più elevati.

La correlazione di F con SiO_2, K e Li, consente di individuare il carattere residuale di questo volatile, la cui abbondanza nelle rocce più primitive fra quelle studiate potrebbe dipendere dal grado di fusione parziale che ha originato il magma.

La grande dispersione di valori per Cl appare possibilmente dovuta alla presenza di un meccanismo ulteriore, consistente in un contributo esterno al sistema, la cui entità sembra sovrapporsi all'effetto di processi petrogenetici.

Introduction

The behaviour of F and Cl during differentiation of magmas has been only rarely considered in recent times up to the last years, probably because an early review concluded that no correlation was observed between the abundances of those elements and the chemical compositions of the rocks (CORRENS, 1956).

The possibility of replacing hydroxyl groups in hydrous minerals has subsequently resulted as an important factor in determining the distribution of both halogens (GREENLAND and LOVERING, 1966; PAUL et al., 1975; NASH, 1976) as well as their affinity for silicate melts and possibility of forming gaseous phases (FUGE, 1977; MURAMATSU and WEDEPOHL, 1979; ROWE and SCHILLING, 1979; HARRIS, 1981).

The role of partial melting and fractional crystallization have been taken into account to explain the actual concentration in different series of rocks (KANISAWA, 1979; ISHIKAWA et al., 1980; SCHILLING et al., 1980; SMITH et al., 1980; AOKI et al., 1981) and the possibility of external contributions has been also considered (BAILEY and MACDONALD, 1975; MUENOW et al., 1979).

Since most of the above mentioned investigations have been carried out on basaltic and alkaline series, a study was undertaken on the rocks pertaining to the Aeolian island arc, of calcalkaline and shoshonitic affinity, to evaluate the characters of F and Cl distribution during advanced stages of magmatic evolution.

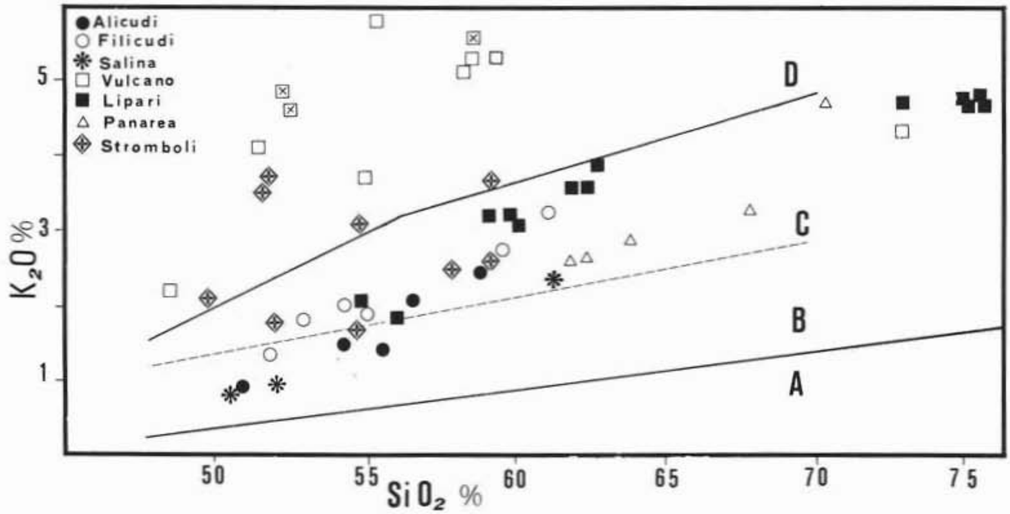

Fig. 1. — Relationship of K_2O versus SiO_2. Crossed open squares refer to lc-tephritic rocks from Vulcano. - *A*: island arc tholeiitic series; *B*: calcalkaline series; *C*: high-potassium calcalkaline series; *D*: shoshonitic series.

These investigations, along with those previously carried out (KLUGER et al., 1975; KIESL et al., 1978; CORADOSSI and MARTINI, 1981) can provide a first contribution on the use of these elements in the interpretation of petrogenetic processes.

