

## Discussion of Peccerillo (2004) “Carbonate-rich pyroclastic rocks from central Apennines: carbonatites or carbonated rocks?”

K. BELL<sup>1\*</sup> and B. KJARSGAARD<sup>2</sup>

<sup>1</sup> Dept. of Earth Sciences, Carleton University, Ottawa, Ontario, K1S 5B6, Canada

<sup>2</sup> Geological Survey of Canada, 601 Booth Street, Ottawa, Ontario, K1A 0E8, Canada

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### INTRODUCTION

Carbonate-rich rocks associated with kamafugites in central Italy are considered by Stoppa and Woolley (1997) to be carbonatites. If this is so, then severe constraints can be placed on any geodynamic model proposed for Italy, given that most carbonatites are intraplate, and some may even be related to plume activity.

The *sine qua non* of Italian geology is that most Cenozoic igneous activity is subduction-related, but this has repeatedly been challenged because many of the rock types found in Italy are absent from active, consuming plate margins. Peccerillo (1998, 2004, 2005) has argued that the carbonate-rich rocks are not in fact carbonatites, but simply reflect the involvement of sedimentary carbonates caught up during migration of small-volume mantle melts through the thick and extensive limestone, marl and evaporite sequences that lie along the length of peninsular Italy. In this contribution we assess both sides of the debate, and attempt to clarify some of the arguments presented. We discuss these in point form:

### DISCUSSION

1). We remain quite unclear about the model that Peccerillo is proposing. The involvement of limestones can take many forms, such as partial assimilation, bulk assimilation, comminution associated with fragmentation of a mantle-derived melt, or simple wall-rock reaction. In Peccerillo's most recent paper (2004), he poses the question as to whether the carbonate-rich rocks “*are magmatic carbonatites, or whether they are fragmented silicate rocks into which calcite has been incorporated during their passage through the sedimentary rocks which they have intruded*”, and also states that the supposed carbonate-rich pyroclastics represent “*fragmented kamafugitic magmas, which have suffered addition of geochemically barren carbonate material from wall rocks*”. Carbonate incorporation then involves “*interaction between silicate magmas and the wall rocks. Such an interaction is not restricted to bulk rock assimilation but may include dissolution of carbonate from the volcanic conduit and re-precipitation, and post intrusion precipitation of carbonate from hot solutions, etc.*”. These statements by Peccerillo (2004) make it difficult, if not impossible, to understand his petrogenetic model for the origin of the carbonate-rich rocks. Although a clearly-stated fate for the sedimentary

\* Corresponding author, E-mail: [kbell@ccs.carleton.ca](mailto:kbell@ccs.carleton.ca)

carbonate-rich wall rock is NOT noted in Peccerillo's paper, we believe there are three potentially viable options: A.) the carbonate wall rock is mechanically incorporated as discrete clasts into the host silicate kamafugite melt and does not geochemically interact with it (closed system), or; B) the carbonate wall rock is assimilated into the host silicate kamafugite magma i.e. it melts, or; C) the carbonate wall rock is mechanically incorporated as discrete clasts into the host silicate kamafugite magma and geochemically interacts with it (open system).

Option A does not account for the geochemical characteristics of the carbonate-rich rocks that differ significantly from the host rock limestones and are fairly similar to typical extrusive carbonatites e.g. Fort Portal – Uganda (Barker and Nixon, 1989). Furthermore, extrusive carbonatites have trace element signatures similar to those of associated co-magmatic silicate igneous rocks. Although these values are generally 2-3 times lower than those of average intrusive carbonatites they are still many times more abundant than sedimentary limestones (Woolley *et al.*, 2005). Geochemical mass balance arguments do not support Option A (wall rock contamination model, see point #6 below).

