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Geochemistry of Fumarolic Deposition of Carbonatite Volcanism in Tamil Nadu

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Abstract: *Carbonatitic volcanic activities are wide spread in several parts of Tamil Nadu. Relicts of weathering products of fumarolic deposits of 4-7 mm dimension as outline films on some calcites in limestone and feldspars in granite pegmatites and thin films of encrustations on net works of cooling cracks in ash flows are observed. Most lava occurs as very small rootless diatremes might have been formed by rapid diffusions of fluids ascending from deep seated ultra low viscosity carbonatitic magma at high PTX through fissures. Air fall lapilli, pisolites and ashes and ash flows of carbonatite lava are studied. SEM images and geochemical chemical compositions show that fumarolic deposits are superficial and subjected to leaching leading to relics of siliceous, ferruginous, carbonaceous and oxidation products. Trends of chemical leaching are just opposite to trends of magmatic evolution of carbonatite lava. In places, the lava is associated with trachyte and tephrite. The fumarolic deposits appear to be younger than carbonatite volcanism manifested in Tamil Nadu from Late-Cenozoic to Recent Period. The enrichments of sodium carbonates over potassium carbonates reveal that the carbonatite volcanism is genetically related to natrocarbonatite eruptions. Spherical and ellipsoidal lapilli from ash flows of Kudangulam and Sattangulam, feeder dykes along ring fractures in south of Sivamalai alkali-complex, discoid lapilli north of Thiruvalangadu from Pleistocene Boulder Bed and thin plates of lapillus along Dharangambadi beach respectively indicate high, medium and low viscosities of carbonatite melts. Release of different quantities of fugitive fluid constituents might have been caused sublimation and deposition of fumarolic materials. .*

Keywords: *Carbonatite volcanism, Fumarolic deposits, Rootless diatremes, Late Cenozoic-Recent, Tamil Nadu.*

1. INTRODUCTION

Carbonatites are relatively rare magmatic carbonate rocks. They occur as intrusive or extrusive forms. Active natrocarbonatite lava eruptions and their pyroclastic materials from Ol Doinyo Lengai, Tanzania are extensively studied in recent years [1-3] to prove magmatic nature of carbonatites. Carbonatite magma derives from mantle source [4]. Temperatures of carbonatite melt ranges between 1000-1500^{o}C [4, 5]. Below these temperatures fumarolic deposits in neighbourhood of some craters or fissures are formed. The wide spread occurrence of carbonatite tuff including air fall tephra of pyroclastic boulders, lapilli, pisolites and ashes in different parts of Tamil Nadu opens a new Era of stratigraphical horizon ranging age between Mio-Pliocene to Recent [6-10] A sketch map showing locations and trend of ash flow tuff is already published [10]. Geological maps for Sivamalai carbonatite lava emplaced along ring fractures [9] and air fall tephra varying from boulder, lapilli, pisolites ashes occurring on calcareous gritty sandstone in Thiruvalangadu [7] and tephrite and carbonatite lava floats occurring along the beach of Dharangambadi are given separately (8).

2. FIELD STUDY

In Sydapuram area near Gudur Town Andhra Pradesh, fumarolic deposit is observed on feldspars in Sivaparvathi pegmatite mine. Nearby a kankar appears to be carbonatite lava occurs 1 m below ground level for a thickness of 1m in Siddhi Vinayaka pegmatite mine. Feldspars in Lakshimi Narayana mine are partially transformed into calcites. Occurrence of well studied Vinjamur eruptive carbonatite lava [11] is not far away from Nellore-Gudur Schist Belt. Fumarolic deposit is seen just 500 m from the east of Pandalgudi Ramco Cement Ltd limestone Mine Officers' Quarters. It is found on a pink coloured calcite for 4-7mm dot and specks of black coloured outline films. About 10 km east of the mine pit at Pandalgudi, carbonatite ash tuff is found near Maravarperungudi village. Net-work of thin films of fumarolic encrustations is found in this ash tuff. Mine cut exposures reveal.circular and semi-circular exposures showing very dark coloured pitchy like coatings along peripheral portions. Aggregates of coalesced pisolites are coated with thin films of dark coloured materials. Brown coloured pisolites aggregates coated with iron oxides are seen at the top of Kudangulam hillock. Ash tuff is coated with dark coloured fumarolic materials in several parts of ash tuff occurring in different places. Dark black coloured coatings are found on crystalline limestone occurring 3 km NW of Eppodumvendran village and 2 km SE of Singikulam village.

A lapillus of porphyritic carbonatite lava is collected on a traverse between Pungavarnattam and Podupatti 5km NW of Eppodumvendran village, Tamil Nadu. In hand specimen it has blades of interpenetration twins of 3x0.5 cm crosscutting another blade of 2.5x0.5 cm amidst very fine-grained calcite matrix. The thickness of blades is less than 1 mm. The rock is extremely silica under saturated and has normative foidal minerals (Table. 1),

Fumarolic deposits and net-work of encrustations Eppodumvendran and Singikulam villages are studied. Peripheral portions are coated with brown coloured materials. About 15 km NE of Eppodumvendran village carbonatite ash tuff is present 1 m below the black cotton soil at Duraiyur village. Near Singikulam, small exposures of carbonatite ash tuff are seen SE of the crystalline limestone between the distances of 5 and 15 km distances trending in NW-SE directions. Ash flow tuff is coated with black coloured materials along joint and weak planes.

Fig1: Between Tisaiyanvilai and Sattangulam (15x5km), an over-turned structural basin is seen.

Fold axial plane of the Tisaiyanvilai-Sattangulam basin dips 60^{o} NW and plunges 45^{o} towards N30^{o}E and its NE termination is displaced and subsided (Fig, 1). The structure has been formed in the Pre-existing Precambrian granite gneisses showing doubly plunging (Fig. 1) overturned fold system which axially faulted and forming a basin structure. It is filled with carbonatite ash flow over which younger terri-sands are filled. Carbonatite tuff occurs below the terri sands over a thickness of 8m in this basin indicating its down-warping nature in this basin and other similar basins. They represent from Kudangulam, Uvari, Tisaiyanvilai-Sattangulam to Kudiraimolzi terri all along the Eastern Coast in this area.