Volcanological and petrological outlines

The Aeolian volcanic arc is one of the main structures in the Mediterranean area and consists of seven islands, including both extinct and active volcanoes. The age of the emerged part of the arc has been found to be younger than 1.0 m.y. (BARBERI et al., 1974), whereas slightly older rocks have been dragged from the Aeolian sea bottom (1.3 m.y., BECCALUVA et al., 1980).

The rock types outcropping along the Aeolian arc range in composition from basic to acidic, and have been emplaced in two stages of activity. The first stage was responsible for the formation of the islands of Panarea, Alicudi, Filicudi and parts of Salina and Lipari, while Stromboli and Vulcano together with the most recent parts of Salina and Lipari were formed during the second.

From a petrological point of view the rocks from the Aeolian arc belong essentially to the calcalkaline and shoshonitic series, although a high-K andesitic series and a lc-tephritic series have been recognized on the basis of K_2O and K_2O/Na_2O versus SiO_2 relationships (BARBERI et al., 1974). The calcalkaline rocks range in composition between basalt and dacite with the single rhyolitic outcrop of Basiluzzo. The high-K calcalkaline suite mainly consists of intermediate terms whereas shoshonitic rocks display a composition ranging from shoshonitic basalt, latite to trachyte. Recently, significant geochemical evidence has been collected which indicates that the rhyolites from Lipari and Vulcano represent the terms of the shoshonitic suite (BARBERI et al., in preparation). The lc-tephritic series is made up of a few lc-tephrites and trachytes outcropping at Vulcano and Stromboli (KELLER, 1980; ROSI, 1980).

The calcalkaline lavas were essentially erupted during the first stage of the arc evolution whereas shoshonitic and lc-tephritic suites characterize the second stage of Aeolian volcanism. Except for the obsidians from Lipari and Vulcano, all the Aeolian rocks have a porphyritic texture with plagioclase and clinopyroxene as main phenocryst phases.

Orthopyroxene is commonly found in basic and intermediate calcalkaline rocks, whereas hornblende and biotite only appear in the most evolved terms. In the shoshonitic

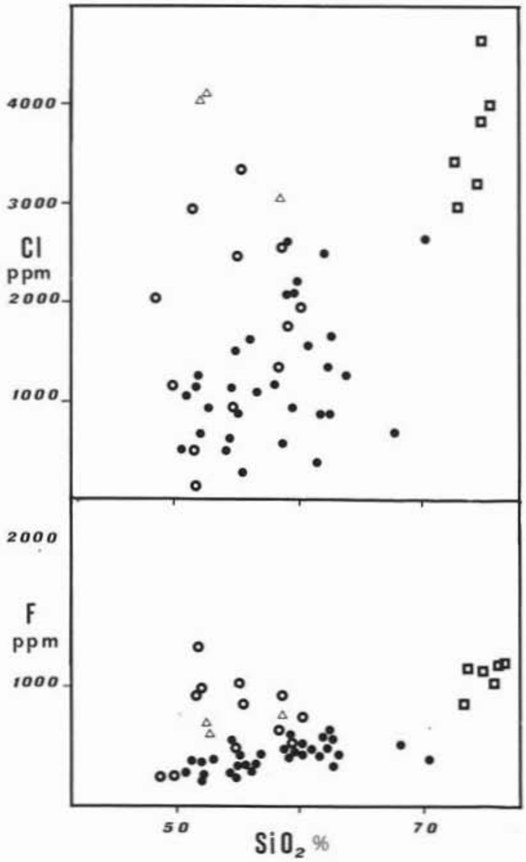

Fig. 2. — Relationship of Cl and F versus SiO_2. - Full circles: calcalkaline series; open circles: shoshonitic series; triangles: lc-tephrites; squares: rhyolites.