Option B, assimilation of carbonate wall rock into kamafugite magma, is highly unlikely. Numerous experimental and theoretical studies have considered the fate of silicate magmas which are contaminated by sedimentary carbonates, with respect to the formation of carbonatite magmas (Watkinson and Wyllie, 1969; Wyllie, 1974, 1989). These studies all indicated that it is not feasible to produce a carbonatite magma by assimilation of sedimentary carbonates into a silicate melt. Furthermore, the general principle that assimilation induces 'thermal death' to any magma is certainly valid, and not consistent with the observation of a variety of carbonatite pyroclastic extrusive rocks. In addition, the presence of mantle debris (e.g. at San Venanzo and Polino; Stoppa and Woolley, 1997) is a testament to the rapid ascent of the kamafugitic and carbonatitic magmas. Option C, mechanical incorporation of carbonate wall-rock into kamafugite magma and open system geochemical interaction is highly problematic. It is difficult to envisage an eruptive mechanism whereby the discrete calcite clasts from the limestone wall-rock assimilate, and now within the kamafugite magma,

could be concentrated to form low silica (<20 wt% SiO<sub>2</sub>) calcite-rich, carbonatite pyroclastics.

2). Field and volcanological observations at Polino, San Venanzo and Cupaello, including measured sections of volcanic deposits with carbonatite base surge, pyroclastic flow, and lapilli tuff beds (Stoppa and Lavecchia, 1992; Stoppa and Woolley, 1997 – their Figure 3), are consistent with the existence of extrusive carbonatite magma. Many of these volcanological features have been noted in other carbonatites worldwide (e.g. Keller, 1981; Hay and O'Neil, 1983; Keller, 1989). Furthermore, volcanic features such as carbonate lapilli, especially spin lapilli (Mittenpergher, 1965; Junqueira-Brod *et al.*, 1999; Stoppa *et al.*, 2003) are evidence for the existence of carbonatite magma. In passing, we also note that Peccerillo (2004) describes the carbonate-rich rocks as pyroclastics, which by definition, form by a process of fragmentation of a magma (Fischer and Schmincke, 1983; Cas and Wright, 1987). Hence, we find this self-contradiction by Peccerillo (2004) confusing – the existence of carbonate-rich magmatic rocks which did not form from a carbonatite magma.

3). Sr isotopic disequilibrium between carbonate and insoluble silicates from Polino and Cupaello, is considered by Peccerillo (2004) to be inconsistent with the carbonatite model, but this only holds true if the carbonatites and silicate lavas were generated from the same melt by magmatic differentiation (crystal fractionation or liquid immiscibility) operating under closed chemical conditions. Other scenarios, however, are possible. The silicate and carbonatitic melts could be derived from sources with quite different isotopic signatures. In addition, the literature includes many examples of Nd, Pb and Sr isotopic differences: 1.) among mineral phases in alkaline silicate rocks e.g. (Simonetti and Bell, 1993), 2.) within carbonatites themselves (Bizzarro *et al.*, 2003), and 3.) between silicate rocks and carbonatites from the same complex (e.g. Bell, 1998; Harmer and Gittins, 1998). Therefore, isotopic disequilibrium on its own cannot be used as evidence against a magmatic origin for the carbonate-rich rocks in Italy. It could simply reflect the involvement of different sources and possible

mixing between melts derived from them, or open-system behavior.

We are surprised that Peccerillo (2004) did not point out that the carbonate-rich rocks from Italy have unusually high  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios (e.g. Castorina *et al.*, 2000), quite different to those measured from most young carbonatites found elsewhere on Earth. Recently, however, intrusive carbonatites from China (Ying *et al.*, 2004) have been reported with high  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios, reflecting the involvement of a mantle source enriched relative to sub-oceanic mantle and similar, in some ways, to the enriched mantle source that underlies Italy (ITEM, Italian Enriched Mantle); the "ITEM" end-member has high Sr, low Nd and intermediate Pb isotopic ratios (Bell *et al.*, 2004; 2005). ITEM is best reflected by data from the Pliocene Tuscan lamproites and from Alpine lamprophyres. The highest  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios are shown by lamprophyric dykes from the Alps. A high ITEM contribution also characterizes potassic rocks of the Roman Region, and the carbonatites of Central Italy. Although the isotopic signature of ITEM is similar to pelagic sediments/upper crust, it forms a distinct end member and differs from isotopic ratios found in present day, subduction-related, igneous rock suites. In addition, the high  $^{87}\text{Sr}/^{86}\text{Sr}$  and low  $^{143}\text{Nd}/^{144}\text{Nd}$  isotopic ratios are similar to K-rich, diamond-bearing lamproites and some kimberlites from cratonic areas suggesting that the source of ITEM could also be the deep mantle (Bell *et al.*, 2004).

