

Reply to: "Carbonate-rich pyroclastic rocks from central Apennines: carbonatites or carbonated rocks? A commentary". A. Peccerillo

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ABSTRACT. — In a recent paper Peccerillo (2004) quoted chemical, including isotopic, mineralogical and field data which he claimed "...cast serious doubt on the hypothesis that the carbonate-rich pyroclastics from central Italy represent carbonatitic magmas.". He concluded that the carbonate is derived from sedimentary limestone. He also noted that he had enunciated these ideas earlier (Peccerillo, 1998) but that he had not been answered and further that "the Editor of this journal [Periodico di Mineralogia] has repeatedly asked me to set down my case again and I have, reluctantly, agreed to do so.". We, some of the principal advocates of the carbonatitic interpretation for the igneous carbonate-rich rocks of central Italy, have, therefore, taken this opportunity to answer, in detail, the various arguments advanced by Peccerillo. We remain firmly convinced that the evidence demonstrates that the carbonate-rich rocks at the five localities of San Venanzo, Cupaello, Polino, Oricola and Vulture are carbonatitic, the carbonate deriving from deep within the mantle, and that sedimentary limestones played no immediate part in their genesis.

RIASSUNTO. — Recentemente Peccerillo (2004) ha riportato dati chimici, inclusi quelli isotopici, e inoltre mineralogici e di campagna, che a suo dire "...gettano

seri dubbi sull'ipotesi che le piroclastiti ricche di carbonato dell'Italia centrale possano derivare da magmi carbonatitici". L'Autore conclude, quindi, che il carbonato è di origine sedimentaria. Egli rimarca di aver avanzato questi dubbi già in precedenza (Peccerillo, 1998) ma di non aver ricevuto alcuna risposta e che inoltre "l'editore di questa rivista [Periodico di Mineralogia] mi ha ripetutamente chiesto di proporre ancora la questione e io, con riluttanza, ho dovuto convenire di farlo". Noi che siamo tra i principali assertori dell'interpretazione in senso carbonatitico delle rocce ignee a carbonato dell'Italia centrale, abbiamo quindi colto l'occasione per replicare, in dettaglio, alle varie obiezioni avanzate da Peccerillo. Siamo fermamente convinti che tutte le evidenze dimostrino che le rocce ignee a carbonato, delle cinque località di San Venanzo, Cupaello, Polino, Oricola e Vulture, siano carbonatitiche e che il loro carbonato derivi in profondità dal mantello e che i carbonati sedimentari non abbiano alcun ruolo diretto nella loro genesi.

KEY WORDS: *carbonatite, sedimentary limestone, Roman Province, Italy.*

INTRODUCTION

In a recent paper Peccerillo (2004) restated more fully a position he had taken earlier (Peccerillo,

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1998, 2003) that the carbonate-rich rocks of the San Venanzo, Cupaello and Polino localities of central Italy are mixtures of kamafugite magma and sedimentary limestone, in contrast to the position of Stoppa and Woolley (1997) who interpreted them as carbonatites, i.e., that the carbonate, as well as the silicate, fractions are igneous in origin. Peccerillo (2004, p. 172) wrote "These objections have been raised previously (Peccerillo, 1998), but have not been answered and have been reiterated here only because it has been repeatedly requested by the Editor of this journal." During the International Geological Congress in Firenze (August, 2004) Peccerillo again stated his position, although his 2004 paper was not at that time available to the present authors. In direct contrast to Peccerillo (2004), we firmly believe that the carbonate-rich rocks at San Venanzo, Cupaello and Polino are carbonatites.