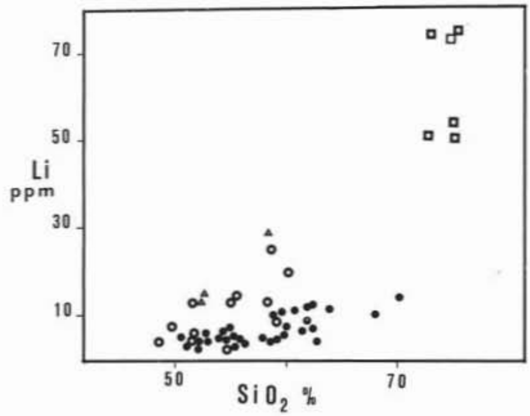

Fig. 3. — Relationship of Li versus SiO_2. - Symbols as in fig. 2.

and lc-tephritic volcanics K-feldspar appears as phenocryst phase in the intermediate terms and olivine is common in basic terms. Microphenocrysts of leucite are confined to the lc-tephritic suite.

Results

The F, Cl and Li values obtained for the analyzed samples are reported in table 1, together with the SiO_2 and K_2O abundances, taken from several authors.

Fig. 1 shows the K_2O versus SiO_2 relationship for the analyzed samples. According to the classification proposed by Peccerillo and Taylor (1976) for the volcanic rocks from orogenic areas, the considered samples plot in two main fields, calcalkaline and shoshonitic, characterized by distinct potassium enrichment. Among the shoshonitic rocks three samples belonging to the lc-tephritic series as recognized by Barberi et al. (1974) have been kept distinguished in this and in the following diagrams.

In fig. 2 *a* the F versus SiO_2 relationship is reported. The calcalkaline rocks display lower F abundances as well as spread values with respect to shoshonitic rocks, mirroring the behaviour of potassium.

Within the calcalkaline suite the F contents increase with SiO_2 up to values of silica around 63 %, displaying a tendency to decrease in the most evolved terms, which appears more clear if only the rocks coming from Panarea are considered (table 1). For the shoshonitic suite F increases with fractionation up to the rhyolitic terms.

Fig. 2 *b* is a plot of Cl versus SiO_2. Essentially, a positive correlation can be noticed for the shoshonitic suite, but with large spread of values. The shoshonitic rocks have an overall higher Cl content with respect to calcalkaline rocks, but with a large compositional overlap, while the lc-tephritic rocks are the most Cl-rich among the analyzed samples.

From the Li versus SiO_2 relationship reported in fig. 3, it can be noticed that Li abundances increase with SiO_2 in both calcalkaline and shoshonitic series, although with a steeper slope in the latter group of rocks. This evidences the residual character