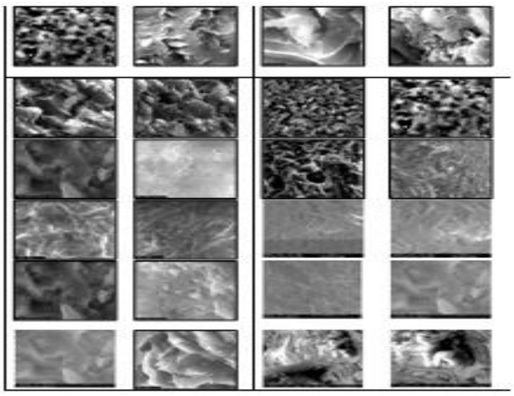

Fig2: Textural and mineralogical compositions lapilli of Dharangambadi (1-4); Thiruvalangadu (5-8); Kudangulam (9-12); Sivamalai (13-16); Podupatti porphyric carbonatite (17-20) and Mylampatti carbonatite lava (21-24). They indicate volcanic feature under higher magnifications calcite glasses have crystalline shapes. They exhibit volcanic texture

Fig3: View of carbonatite ash tuff with conchoidal fractures and laminations brought from neighbourhood well excavation in Kudiraimolzi Teri for construction of Mulaithenda Ayyanar temple: Photograph is made by Arun Kannan, Google.co.in.com.

3. REVIEW OF CARBONATITE VOLCANISM IN TAMIL NADU

Carbonatite lapillus is identified by presence of spherulitic texture in thin section. Matrix is composed of very fine-grained calcite. In fine-gtained matrix relatively larger phenocrysts are grown near approach of outer periphery of vesicle or micro-fracture towards central cavity. Under higher magnification of matrix shows crystalline plate-lets of calcites of 1 to 5μm sizes. EDAX analyses show that some cavities are partially filled with tabular crystals of pyrochlore, baddeleyite, barite and topaz only at peripheral portion. The petrographical features of lapilli from different parts of Tamil Nadu show magmatic texture and mineral compositions (Fig. 2.). In several places ash tuff is composed of pyroclastic lapilli. Lapillus is enriched with volatile concentrations of F, Cl, P_2O_5, and SO_3. High concentrations of elements like Hf, Ta, Zr, Nb, Y, Pb, Ni, Co and LREE < HREE are found in degassed lapilli. It has high ratios of Sr/Ba indicating its deep seated origin [5]. Pink coloured pyroclastic materials are younger to black coloured materials. Lapilli and pisolites are air fall pyroclastic materials present in ash flow tuff. Grey and pink coloured carbonatite lapilli are collected in the year 1983 from Kudangulam but only after finding these pyroclastic materials varying from boulder, lapilli, pisolites and ashes in a Pleistocene boulder bed of calcareous gritty sandstone carbonatite volcanism is conformed in the year 1998 during the course of clay mineral investigation in this area [7]. Association with soda-trachyte [12], olivine tephrite and carbonate-tephrite [13] carbonatite dykes [14] and pocket of phlogopite and apatite [15] in Kudangulam area, further supports volcanic activity in this area. Wide spread basaltic rocks are seen in Kudangulam, Sanganeri, Thiruvambalapuram and Kanakkankulam near Kuttam village [12, 13]. Fluidization of country rocks of granite gneisses into kaolinite near Tisaiyanvilai releases rounded pure quartz and kaolinite. A large deposit of Kaolinite occurs in Idinthakarai and Vijayapathi [16] for a depth up to 10m near Kudangulam. White sands and kaolinite are found in Muttam. Red coloured kaolinite occurs in Surankudi village. Fumarolic deposits of gypsum is seen 2 km NE Thalikattupuram village near Vilathikulam Town in association with rootless ferrocarbonatite diatremes (<50x10 cm). Carbonatite lava diatremes are found 2km north of Mylampatti in association with carbonate tephrite. The tephrite has notable trace elements of Mo and molybdenite mineralization is present in limestone quarry nearby located 2 km north of Palayam and 5 km NE in crystalline limestone mine at Dolipatti village. Many individual discontinuous diatremes of carbonatite lava trending ENE-WSW direction are found near Alambadi limestone mine area [17].

Globular lapilli occur in ash flows of Kudangulam and Sattangulam [10]. The lapilli present in Thiruvalangadu area are discoid in form and pisolites and ashes are relatively plate like in form. They are air fall tephra and their shapes are attained by air thrown semi-solid hot fluid at some heights from crater, falling down to the surface. Along Dharangambadi beach very flat (10x7x0.5 cm) grey and pink coloured lapilli are collected and a carbonate tephrite is also collected at Arasalar River mouth in Karaikal Town of Pondicherry State along the border of Tamil Nadu. Very flatness of pyroclastic material thrown from a submarine crater indicates that the lava might have been under low viscous state. From this fact, a submarine volcano on 20th January 1757 might have been erupted and thrown pyroclastic of carbonatitic and basaltic materials [18, 19] is to be reassessed.

Dark grey carbonatite lava occurs in ring fractures 5 km south of Sivamalai alkali complex, Tamil Nadu [9]. A rootless basaltic eruption took place on 26th May, 1996 [20 -22] in Sirukinaru hamlet near Uthiyur village. Extensive deposits of kankar for a maximum depth of 4 m are seen in this area and it relatively appears to be a carbonatite ash tuff in NE and SW portion of this complex but no lapilli are seen on this ash tuff. Barite is a notable accessory mineral in the grey coloured carbonatite lava which is emplaced along Vattapparai ring fractures in Sivamalai area. Gypsum is mined in SE portion and it might have been formed as fumarolic deposit in this place. Rapid effusions of volcanic lava flows of silicate perovskite derived from deep mantle source occur in this area and several other parts in Tamil Nadu are reported [22]

Voluminous ash flow tuff carrying pisolites and lapilli is observed in Kudangulam and Sattangulam. In places, Coalesced pisolites occur as concretions but without any nucleus at cores. Aggregates of more than two globular pisolites appear to be formed single lapillus. In places, the ash tuff is very fine-grained, homogeneous and exhibits lamination. Broken surfaces of ash tuff show conchoidal fractures and it is used as local construction material (Fig.2).