4). Given the explosive (pyroclastic) nature of the carbonate-rich rocks (i.e. open-system behaviour), it is not surprising that their  $\delta^{18}\text{O}$  values are much higher than those typical of plutonic carbonatites. It should also be remembered that the  $\delta^{18}\text{O}$  for carbonatites can be quite high, the result of Rayleigh fractionation during magma evolution (Deines, 1989).

As Peccerillo (2004) points out, the high  $\delta^{18}\text{O}$  values of carbonates can result from post-crystallization exchange with ground water, so nothing is to be gained by comparison of the carbonate-rich rocks with others. Peccerillo (2004) also emphasizes the high  $\delta^{18}\text{O}$  in calcite from the carbonate-rich volcanic rocks, and draws attention to the high values in *fresh* pyroxene and olivine of silicate lavas supposedly in support of his argument of interaction with wall rocks.

This feature we consider inconsistent with wall-rock reaction involving assimilation and/or fluid exchange. We would rather attribute the high  $\delta^{18}\text{O}$  in the *fresh* pyroxene and olivine of silicate lavas to an unusual mantle source (e.g. Vollmer, 1989). The high  $\delta^{18}\text{O}$  values also characterize most of the Roman Comagmatic Province rocks, including those from primitive magmas such as leucitites (e.g. Holm and Munksgaard, 1982; Ferrara *et al.*, 1985; Vollmer, 1989). High  $\delta^{18}\text{O}$  values have also been found in both melilitites and carbonatite (Stoppa and Woolley, 1997).

5). The Italian carbonate-rich rocks are pyroclastic, and we believe it is highly problematic to compare their geochemistry to plutonic carbonatites. So far about fifty extrusive or pyroclastic carbonatites had been identified worldwide (Woolley and Church, 2005). In general terms, the trace element contents of pyroclastic carbonatites are lower than those of plutonic carbonatites (Woolley and Church, 2005), which in part is due to the incorporation of fragments of country rocks or crustal xenocrysts, and their general open system behaviour. Peccerillo (2004) is correct in noting that the chemical compositions of the Italian carbonate-rich rocks do not look like worldwide carbonatites – but we suggest this is a reflection of the source, and that comparing Italian pyroclastic carbonatites to world average plutonic calcite carbonatite (sövite) is meaningless.

6). Then there is the question of the trace element contents of the carbonate-rich rocks, or calcite-rich kamafugite which have lower (Cupaello) or similar (San Venanzo) concentrations to those in the accompanying calcite-poor kamafugites. Implied in the trace-element enrichment (relative to barren limestone) is a nebulous model involving transfer of the trace element pattern of the silicate melt to the carbonates. Peccerillo (2004) states "*The carbonate-rich rocks are depleted in several trace and major elements relative to kamafugitic magmas from the same volcano*" and applies a straight dilution argument to resolve the problem i.e. silicate rocks, and silicate rocks which have added sedimentary material. The main flaws in such an argument are that: 1) The carbonatites have elevated MREE/HREE ratios as compared to the Apennine sedimentary carbonates (see