TABLE 1
Analytical results for the studied samples

sample			SiO_2	K_2O	F	Cl	Li	rock type
Alicudi	Al	2	54.25	1.52	290	485	4	basaltic andesite
	Al	10	56.50	2.08	365	1080	3	high-K andesite
	Al	13	50.97	0.90	395	1040	3	basalt
	Al	18	58.59	2.45	500	550	4	high-K andesite
	Al	23	55.43	1.41	355	245	3	basaltic andesite
Filicudi	Fi	1	60.87	3.06	495	1565	11	high-K andesite
	Fi	5	59.45	2.78	535	900	11	high-K andesite
	Fi	8	52.83	1.86	400	930	6	high-K bas. andesite
	Fi	16	51.82	1.38	380	1120	5	basalt
	Fi	30	54.35	2.02	555	595	6	high-K bas. andesite
	Fi	40	55.00	1.90	355	900	7	high-K bas. andesite
Salina	Sa	76	61.40	2.40	430	345	6	high-K andesite
	Sa	113	50.50	0.80	290	480	5	basalt
	Sa	210	52.00	0.90	230	680	3	basalt
Lipari	IL	2	75.23	4.88	1210	4700	54	rhyolite
	IL	3	59.04	3.21	605	2580	10	high-K andesite
	IL	4	72.98	4.68	880	2970	51	rhyolite
	IL	5	75.16	4.95	1050	3680	51	rhyolite
	IL	12	62.62	3.87	375	1650	4	high-K andesite
	IL	24	60.04	3.13	535	2170	7	high-K andesite
	IL	25	62.32	3.61	590	1325	7	high-K andesite
	IL	26	62.00	3.58	500	2500	12	high-K andesite
	IL	31	59.80	3.20	485	2090	6	high-K andesite
	IL	34	56.06	1.90	305	1650	4	high-K andesite
	IL	35	54.94	2.09	460	1500	5	high-K bas. andesite
	IL	40	75.03	4.91	1165	3120	73	rhyolite
	IL	42	75.43	4.86	1250	3850	75	rhyolite
Vulcano	V	40	58.60	5.60	910	2570	25	latite
	V	129	51.50	4.10	950	2930	13	shoshonitic basalt
	V	147	48.50	2.20	280	2030	5	shoshonitic basalt
	V	154	55.40	5.80	860	3340	14	shoshonite
	VL	11	55.00	3.70	1035	2440	13	shoshonite
	IV	2	58.80	5.18	650	1315	13	latite
	IV	21	59.60	5.43	770	1970	20	latite
	IV	25	72.60	4.36	1210	3340	74	rhyolite
	VO	6	58.50	5.30	750	3045	29	latite
	VO	18	52.50	4.60	605	4050	15	shoshonite
	VO	24	52.35	4.85	700	4130	13	shoshonite
Panarea	PRX	1	63.92	2.81	430	1240	11	high-K andesite
	PRX	2	67.89	3.24	540	645	10	high-K andesite
	PRX	4	70.25	4.73	405	2695	14	rhyolite
	PRX	5	62.34	2.64	640	860	12	high-K andesite
	PRX	6	61.76	2.63	575	865	9	high-K andesite
Stromboli	B	12	59.01	2.60	420	2090	5	high-K andesite
	B	34	57.89	2.51	455	1150	5	high-K andesite
	B	42	54.58	1.71	245	1130	5	high-K bas. andesite
	C	13	59.18	3.66	535	1750	9	latite
	D	12	54.68	3.12	490	915	4	shoshonite
	D	10	52.00	1.80	285	1240	4	basalt
	B	20	51.61	3.56	1310	490	5	shoshonite basalt
	D	1	51.70	3.76	975	150	6	shoshonite basalt
	St	75	49.77	2.15	270	1140	7	shoshonite basalt

Values for SiO_2 and K_2O, expressed in percent, are from several authors; F, Cl and Li are given in ppm. Atomic absorption spectrophotometry has been used for Li determination, and selective-ion electrode potentiometry for F and Cl.

TABLE 2
Description of samples

Alicudi	Al 02	Dirituso	Lava flow
	Al 10	Filo dell'Arpa	Lava dome
	Al 13	Galera	Lava flow
	Al 18	Montagna	Lava dome
	Al 23	Galera	Dike
Filicudi	Fi 1	Capo Graziano	Lava dome
	Fi 5	Montagnola	Lava dome
	Fi 8	Zucco Grande	Lava flow
	Fi 16	Fossa delle Felci	Lava flow
	Fi 30	Monte Terrione	Lava flow
Salina	Sa 76	Paolonoci	Lava flow
	Sa 113	Erbe Bianche	Lava flow
	Sa 210	Serro del Capo	Bomb
Lipari	IL 2	Forgia Vecchia	Obsidian
	IL 3	Monte S.Angelo	Lava flow
	IL 4	Giardina	Lava flow
	IL 5	Capistello	Lava flow
	IL 12	Monte S.Angelo	Lava flow
	IL 24	Costa d'Agosto	Lava flow
	IL 25	Monte S.Angelo	Lava flow
	IL 26	Timpone Carrubbo	Lava flow
	IL 31	Costa d'Agosto	Lava flow
	IL 34	Monterosa	Lava flow
	IL 35	Monterosa	Lava flow
	IL 40	Pilato	Obsidian
	IL 42	Vallone Bianco	Obsidian
Vulcano	V 40	Palizzi	Lava flow
	V 129	Lentia	Dike
	V 147	La Sommata	Bomb
	V 154	Punta Luccia	Lava flow
	VL 11	Lentia	Lava flow
	IV 2	Porto	Lava flow
	IV 21	Punte Nere	Lava flow
	IV 25	Pietre Cotte	Obsidian
	VO 6	Vulcanello	Lava flow
	VO 18	Vulcanello	Lava flow
	VO 24	Vulcanello	Lava flow
Panarea	PRX 1	Punta Falcone	Lava dome
	PRX 2	La Fossa	Lava flow
	PRX 4	Basiluzzo	Lava dome
	PRX 5	La Loca	Lava flow
	PRX 6	Western coast	Lava flow
Stromboli	B 12	Petrazza	Lava flow
	B 34	Serro Monaco	Lava flow
	B 42	Serro Monaco	Lava flow
	C 13	Upper Vancori	Lava flow
	D 12	Middle Vancori	Lava flow
	D 10	NE sector along coast	Lava flow
	B 20	Sciara del Fuoco	Lava flow
	D 1	Sciara del Fuoco	Lava flow
	St 75	Sciara del Fuoco	Lava flow