Similar ash flows are found below the dune sands exposed from Kudangulam, Uvari, Tisaiyanvilai, Sattangulam in to Kudiraimolzi Terri in the north. The tuff discontinuously expose just above granite gneisses along an arc like fracture >100 km N-S width between Uttumalai and Vijyanarayanam including exposures in Shamugarengapuram Radhapuram and Kudangulam. The ash tuff extends further to Surendai, Sendamaram. Mahedravadi and is in the vicinity of Sankarankoil Town. A large quantity of ash flow is found in the east Eluvamugi, Pidaneri, Pannaikulam and Payankulam. Again, in the east, the ash flow extends 1 m below black cotton soil in Kayattar, Parutthivilai, Duraiyur, Muthulapuram, Nagalapuram and Maravarperungudi [10].

Near Hogenekal, ash tuff is found in Chinnaru River valley and about 6 km N of northern bank of Cauvery River. Sovite intrusives are reported in this area [23]. In Walaiyar crystalline limestone mine carbonatite lava associated with kaolinite [16]. The ash tuff deposit is described as tufaceous limestone [24] by earlier workers [24-28]. Mio-Pliocene shells and gastropods which are earlier ones seem to be adhered and included in the tufaceous limestone which is younger to the age of these fossils along coastal tracts. Narayanaswamy [25, 26] by observing presence of abundant carbonate lapilli as inclusions in the tufaceous limestone occurring between Pannamparai and Sattangulam, he classified this part of tufaceous limestone as a separate stratigraphical unit as Pannamparai Formation [25, 26], Detailed petrographical and field studies, such inclusions occurring in southern parts of Tamil Nadu show that they are carbonatite pisolites and lapilli that prove that the tufaceous limestone is a volcanic ash flow tuff spread in several hundreds km^2. Many of them are found on either side of river valleys which are deeply fractured. The flow is restricted to a depth of <4 m due to surface erosion.

4. METHODOLOGY

Using High Resolution Scanning Electron Microscope attached with Energy Diffusive X-ray Micro Analyses (EDAX) probe in the Department of Material Science and Metallurgical Engineering, Indian Institute of Technology Madras, Chennai, 600036. The (EDAX) were made in different parts of a scarped fumarolic material of 7 mm dimension with thickness of 0.2mm on calcite from a crystalline limestone occurring between Eppodumvendran and Podupatti villages. Similar studies were made on the fumarolic samples collected 2 km SE of Singikulam village. With limited facilities available and bulk and spot EDAX analyses of individual minerals were detected. The analyses were recalculated on the basis of their oxides. Rittmann's normative mineral proportions were calculated [29] and listed in the Table 1-7. Geochemical interpretations are made by using major and trace elements present in the rock by constructing suitable variation diagrams.

5. PETROGRAPHY AND GEOCHEMISTRY

SEM images show that fumarolic materials have large volume of empty spaces for free growth of large mineral grains. Sizes range around 1μm diameter is globular and rounded in shape but they are crystalline nature. Most of larger grains are prisms, laths and needles, Notable sizes ranges between 1.5x3.0 and 4x0.5 μm. Both tabular and needle like crystals are seen. The sample exhibits homogeneous hypidiomorphic granular texture. Free growth of crystals are seen adjacent to the boundaries of cavities and cracks.

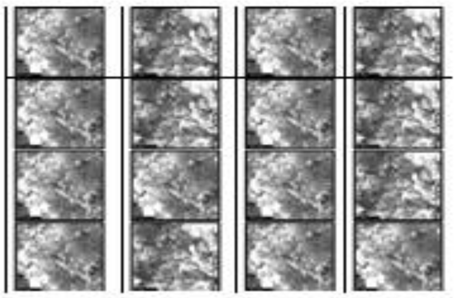

Fig4: SEM images and EDAX analyses of fumarolic materials deposited on calcites present in crystalline limestone occurring between Eppodumvendran and Podupatti villages,

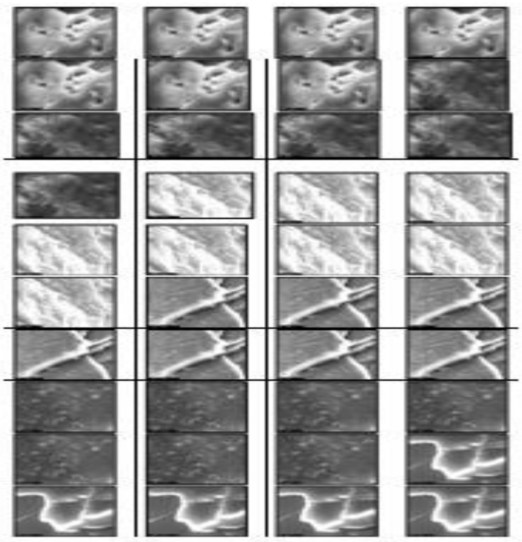

Fig5: EDAX images of fumarolic material in Singikulam village

The porphyritic carbonatite lava (Table 1) from Podupatti village is composed of large amount of calcite, portlandite and volatiles. The carbonatite lava is silica undersaturated and depending upon degassing of volatiles from lava, it is silica saturated under volatile enriched state. Silica undersaturated lava with excessive CO_2 alkali carbonate is formed. Silica undersaturated carbonatite lava produces foidal minerals during cooling under water free anhydrous state. Portlandite forms in Ca-rich hydrous carbonatites.

Scanning electron images show a massive fumarolic material has number of linear cavities along crystal boundaries. Sizes of cavities are ranging from >0.5μm to <5μm in length and width around 0.5 μm. In places tabular crystals of 6x3 μm are seen. Linear cavities up to 6x1.5 μm are seen. Crack of 7x 01 μm is developed narrowing at one end amidst a massive material. Plates of massive minerals (16x8 μm) overlap one over other. Rhombohedral plates of 6x1.5 μm are seen. Crack of 7x 01μm is developed narrowing at one end amidst a massive matrix. Plates (16x8 μm) of minerals overlap one over other. Rhombohedral plates of 50x50μm are seen. A fine-grained needle of 50x0.5 μm is seen in a massive fine-grained holohyaline matrix. Very small crystals >2 μm are also found in the matrix. Prisms of 20x5μm are seen in some places. The texture of fumarolic material varies from holocrystalline to holohyaline. Fumarolic materials from Eppodumvendran are massive and more altered than Singikulam materials.