Whether or not the carbonate-bearing igneous rocks of central Italy are carbonatites is of considerable interest, and importance, in the light of the numerous papers that have been published in recent years on the petrogenesis of the Roman Province, *sensu lato*. Carbonatite involvement in the petrogenesis of the Province has been proposed previously by a number of authors (Marinelli, 1975; Cundari, 1979; Locardi, 1985) before the first description of a carbonatite in Italy (Stoppa and Lupini, 1993). The complexity of Italian volcanism both in terms of the broad range of rock types involved, the structural setting and the apparent unusual nature of the mantle beneath central and southern Italy is manifest. Wherever carbonatites are found, although small in volume, we believe that they raise petrogenetic considerations that are important, and should be considered as such.

REPLY TO PECCERILLO'S OBJECTIONS TO CARBONATITE INTERPRETATION

Peccerillo lists six main arguments that he says have been advanced in favour of "their being carbonatites". These basically cover the same ground as the three points of his 1998 paper with the addition of further discussion on Sr isotope equilibria and C stable isotopes. He then raises objections to each of these in turn. We will answer these points, in the same order.

Peccerillo goes on to discuss producing the carbonate-rich rocks by the mixing of kamafugitic lavas with "barren carbonate material, such as the Apennine limestones". This point is illustrated by a mixing diagram of La and Th for lavas from Cupaello and San Venanzo with limestone. We do not address the mixing model because it is tackled independently and fully by D.K. Bailey in a separate contribution (this issue).

1. *High concentrations of Large Ion Lithophile Elements (LILE) in carbonatites*

We did not use this argument to identify the carbonate-rich rocks as carbonatites, but we did note that these rocks, like typical carbonatites, do have high tenors of LILE. We are aware that high LILE values are not diagnostic and may be found in other igneous 'silicate' rocks, which certainly cannot be said for sedimentary limestones.

It should be remembered that the Italian carbonatites are essentially extrusive, and that this is an important factor that should be taken into account when discussing their whole-rock chemistry. The Polino occurrence is a small diatreme, but certainly a very shallow one, and probably had an extrusive facies. We know that there are significant differences in chemistry between 'extrusive' and 'intrusive' carbonatites (Woolley and Church, in press). The average values for a range of elements in carbonatites presented by Woolley and Kempe (1989) were based on a large database but almost all the analyses were derived from intrusive carbonatites. About 50 occurrences of extrusive carbonatite have been described and, apart from the recent lavas of Oldoinyo Lengai and a few lava flows, almost all of them have LILE values below the Woolley and Kempe (1989) averages, but are still enriched relative to limestone, and all of the Italian carbonatite data conform to this pattern.

2. *REE patterns*

It is pointed out by Peccerillo (1998, 2004) that the carbonate-rich volcanic rocks have highly fractionated REE, including negative Eu anomalies, as do both venanzite and coppaellite, as well as leucite tephrites of the Roman Province. He continues "These features are closer to ultrapotassic Roman Province rocks than to carbonatites". We agree that the negative Eu anomaly parallels

that found in Roman Province rocks, and Stoppa and Woolley (1997) have already referred to the negative Eu anomaly in venanzite and coppaelite, both plagioclase-free rocks. We must point out, however, that Peccerillo did acknowledge that Stoppa and Woolley (1997) "have noted that carbonatites do not normally have negative Eu anomalies, although some do". It might be noted that Eu anomalies also occur in, for instance, some Tanzanian carbonatites (Stoppa, 2003), and Dr. M.J. Le Bas (pers. comm., 2005) has found Eu anomalies in some alvikites.

We assume that Peccerillo compared the Roman Province data with the average carbonatite REE pattern of Woolley and Kempe (1989), which he gives on his diagrams. As already pointed out, however, the average values of Woolley and Kempe (1989) are overwhelmingly based on intrusive carbonatite data, while chemical data from extrusive carbonatites (Church, 1996; Woolley and Church, in press) show that the incompatible elements, and notably the REE, Sr, Ba and Nb, are significantly lower than in the intrusive carbonatites.

A second feature which reduces the REE (and other LILE abundances), is secondary recrystallisation of the calcite. This particular problem is discussed more fully below.