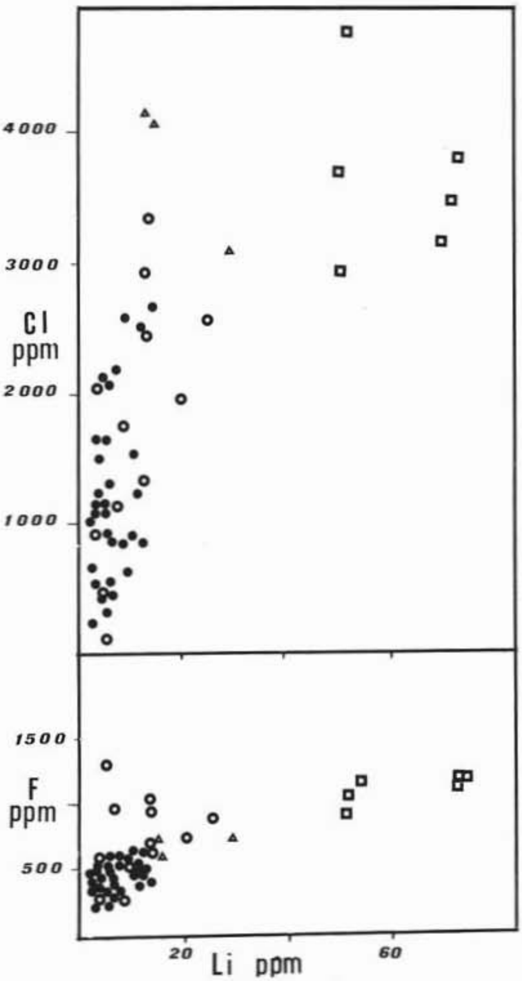

Fig. 4. — Relationship of Cl and F versus Li. - Symbols as in fig. 2.

for this element which, accordingly, can be used as an index of differentiation.

Based on the relationship of Cl and F against Li, it can be better evidenced that the increase of F and Cl is stronger in the early and middle stages of differentiation, whereas in the late stages the abundances of the two elements tend to keep constant against strong increase of Li content (fig. 4 *a, b*). This relationship can be interpreted as an evidence that both calcalkaline and shoshonitic magmas behave as close systems for Li but not for F and Cl in the late stages of differentiation.

In fig. 5 F and Cl against K_2O relationships are shown. A good positive correlation between potassium and the two halogens is evident, in accordance with the similar relation observed by AOKI et al. (1981) for worldwide distributed volcanic rocks.

Finally, the Cl versus F relationship is shown in fig. 6, where a positive correlation between the two elements is apparent.