Table1: EDAX analyses of Podupatti Porphyritic carbonatite lapillus from Podupatti village. Trace elements are in parts per million (ppm).

	115	116	117
SiO2	24.26	4.83	2.54
Al2O3	7.83	2.00	1.39
FeO	0.91	0.84	1.02
MgO	3.17	11.77	3.46
CaO	28.49	48.17	57.23
Na2O	0.63	0.32	0.87
K2O	8.16	1.33	0.99
TiO2	0.45	0.26	0.43
ZrO2	1.87	2.13	3.71
Nb2O5	0.00	0.00	0.00
Y2O3	0.66	1.88	2.10
F	0.62	0.00	0.74
P2O5	1.21	1.13	1.48
SO3	1.71	1.62	1.72
Cl	0.50	0.27	0.60
CO2	19.53	23.45	21.72
SUM	100	100	100
	115	116	117
zr	1.57	1.76	3.18
anh	2.20	2.07	2.33
xeno	0.63	1.76	2.01
hl	1.47	0.83	1.80
fl	2.62		3.13
ap	1.52		0.27
il	0.63	0.31	0.53
mt	0.05	0.05	0.05
lar		3.88	
cc	46.57	55.05	52.39
port		28.76	32.40
cpx	18.51		
an		2.33	
ne		0.31	
ks	24.23	2.79	1.91
	100	99.9	100

Ba	350	0	220
Sr	700	440	1110
Pb	250	0	160
V	250	30	200
Co	530	170	430
Ni	530	570	690
Hf	810	220	890
Ta	830	220	950
La	970	170	720
Ce	570	360	480
Nd	660	220	640
Eu	290	50	220
Dy	550	460	530
Yb	430	110	370
Lu	210	0	130

Table2: shows chemical compositions of fumarolic materials on calcites of limestone between Eppodumvendran and Podupatti

	1	2	3	4	5	6	7	8
SiO2	34.76	27.20	37.25	48.31	23.20	12.64	57.21	24.86
Al2O3	6.36	6.21	7.61	8.18	7.03	2.24	4.12	6.52
FeO	7.13	3.76	1.73	3.24	3.55	2.72	1.46	31.23
MgO	0.55	0.90	0.60	0.60	0.84	0.29	0.44	0.47
CaO	10.99	24.81	17.57	11.58	13.05	10.30	2.56	4.98
Na2O	0.07	0.15	0.21	0.00	0.29	0.00	0.15	0.00
K2O	2.11	1.39	1.82	4.95	1.67	0.77	1.27	1.83
TiO2	0.54	0.22	0.34	0.77	0.59	0.05	0.18	0.63
P2O5	0.10	0.19	0.15	0.23	0.00	0.00	0.21	0.54
F	0.32	0.31	0.22	0.38	0.76	0.20	0.16	0.62
Cl	0.06	0.06	0.18	0.32	0.03	0.07	0.20	0.26
SO3	0.16	0.21	0.44	0.25	0.31	4.34	0.54	0.38
CO2	31.98	27.46	22.16	12.13	43.79	59.89	25.91	14.41
Sc2O3	0.23	0.35	0.67	0.55	0.35	0.23	0.37	0.60
Cr2O3	0.10	0.21	0.35	0.58	0.23	0.21	0.25	0.40
CoO	0.21	0.20	0.31	0.46	0.37	0.30	0.35	0.34
NiO	0.08	0.25	0.28	0.25	0.21	0.31	0.20	0.14
V2O5	0.13	0.23	0.45	0.73	0.23	0.17	0.40	0.36
Rb2O	0.29	0.64	0.94	0.98	0.59	0.47	0.82	0.93
SrO	0.48	0.50	0.97	1.45	0.42	0.91	0.65	1.57
Y2O3	0.94	1.56	1.70	1.32	0.57	1.05	0.73	2.73
ZrO2	1.11	1.46	2.49	2.16	0.71	1.47	0.87	2.52
Nb2O5	1.33	1.72	1.57	0.60	1.21	1.39	0.96	3.68
	1	2	3	4	5	6	7	8
zr	1.50	2.03	2.94	2.37	1.14	1.73	1.23	4.30
caf2	1.40	1.35	0.90	1.78	1.56	0.91	0.78	3.16
anh	0.21	0.34	0.68	0.36	0.42	5.48	0.78	0.63
ap	0.16	0.45	0.28	0.47				
il	0.75	0.34	0.45	1.18	0.73	0.10	0.22	1.01
mt	0.59	0.34	0.11	0.24	0.26	0.20	0.11	3.23
nacl	0.21	0.23	0.57				0.67	
kcl								0.89
na2c			0.17		0.73			
k2c	3.65	2.70	3.34	9.35	2.70	1.22	2.18	3.04
rb2c	0.16	0.23	0.40	0.47	0.26	0.15	0.34	0.51
cc	20.20	48.56	34.84	23.92	22.15	13.39	3.91	10.37
mgc	1.50	2.48	1.70	1.78	2.18	0.70	1.23	1.52
fec	8.70	4.85	2.04	0.36	3.85	3.35	1.79	27.07
al2c3	16.82	10.59	13.24		17.94	5.58	11.34	
sic	24.83				39.00	19.58	40.79	
sc2c3					0.16	0.25	0.45	
cr2c3					0.16	0.41	0.45	
coc					0.26	0.41	0.56	
nic					0.26	0.41	0.34	
vc					0.05	0.10	0.22	
y2c3					0.16	0.41	0.45	
hy								20.49
sil			4.75	14.21				12.14
qz	18.65	24.51	32.01	41.68			32.18	9.74
sc2o3	0.11	0.17	0.28	0.24				0.25
cr2o3	0.05	0.17	0.28	0.47				0.32
coo	0.16	0.17	0.23	0.36				0.32
nio	0.05	0.06	0.23	0.18				0.13
v2o5	0.05	0.06	0.11	0.24				0.13
y2o3	0.21	0.39	0.45	0.36				0.76
co2					6.14	45.61		

Table3: EDAX Chemical composition Fumarolic material on crystalline limestone Podupatti-Eppodumvendran

[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]