Peccerillo, 2004; Fig. 2b); 2) The carbonatites have LREE (La to Nd) that form a smooth REE pattern on a chondrite normalized plot, in contrast to the Apennine sedimentary carbonates, which commonly exhibit positive Ce or Nd anomalies and in general have irregular, not smooth, LREE patterns (see Peccerillo, 2004; Fig. 2b); 3) Carbonatites have extended-element patterns that closely mimic those of the kamafugites. Addition of sedimentary limestone (which has a disparate, extended-element pattern to the kamafugites) to kamafugite will produce an extended element pattern intermediate between that of the limestone and that of the kamafugite, but this is not observed. In Table 1, we have modelled the addition of 40%, 50% and 60% limestone to Cupaello kamafugite. We then compare this calculated “carbonatite” composition to the composition of Cupaello carbonatite (Table 1, Figure 1). It is clear from Figure 1, an enrichment/depletion diagram, that limestone addition to kamafugite cannot explain the geochemistry of the carbonatites, because of decoupling between major and trace elements and also between some major elements, and some trace elements. The only likely way that the carbonates can acquire the trace element signatures of the silicate rocks is by carbonate wall-rock assimilation and subsequent formation of a carbonatite magma, which we do not believe is a plausible mechanism.

### *Concluding statements*

In the absence of any clearly-defined model by Peccerillo (2004), it is difficult to assess his proposal for the origin of the carbonate-rich rocks found in central Italy. Possible models involving limestone wall rock assimilation/incorporation/interaction are all weak or flawed. The observation that carbonate-rich pyroclastic rocks occur in central Italy, indicates the former existence of carbonatite magmas. In this regard, we find it strange that Peccerillo (2004), who is so disposed to subduction-related processes, did not invoke melting of a carbonated slab because of his statements that “*a mantle contaminated by subduction-related upper crust*” is considered to play a key role in generating the “*unusual mantle from which many of the Italian mafic, and strongly silica-undersaturated melts were derived*”.

We also disagree with Peccerillo that Popperian falsification is hardly applicable to the Earth Sciences. If something is not falsifiable, at least in principle, then there is very little difference between science and witchcraft. The comment that crucial data or experiments rarely exist in geology can be contested. Field observations, geochemical data and geophysical experiments form part of the data sets used by Earth scientists. Anyone familiar with the history of ideas involved with plate tectonics realizes that observations were critical, in first formulating a hypothesis and subsequently, along with other information, in establishing a theory.

Peccerillo’s contributions involving carbonatitic magmatism in Italy seems to be a replay of the ideas proposed, discussed and then discarded by earlier workers in the early- to mid-twentieth century regarding the role of limestone in the origin of carbonatites and the formation of silica-undersaturated magmas (e.g. Daly, 1910; Brögger, 1921; Dixey and Smith, 1929; Daly, 1933; von Eckerman, 1948). Silica-undersaturated silicate extrusive rocks are common in central Italy and some are isotopically distinct with high  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios, yet no one in recent years has advocated the involvement of limestones in generating such rocks. Rather their unusual isotopic signatures simply reflect the involvement of the isotopically-distinct, ITEM source. The gross similarities of the isotopic ratios in the carbonate-rich rocks and some of the silica undersaturated rocks, coupled with the gross dissimilarities between the carbonate-rich rocks and the isotope ratios found in the sedimentary limestones (Bell and Castorina, unpublished data), suggest that those rocks with high  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios are primary, and simply reflect an enriched, heterogeneous mantle source. Peccerillo’s ideas are clearly at odds not only with a wide range of field observations but with geochemical data that are best interpreted as supporting the idea that the carbonate-rich rocks at Polino, San Venanzo and Cupaello are, in fact, carbonatites.

To summarise, our main points of concern are: 1. the lack of any consistent explanation in Peccerillo’s model for the role and ultimate fate of sedimentary carbonates (are they remobilized?, assimilated? fragmented?) and 2. an adequate reason for the marked differences in chemical signatures between the carbonate-rich, igneous