Discussion

The behaviour of both F and Cl during the magmatic processes has recently received some attention, and much of the interest arises from the possibility of evaluating through their variations the role of some mineralogical phases (e.g. apatite, biotite, hornblende) during petrogenetic processes

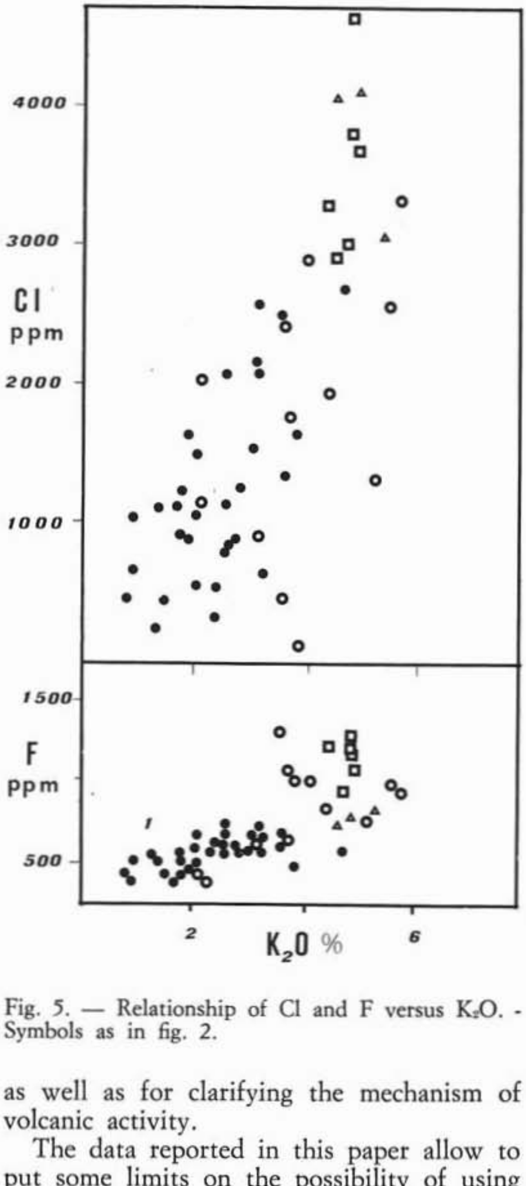

Fig. 5. — Relationship of Cl and F versus K_2O. - Symbols as in fig. 2.

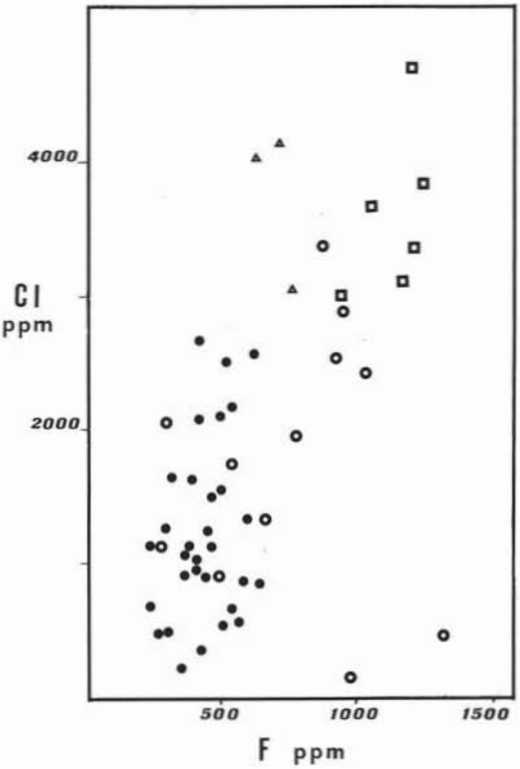

Fig. 6. — Relationship between Cl and F. - Symbols as in fig. 2.

as well as for clarifying the mechanism of volcanic activity.

The data reported in this paper allow to put some limits on the possibility of using F and Cl variations in recognizing and modelling some magmatic processes as crystal-liquid fractionation or degree of partial melting.