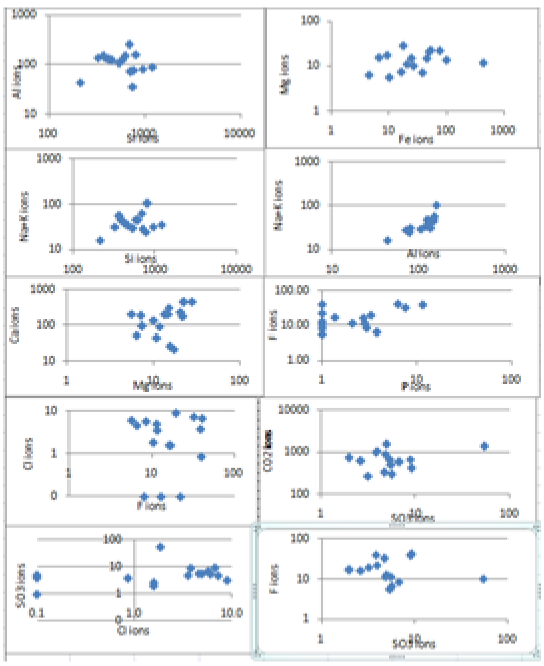

Fig4a: Chemical compositions of fumarolic material deposited on calcite in crystalline limestone occurring between Eppodumvendran and Podupatti

Table4: EDAX compositions of Singikulam fumarolic materials are shown.

	1	2	3	4	5	6	7	8	9	10
SiO2	90.06	73.28	75.41	88.43	73.17	84.40	76.51	8.84	8.65	8.76
Al2O3	1.28	1.14	1.15	1.68	1.08	1.32	1.00	0.47	0.54	0.54
FeO	4.60	8.16	5.43	4.87	8.11	7.59	8.24	87.96	88.92	88.27
MgO	0.14	0.29	0.29	0.45	0.40	0.29	0.13	0.30	0.36	0.35
CaO	0.27	0.06	0.31	0.16	0.18	0.11	0.21	0.23	0.20	0.23
Na2O	-	0.26	0.26	0.00	0.26	0.15	0.20	0.00	0.00	0.00
K2O	0.14	0.07	0.03	0.07	0.00	0.00	0.04	0.05	0.01	0.02
TiO2	-	0.00	0.13	0.00	0.11	0.00	0.10	0.02	0.00	0.04
P2O5	0.50	0.13	0.34	0.00	0.00	0.00	0.18	0.02	0.02	0.03
F	0.53	0.79	0.00	0.17	0.55	0.57	0.21	0.18	0.15	0.10
CO2	-	12.90	13.25	0.00	11.91	2.75	10.92	0.86	0.35	0.83
SO3	0.39	0.23	0.42	0.00	0.15	0.00	0.00	0.05	0.06	0.10
Sc2O3	0.15	0.00	0.23	0.10	0.12	0.00	0.11	0.06	0.05	0.06
CoO	0.06	0.14	0.21	0.06	0.26	0.05	0.23	0.05	0.02	0.04
NiO	0.07	0.09	0.18	0.12	0.22	0.10	0.19	0.04	0.04	0.04
SrO	-	0.55	0.61	0.89	0.73	0.89	0.36	0.15	0.18	0.09
Y2O3	-	0.63	0.40	0.90	0.66	0.45	0.63	0.19	0.11	0.17
ZrO2	1.81	0.44	1.34	0.81	1.13	0.72	0.75	0.25	0.15	0.17
Nb2O5	-	0.85	0.00	1.27	0.96	0.61	0.00	0.28	0.17	0.15
zr	1.82	0.81	1.29	1.48	1.54	0.97	0.71	0.42	0.28	0.28
anh	0.61	2.26	0.59					0.14	0.14	0.14
caf2	1.58			0.55	1.6	1.75	0.65	0.63	0.56	0.35
ap	1.15	0.29	0.76				0.47			
il			0.23		0.12		0.12			0.14
mt	0.06	0.81	0.59	0.55	0.89	0.85	0.88	10.34	10.45	10.34
cor										
na2co3		1.39	0.70		0.83	0.48	0.35			
k2co3		0	0.12				0.12	0.14		
cc		-				0.12	0.35		0.28	0.28
mgc					1.06	0.85	0.35	0.99	0.84	1.27
fec		12.54	7.51		11.47	6.65	11.08	1.55		1.13
al2c3		3.19	3.40		3.13		2.95			
sic	5.28	15.21	20.77		13.59		12.15			
scy2c3		0.23	0.47		0.65		0.47			
conico3		0.35	0.58		0.70		0.70			
cpx										
hy				8.14		3.74		19.56	19.07	19.55
sil	2.00			1.29		2.36		0.99	1.19	0.77
or	0.91			0.31						
ab										
an				2.77						
qz	86.39	62.74	62.62	84.40	64.42	81.87	68.63			
scy2o3	0.06			0.31		0.24			0.14	
conio	0.07			0.09		0.12			0.14	0.14

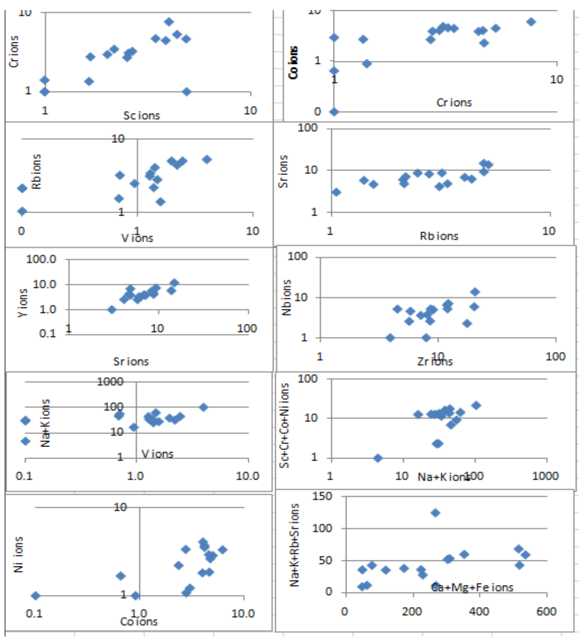

Fig4b: Chemical compositions of fumarolic material deposited on calcite in crystalline limestone occurring between Eppodumvendran and Podupatti

6. DISCUSSION

Fumaroles are fissures through which volcanic gases escape into the Earth's surface and sublimated as fumarolic products [30]. Such deposits are highly soluble in meteoric water. At present only relics are remain. Identification of fumarolic minerals is rather complex and needs sophisticated instrumentation techniques and knowledge to assume original ones.