3. Trace element contents of calcite

"The high trace element contents of calcites are not exclusive to carbonatites" (Peccerillo, 2004). Peccerillo notes that calcite in skarns from the Lascar volcano of Chile show LREE enrichments up to 70 x chondrite, which is rather lower than the San Venanzo, Cupaello or Polino rocks, in which La, for instance, lies in the range 200-800 x chondrite, and that high values of Sr and Ba are found in skarns in the Alban Hills. No mention of Ba is to be found in the paper by Federico and Peccerillo (2002) on the Alban Hills skarns and there are no data for REE. However, they did apparently find a maximum SrO value of 10.03 wt% in one specimen. These comparisons are not relevant to the present discussion because we are not dealing with skarns. At Polino, for instance, small xenoliths of limestone can be clearly seen (e.g. Stoppa and Lupini, 1993, Fig. 2A; and Fig. 1A of this paper) and the walls of the diatreme can

also be examined. In neither case is there any sign of skarn development.

Stoppa and Woolley (1997, Table 1 and Fig. 6) presented a great deal of data for BaO, SrO and REE (all wt%) for calcite from San Venanzo, Cupaello, Polino and Monticchio (Vulture). The REE data include many analyses with values 0.1-0.4 wt% i.e. 1,000 to 4,000 ppm. There are also values <0.1 wt% but these have been found only in secondary calcite. Similar distributions of values are demonstrated by SrO (0-1.5 wt%) and BaO (0-0.7 wt%).

No one has considered these occurrences to be skarn deposits on the basis of field observations, and their geological setting is not conducive to skarn formation. The IUP (Intra-appennine – ultra-alkaline – province) occurrences are all monogenetic cones or diatremes and must have been emplaced very rapidly and, probably, under supercooled conditions.

Even the textures are inconsistent with skarn formation. The calcite in all the Italian carbonatite occurrences (San Venanzo, Cupaello, Polino, Oricola and Vulture) is fine-grained and when fragments of sedimentary limestone are present, e.g. at San Venanzo and Polino, they are readily distinguished, consisting of irregular pieces of a range of sizes and shapes (Fig. 1A and B). We cannot envisage a mechanism that would generate from sedimentary limestones the uniform, fine-grained texture of the carbonate-rich rocks found at all five Italian occurrences. The texture of the carbonate is more consistent with melt crystallization, and similar to that displayed by carbonatitic tuffs at many localities outside of Italy.

4. Minerals unique, or not unique, to carbonatites

Peccerillo (2004) writes "Minerals such as baddeleyite, Zr-rich garnet (~5% ZrO₂), Th-rich pyrochlore, REE-rich perovskite are not unique to carbonatites and have been reported from skarn xenoliths in the Alban Hills (Federico *et al.*, 1994; Federico and Peccerillo, 2002)". We agree with this statement and wish to point out that we did not say or imply that these minerals are unique to carbonatites. Indeed, only Zr-rich garnet of this list occurs in Italian carbonatites.

5. Sr isotopes

Peccerillo (2004, p. 170) claims that the Sr isotopic compositions of the Polino and Cupaello carbonatites (in fact the sample PPC-3d that he cites is from San Venanzo) have different Sr isotope ratios for the silicate and carbonate fractions, concluding that “Clearly, the silicate and carbonate fractions are not in isotopic equilibrium.” This observation is the exception rather than the rule, as the majority of the available isotopic data indicate substantial homogeneity at the fourth decimal place for the same outcrop (Castorina *et al.*, 2000, Table 2). Thus, it appears that Peccerillo took a small, unrepresentative set of the data of Castorina *et al.* (2000) which he discussed in isolation.