TABLE 1  
Mixing models

Sample	CUP		CM	CM60	CM50	CM40	CM60 vs		CM50 vs		CM40 vs	
	kamafugite	CUP carbonatite					CUP carb	CUP carb	CUP carb	CUP carb	CUP carb	CUP carb
SiO <sub>2</sub>	43.6	19.35	0.11	17.51	21.86	26.2	0.9	1.13	1.35			
TiO <sub>2</sub>	1.15	0.41	<b>0.05</b>	0.49	0.6	0.71	1.2	1.46	1.73			
Al <sub>2</sub> O <sub>3</sub>	8.01	2.65	<b>0.05</b>	3.23	4.03	4.83	1.22	1.52	1.82			
Fe <sub>2</sub> O <sub>3T</sub>	7.74	2.76	<b>0.05</b>	3.13	3.90	4.66	1.13	1.41	1.69			
MnO	0.11	0.06	<b>0.05</b>	0.07	0.08	0.09	1.35	1.45	1.56			
MgO	11.90	7.96	0.13	4.84	6.02	7.19	0.61	0.76	0.90			
CaO	13.60	31.96	56.97	39.62	35.29	30.95	1.24	1.10	0.97			
Na <sub>2</sub> O	0.23	0.40	<b>0.05</b>	0.12	0.14	0.16	0.31	0.35	0.40			
K <sub>2</sub> O	5.56	0.93	<b>0.05</b>	2.25	2.81	3.36	2.44	3.03	3.63			
P <sub>2</sub> O <sub>5</sub>	1.19	0.58	<b>0.05</b>	0.51	0.62	0.73	0.88	1.08	1.28			
Rb	491	152	5.0	199	248	297	1.31	1.63	1.95			
Ba	4218	918	<b>2.5</b>	1689	2110	2532	1.84	2.3	2.76			
Sr	3857	1167	11.4	1611	1986	2360	1.38	1.7	2.02			
Th	116	41.5	22	59.6	69.0	78.4	1.44	1.66	1.89			
Nb	45	20.5	<b>2.5</b>	19.5	23.8	28.0	0.95	1.16	1.37			
Zr	837	283	<b>2.5</b>	336	420	503	1.19	1.49	1.78			
Cr	60	30	<b>2.5</b>	26	31	37	0.85	1.04	1.23			
Ni	56	28	6	26	31	36	0.93	1.11	1.29			
Y	46	15	11	25	29	32	1.67	1.9	2.13			
La	271	95	1.8	110	136	163	1.15	1.44	1.72			
Ce	519	184	15.9	217	268	318	1.18	1.45	1.73			
Nd	229	81.7	2.5	93.1	116	138	1.14	1.42	1.69			
Sm	40	15.1	0.8	16.5	20.4	24.3	1.09	1.35	1.61			
Eu	6.5	2.9	0.10	2.7	3.3	3.9	0.92	1.14	1.36			
Gd	25.9	12.2	0.70	10.8	13.3	15.8	0.88	1.09	1.3			
Yb	2.6	0.9	0.10	1.1	1.3	1.6	1.2	1.48	1.76			
Lu	0.4	0.1	0	0.2	0.2	0.3	1.84	2.2	2.56			

Cupuello (CUP) kamafugite from Peccerillo (2004); Cupuello (CUP) carbonatite (average of 2) from Stoppa and Woolley (1997); "Calcare Massiccio" (CM) limestone from Stoppa et al. (2005). CM60 = 60% CM limestone + 40% CUP kamafugite; CM50 = 50% CM limestone + 50% CUP kamafugite; CM40 = 40% CM limestone + 60% CUP kamafugite; CM60 vs CUP carb = (60% CM limestone + 40% CUP kamafugite)/CUP carb; CM50 vs CUP carb = (50% CM limestone + 50% CUP kamafugite)/CUP carb; CM40 vs CUP carb = (40% CM limestone + 60% CUP kamafugite)/CUP carb. Values in bold are one half the detection limit for the purposes of the calculations.