Table5: EDAX compositions of Singikulam fumarolic materials are shown.

	11	12	13	14	15	16	17	18	19	20
SiO2	8.64	8.68	8.83	20.45	19.27	19.24	19.25	18.04	19.62	19.88
Al2O3	0.48	0.56	0.49	1.03	0.91	1.05	1.02	0.84	0.85	1.06
FeO	89.19	88.08	87.89	53.08	53.96	55.81	55.23	54.82	56.16	55.98
MgO	0.31	0.42	0.38	9.16	8.49	8.47	8.52	8.01	8.44	8.89
CaO	0.19	0.21	0.20	2.53	2.28	2.51	2.38	2.28	2.46	2.46
Na2O	0.00	-	0.02	0.26	0.11	0.17	0.20	0.26	0.16	0.22
K2O	0.04	0.02	0.02	0.03	0.06	0.06	0.08	0.12	0.12	0.08
TiO2	0.03	0.06	0.02	0.00	0.00	0.00	0.07	0.01	0.09	0.09
P2O5	0.03	0.06	0.04	0.06	0.10	0.06	0.09	0.00	0.00	0.05
F	0.13	0.14	0.16	0.26	0.08	0.00	0.27	0.20	0.06	0.00
CO2	0.00	0.78	1.05	11.37	12.59	11.15	11.26	13.80	9.96	9.70
SO3	0.02	0.09	0.08	0.20	0.19	0.15	0.21	0.16	0.19	0.15
Sc2O3	0.04	0.05	0.05	0.05	0.13	0.03	0.09	0.08	0.12	0.13
CoO	0.07	0.05	0.05	0.05	0.06	0.11	0.08	0.05	0.08	0.09
NiO	0.04	0.03	0.04	0.05	0.06	0.07	0.08	0.06	0.10	0.07
SrO	0.12	0.17	0.12	0.41	0.23	0.32	0.28	0.29	0.38	0.24
Y2O3	0.19	0.22	0.18	0.26	0.36	0.21	0.19	0.39	0.45	0.30
ZrO2	0.19	0.23	0.23	0.42	0.63	0.19	0.40	0.30	0.36	0.43
Nb2O5	0.28	0.15	0.14	0.32	0.49	0.42	0.31	0.30	0.40	0.19
zr	0.43	0.42	0.43	0.48	0.85	0.48	0.49	0.36	0.50	0.49
anh		0.14	0.14	0.24	0.24	0.24	0.37	0.24	0.25	0.24
caf2	0.43	0.56	0.58	0.72	0.30		0.86	0.54	0.19	
ap		0.21	0.22	0.18	0.18	0.18	0.18			0.18
il		0.14							0.12	0.12
mt	10.62	10.37	10.63	5.35	5.45	5.64	5.65	5.54	5.82	5.68
cor										
na2co3				0.72	0.36	0.48	0.55	0.72	0.50	0.67
k2co3				0.12	0.22	0.12	0.18	0.18	0.31	0.18
cc		0.14		4.93	4.48	5.33	4.30	4.70	5.57	5.13
mgc		1.41		25.50	25.56	25.21	22.62	23.96	19.69	21.12
fec		0.99			4.24		0.25	8.43		
al2c3										
sic										
scy2c3										
conico3										
cpx										
hy	18.96	20.04	20.31	39.45	37.13	37.39	38.12	34.92	39.07	38.82
sil	0.43		1.08	1.80	1.64	1.94	1.84	1.57	1.61	1.95
or										
ab										
an										
qz										
scy2o3	0.07	0.14	0.07	0.12	0.12	0.06	0.25	0.12	0.25	0.12
conio	0.14	0.14	0.14	0.12	0.06	0.12	0.25	0.12	0.12	0.12
cao										
mgo										
feo	68.85	65.42	66.38	20.26	19.26	22.79	24.15	18.60	26.07	25.15
k2o										

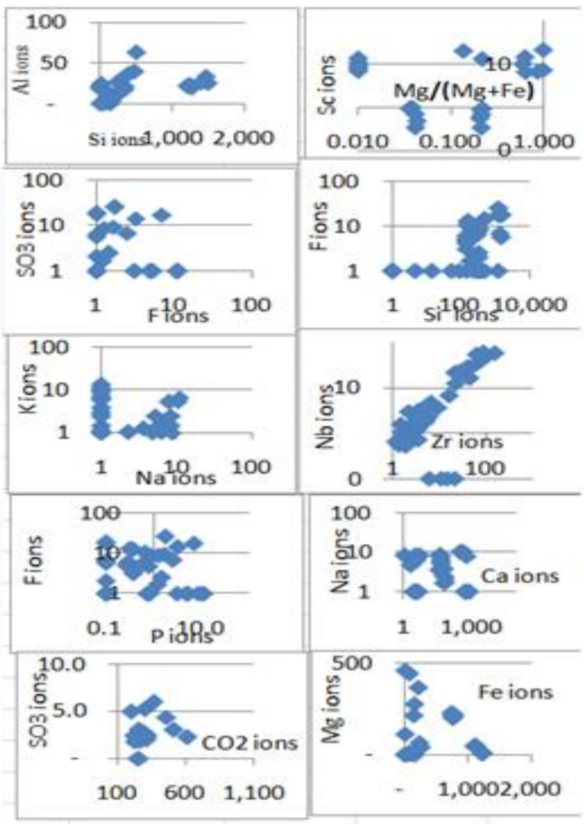

Fig5: shows bi-variation diagrams for fumarolic material occurring in calcite from crystalline limestone Singikulam. There are 4 groups formed by leaching.

Table6: EDAX compositions of Singikulam fumarolic materials are shown.