Fluorine

The good positive correlation observed for F with Li and K_2O, as well as the increase of F abundances with increasing silica contents in both calcalkaline and shoshonitic series, at least in the basic-intermediate range, indicates that fluorine behaves as a residual element. In addition, the narrow range of F values shown by calcalkaline suite at any silica content suggests that during the evolution no important loss or gain of this element occurred in the magma which, accordingly, behaves as a closed system for F, at least in the early-medium stages of differentiation. The decrease of F in the acidic rocks from Panarea can be interpreted either as due to loss of F as volatile phase or to its incorporation into a crystallizing F-rich phase. The first possibility is suggested by equilibria of the type:

$$2\,MeF + SiO_2 + H_2O \leftrightarrows Me_2SiO_3 + 2\,HF$$

where Me mainly represents Na or K, according to which a decrease in solubility of F follows the increase in silica concentration (Kogarko et al., 1961; 1968).

The separation of a F-rich mineral is also a suitable mechanism for explaining the F depletion in the acidic rocks since rather abundant hornblende phenocrysts occur in the rocks from Panarea (ROMANO, 1973). The presently available data do not allow to discriminate between the two hypotheses.

Apatite and biotite fractionation instead is not expected to have played an important role, because accessory amounts only of the minerals can be found in these rocks.

Within the shoshonitic series, F values are more scattered for a given silica content, a feature which is also shown by potassium.

F contents still increase with silica and, unlike the calcalkaline series, this trend continues in the acidic range, although with a flatter slope.

This different behaviour can be accounted for by the increase in solubility of F in shoshonitic magma with respect to the calcalkaline one due to its higher concentration in alkali metals, whose positive role appears evident from the above mentioned equilibrium.

However, an alternative possibility is that hornblende is not present in the shoshonitic rocks so that the depletion of fluorine by crystal-liquid fractionation processes is inhibited.

Chlorine

The positive correlation between Cl and F evidences the common geochemical character of the two elements. The increase of Cl with increasing K_2O and Li also points to a residual character for Cl. The large scattering of its values, however, in both calcalkaline and shoshonitic suites at any degree of silica enrichment clearly indicates that important losses or gains of Cl as volatile phase occurred in magma which, accordingly, behaved as an open system for Cl at any stage of evolution. Such conclusion precludes the possibility of using Cl abundances for petrogenetic interpretation at least in the studied suites.

Conclusions

The F, Cl and Li abundances determined in a large number of volcanic rocks from the Aeolian arc have shown some characteristics which can help in understanding petrogenetic processes.

Because of its defined residual character, Li appears to represent for the studied area a good differentiation index; few data only, however, are available about the distribution of this element in other magmatic suites and any comparison on a general basis is not possible yet.

The good positive trends displayed by F against K_2O as well as the small spread of values shown by this element expecially in the calcalkaline suite, seem to exclude important losses or gains of F as volatile phase and support the idea that the F variations can be used for qualitative petrogenetic interpretation at least in the basic and intermediate ranges.

The close correlation between K_2O and F for worldwide distributed volcanics found by AOKI et al. (1981) as well as the distinct F enrichment observed in volcanics belonging to the potassium series and high-potassium series from Mts. Ernici (CIVETTA et al., 1981; MARTINI, unpublished data) strongly support this conclusion.

Instead, the behaviour of F during the late stages of magmatic differentiation still needs investigation and evaluation.

Also worth of study is the possibility of using F abundances for quantitative or even semiquantitative modelling, once a sufficient quantity of data have been collected on the abundances of F for mineral-groundmass pair in several volcanic rocks.

The Cl variations, although similar to those of F, are characterized by large scattering at any degree of evolution, and this has been interpreted as evidence for losses and gains of Cl as volatile phase in magmas. These processes have overshadowed the Cl variations connected with the main petrogenetic processes and preclude the possibility of using this element for petrogenetic interpretation, at least in the considered suites.

Acknowledgement. — The samples from Alicudi, Filicudi and Panarea have been kindly provided by L. VILLARI, those from Stromboli by M. ROSI, and those from Salina, Lipari and Vulcano by the Institute of Mineralogy and Petrography of the University of Pisa.

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