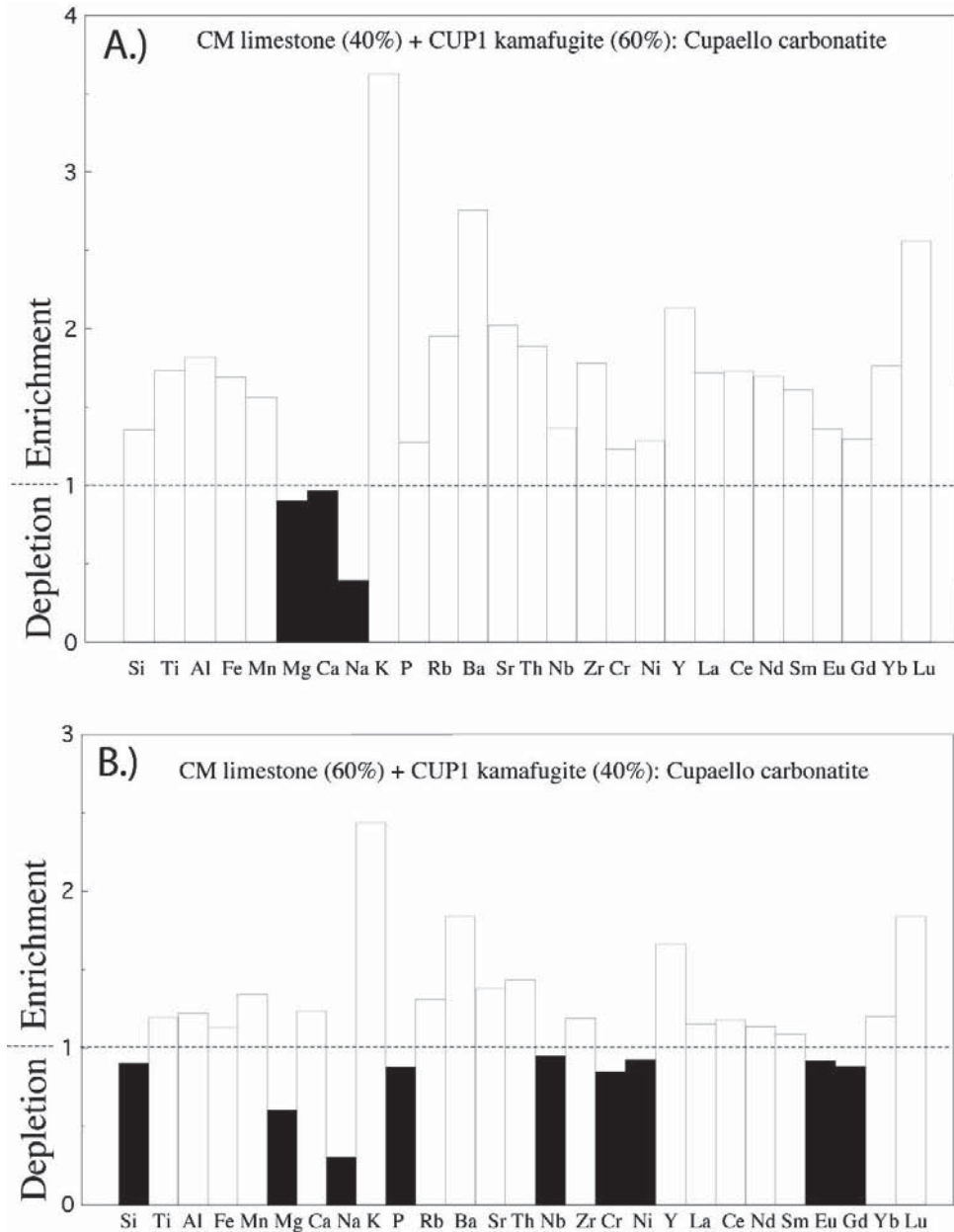


Fig. 1 – Enrichment/depletion diagram comparing the calculated composition of a kamafugite with 40% or 60% limestone assimilation. In A.) note that for 40% limestone assimilation the Mg, Ca and Na levels are comparable between the calculated composition and the Cupaello carbonatite, but all other major elements and trace elements are enriched (1.3x to 3.6x). In B.) note that for 60% limestone assimilation, enrichment levels are reduced (1.1x to 2.4x), but Si, Mg, Na, P, Nb, Cr, Ni, Eu, Gd are depleted (0.6x to 0.93x) in the calculated composition as compared to the Cupaello carbonatite. Black areas indicate depletion.

rocks and limestone. Although arguments can be made against any one of these points when taken on their own and out of context, in summing up the evidence we are forced to conclude that the carbonate-rich rocks are, indeed, carbonatites and have little to do with sedimentary carbonates.

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