	21	22	23	24	25	26	27	28	29	30
SiO2	18.86	9.65	10.14	10.01	9.66	9.73	23.83	8.31	3.16	14.78
Al2O3	0.93	0.31	0.37	0.20	0.33	0.28	1.87	0.00	0.00	1.36
FeO	54.88	80.85	79.43	81.57	80.84	80.74	15.08	0.00	18.40	10.07
MgO	8.36	2.05	1.91	1.81	1.97	1.68	14.73	4.47	1.64	8.60
CaO	2.39	3.50	3.75	3.85	3.63	3.67	33.76	45.49	31.63	29.08
Na2O	0.20	0.05	0.06	0.07	0.08	0.12	0.00	0.00	0.00	0.00
K2O	0.05	0.01	0.02	0.05	0.04	0.05	0.34	0.61	0.18	0.42
TiO2	0.07	0.01	0.04	0.03	0.06	0.06	0.00	0.00	0.00	0.00
P2O5	0.06	0.03	0.09	0.12	0.02	0.05	0.00	0.00	0.00	0.00
F	0.26	0.38	0.05	0.29	0.39	0.32	0.00	0.00	0.00	0.00
CO2	11.26	1.81	2.23	0.00	1.47	1.76	0.00	0.00	0.00	0.00
SO3	0.24	0.06	0.13	0.13	0.02	0.06	0.00	0.00	0.00	0.00
Sc2O3	0.05	0.07	0.10	0.14	0.05	0.13	1.96	2.88	2.73	1.37
CoO	0.07	0.01	0.04	0.05	0.06	0.06	0.65	0.00	0.38	0.25
NiO	0.07	0.00	0.06	0.05	0.05	0.05	0.38	0.00	0.36	0.36
SrO	0.38	0.22	0.26	0.31	0.31	0.10	1.06	7.45	8.41	6.30
Y2O3	0.53	0.29	0.35	0.27	0.24	0.24	3.71	4.21	7.61	5.51
ZrO2	0.52	0.33	0.50	0.27	0.36	0.40	2.62	10.67	12.45	7.62
Nb2O5	0.84	0.36	0.46	0.78	0.43	0.49	0.00	15.90	13.05	14.27
zr	0.86	0.55	0.83	0.69	0.62	0.68	2.52	11.58	8.72	18.63
zro2								1.51	3.95	
nb2o5								5.03	4.03	
anh	0.37	0.14	0.28	0.28		0.14				
caf2	0.73	1.38	0.21	0.96	1.24	1.02				
ap			0.21	0.34		0.20				
il	0.12		0.14		0.12	0.14				
mt	5.63	9.32	9.17	9.37	8.37	9.22	1.62		2.55	1.21
cor										
na2co3	0.55				0.31	0.41				
k2co3	0.12				0.12	0.14				
cc	4.65	5.52	6.90		3.72	5.05				
mgc	25.34									
fec	0.86									
al2c3										
sic										
scy2c3										
conico3										
cpx				21.09	19.03	21.23				
hy	36.71	21.19	22.21				40.13			12.09
sil	1.65	0.62	0.76		0.68	0.55				
or				0.34			2.10			3.20
ab				0.69						3.20
an				0.34			4.20			
qz										
scy2o3	0.12	0.07	0.14	0.14	0.06	0.07	1.14	4.53	4.68	2.70
conio	0.12		0.14	0.14	0.12	0.14	1.68		0.82	0.57
cao							35.21	68.04	53.04	40.54
mgo							11.40	9.31	3.37	11.95
feo	22.15	61.21	59.03	65.61	65.59	61.02			18.30	5.90
k2o									0.33	

Table7: EDAX Compositions of Singikulam fumarolic materials are shown

	31	32	33	34	35	36	37	38	39	40
SiO2	19.87	28.68	30.22	5.00	0	0.27	0	0	0	0.84
Al2O3	1.63	2.01	3.21	0.34	0	1.16	0.97	1.05	0.94	1.23
FeO	10.82	5.00	0.00	16.37	0	11.54	-	0.00	0.00	0.00
MgO	11.10	17.81	18.44	2.59	0	0.00	-	0.00	0.00	0.00
CaO	19.60	27.22	36.20	41.98	42.82	53.05	57.43	64.55	63.05	58.92
Na2O	0.32	0.34	0.25	0.00	0.00	0.00	-	0.00	0.00	0.00
K2O	0.25	0.31	0.25	0.51	0.00	0.28	0.28	0.12	0.44	0.00
TiO2	0.06	0.15	0.00	0.00	0.00	0.00	-	0.00	0.00	0.00
P2O5	0.22	0.23	0.00	0.00	0.00	0.80	0.78	0.00	0.38	0.62
F	0.00	0.44	0.00	0.00	0.00	0.00	-	0.00	0.00	0.00
CO2	20.05	8.78	0.00	0.00	0.00	16.09	27.08	11.38	14.11	22.63
SO3	0.35	0.40	0.00	0.00	0.00	0.48	0.18	0.00	0.20	0.24
Sc2O3	0.89	0.90	0.99	1.78	1.84	1.22	1.21	0.95	1.47	1.06
CoO	0.36	0.30	0.14	0.81	0.00	0.17	-	0.00	0.00	0.00
NiO	0.22	0.16	0.00	0.55	0.00	0.08	0.08	0.00	0.00	0.00
SrO	1.80	1.47	1.79	5.66	9.72	2.17	1.57	3.37	2.25	2.19
Y2O3	2.83	2.03	2.51	5.33	11.72	2.92	2.09	3.22	4.62	2.64
ZrO2	3.77	1.99	2.73	8.93	18.59	5.44	2.66	6.94	4.93	3.45
Nb2O5	5.83	1.79	3.26	10.16	15.31	4.33	5.67	8.44	7.60	6.17
zr	5.91	3.03	3.89	10.50		1.10				1.64
zro2					13.46	4.31	1.26	3.56	2.51	0.82
nb2o5				2.77	5.17	1.77	1.20	2.03	1.82	1.35
anh	0.46	0.66				1.33	0.23		0.25	0.35
caf2		1.38								
ap	0.46	0.53				3.20	1.66		0.82	1.41
il	0.12	0.26								
mt	1.04	0.53		1.97		2.10				
cor						2.54	1.08	1.33	1.13	1.41
na2co3	0.87	1.12								
k2co3	0.46	0.66				0.99	0.51	0.19	0.88	
cc	41.25	25.02				80.22	69.86	32.66	39.72	60.29
mgc	10.66									
fec										
al2c3										
sic										
scy2c3										
conico3										
cpx										
hy	30.59	57.14	43.43							
sil	2.78	3.88								
or			1.43	2.55						
ab			2.29							
an			7.14							
qz										
scy2o3	0.81	0.92	0.86	2.77	4.99	2.09	0.91	1.39	1.32	1.06
conio	0.46	0.39	0.11	1.31		0.33	0.06			
cao			36.46	58.53	76.38		23.2	58.83	51.5	31.67
mgo	2.26	3.95	4.40	4.66						
feo	1.85	0.53		14.65						

Fumaroles generally contain between 1 and 2 volume percent of HF [31] which indicates that HF play critical role in scoring and reacting with silicate minerals. HF presents a notable volume in vapour phase of fugitive constituents in carbonatite melt. Carbonatite lava is composed of significant quantity of F, Cl and SO_3. Degassing of volatiles and increasing of viscosities of melts play critical role on stabilization of minerals under ambient PTX conditions. Deep-mantle derived carbonatite lava has ultralow viscosity [32] and therefore, it rapidly ascends little degassing upwards without loss of heat energy either by friction and cooling [22] . Bi-variable chemical components indicate that trends of magmatic evolution but towards opposite direction to magmatic evolution but due to chemical leaching of fumarolic materials negative trend. Fig, 4a, 4b and 5 distinguish 3 or 4 groups of fumarolic trends of alterations between major and minor elements and oxides. Within these groups there exist linear positive or negative linear variations.. During silicification, Al positively increases with Si. Negative correlation of (Na+K) against Si or Al distinctly show leaching and removal of (Na+K) during the course of leaching of alkali constituents. Ca vs Mg show negative correlation between altered products. S vs P represent the alteration process. SO_3 vs Cl and F vs SO_3 show similar trends of negative correlations indicating the effects of leaching process.

Minor elements represent in the Fig. 4b. clearly show negative correlation during the processes of leaching. They exhibit linear negative correlations during the course of weathering and meteoritic leaching. In Fig:5, Sc vs Mg/(Mg+Fe) distinguishes 4 groups of alteration stages, Nb vs Zr shows perfect negative linear trends. F vs Si represents increase of F against Si which maintains constant stage of Si saturation with increasing F, Mg vs Fe show a strong negative correlation due Fe enrichment Other bi-variable components, though they are scattered show similar negative correlation by enrichment of Si, Fe, carbonates and oxides. .It is known that the rocks are subjected silicification, enrichment of iron, carbonation and oxidation from their chemical alterations.

The carbonatite lava is relatively silica alumina poor. They are composed with significant amount of sulphates like gypsum, anhydrite and xenotime. Halite and fluorite are seen. Silica undersaturated lava normative nepheline and kalsilite are produced on dry basis. Under hydrous state, portlandite forms at ambient state of crystallization with lower concentration of CO_2. This may be happened due extreme quantity of degassing of CO_2 and anhydrous volatiles relative to water vapour. Since the composition fumarolic materials are subjected to chemical leaching and removal of chemical components from the source to their present compositions cannot be correlated with either original carbonatite lava or ash tuff. Some fumarolic samples (Table 2 and 3) in Eppodumvendran-Podupatti have excessive CO_2. The sample No. 8 has very high total iron content due to ferruginous transformation of the rock. Significant amount of ferro-carbonates are present in this sample. In addition to that aluminium and silicon carbonates are present by excessive CO_2. Though most of Na_2CO_3, K_2CO_3, NaCl and KCl are readily soluble in meteoric water and their remnants are still present. K_2CO_3 is dominant over Na_2CO_3.The solubility of sodium carbonate is lower than potassium carbonate [31].

The original fumarolic material should be enriched with more quantity of sodium carbonate. This is one of the characteristic features of natrocarbonatite and its residual components. The relatively (Table 2 & 3) higher contents of Si is due to enrichment by leaching of other components due to removal of Fe, Mg, Ca, $Fe.^{2}+$, Na and K. The silicate mineral structure is easily breakable by enrichment of F which transforms into HF under hydrous state. HF easily dissolves all chemical constituents by breaking silicate structure by dissolving into meteoric water. Combined effects of Cl, SO_3 CO_2, and P_2O_5 such processes are activated. Most bi-variable components show positive linear variations due to their magmatic originalities but their dissolution effects by circulations of meteoric water on fumarolic deposits, positive trends are partially reversed in many cases (Fig. 4a, 4b and 5) Hence, there exist some uncertainty in interpreting the results.

Similar content of fumarolic deposits and net-work of encrustations are seen on limestone exposed 2 km SE of Singikulam village. In Table 4-7, fumarolic materials are highly leached out by meteoric water and SiO_2 content extremely increased. Total FeO content increases for SiO_2. CaO is extremely low. $FeCO_3$ is seen in notable amount. Studying Table 4-7 and Fig. 5, apart from magmatic variations, the fumarolic materials are subjected to leaching. The linear variation of Nb against Zr, Ni against Co, and Y against Sr ions are little altered by meteoric circulation and they show positive linear variations during magmatic evolution. However, four distinctive groups discriminated are by i) silicification caused enrichment of Si and Al consequent removal Si, Al, Fe, Mg, Ca, Na and K in silicate minerals: ii) ferruginous alterations are caused by reaction and addition of iron from meteoric water: iii) carbonation by addition of bi-carbonate materials from meteoric water: iv) oxidation of fumarolic material by intermittent circulation of meteoric water, time interval between circulation gap promotes oxidation. During course of time, fumarolic films will be complexly erased out by chemical weathering processes. Yet, there are some relics still present in dark or brown films which should be more studied to prove fumarolic deposition of carbonatite volcanism in Tamil Nadu.

7. CONCLUSIONS

The geochemical studies reveal that the following fumarolic minerals are possible to identify: quartz, goethite, portlandite, brucite, ilmenite, gypsum, anhydrites, barite, strontianite, galena, fluorite, vanadate, xenotime, apatite, williamite zircon, calcite, dolomite, ferro-carbonate, pyrochlore, topaz thermonatrite, trona, halite, sylvite, sillimanite, hypersthene, corundum, ilmenite, hematite and magnetite. Since, the role meteoric water plays a critical role in the origin of most of the minerals the compositions and textures of fumarolic minerals are totally changed. These fumarolic minerals may be identified and confirmed by using multiple sophistication instrumentation techniques. Extensive field traverses may be revealed still more occurrences of fumarolic deposits of carbonatite volcanism in some other parts of Tamil Nadu.

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