The variation of $^{87}\text{Sr}/^{86}\text{Sr}$ amongst the Polino and Cupaello carbonatites, and the San Venanzo carbonatitic rocks and kamafugites, are in the range between 0.710075 and 0.71193. It is noteworthy that this variation is comparable with, or lower than, that observed in the classical East African carbonatites (0.70307-0.70505; Bell and Tilton, 2001) and other regions (Dunworth and Bell, 2001). The variation between the silicate and carbonate fractions of the Italian carbonatites and kamafugites is much lower. For example, in San Venanzo rocks, the carbonate and silicate fractions in a kamafugite lava (average of three samples) gave a difference of 0.000448 (± 0.000011), a carbonatitic tuff sample (PPC 9f) gave 0.000023 (± 0.000014) and a carbonatitic

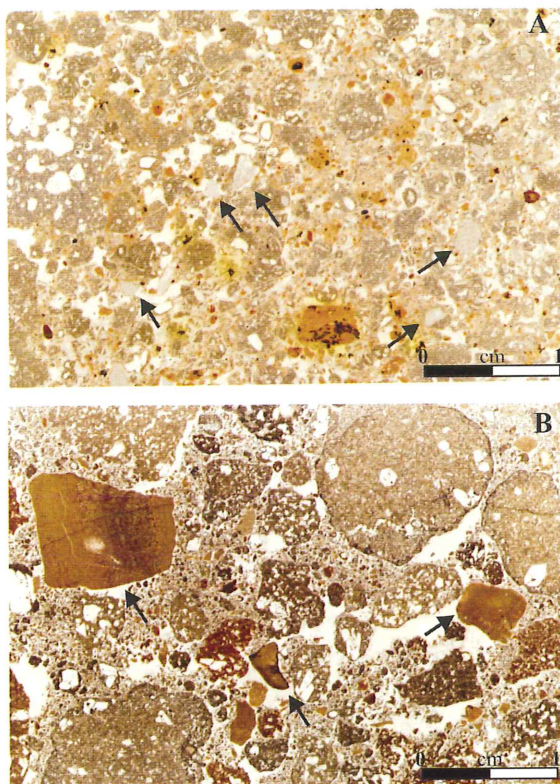


Fig. 1 – Photomicrographs, in reflected light, of San Venanzo and Polino carbonatitic and carbonatite tuffisites. A. Carbonatite tuffisite from Polino (PO2-4). The darker clasts are sedimentary rock fragments. Note the white, fresh limestone fragments (indicated by arrows). All the lapilli and the fine-grained matrix are composed of carbonatite. B. Carbonatitic tuffisite (PPC-9f). Large melilitite lapilli and sharp marly limestone fragments (indicated by arrows) in a fine-grained carbonatitic matrix. The 2 cm scale bar is the same for both figures.

melilitolite sill (average of two sample) gave 0.000041 (± 0.000012) (data from Castorina *et al.*, 2000). The reproducibility of the $^{87}\text{Sr}/^{86}\text{Sr}$ ratios is 4 parts in 70,000.

Silicate minerals in these rocks show larger variation (sanidine in carbonatitic tuff of San Venanzo gave 0.710744, clinopyroxene in Cupaello carbonatite gave 0.711267 and Polino phlogopite gave 0.715371). It is notable that such differences are also observed in other alkaline carbonatite massifs (Dunworth and Bell, 2001).

Different outcrops are spatially resolved in terms of the Sr-Nd diagram (Fig. 2), and are considered by the authors to be primary and link different melilititic-carbonatitic magma batches to

an isotopically heterogeneous mantle source, such as that proposed by Bell *et al.* (2004). Significant intra-outcrop differences have been noted in Cupaello carbonatite and kalsilitite pairs (COP8 and COP11 of Stoppa and Cundari, 1995) and in the silicate fraction of a single carbonatitic tuff layer at Pian di Celle (San Venanzo, sample PPC3d, Fig. 2). The last example was used by Peccerillo as his main point of criticism of the Sr isotope equilibrium between the carbonate and silicate fractions of Italian kamafugites and carbonatites. The trends point towards enrichment of the silicate fraction in ^{87}Sr .

Peccerillo (2004) suggests that the isotopic disequilibrium between carbonate and silicate

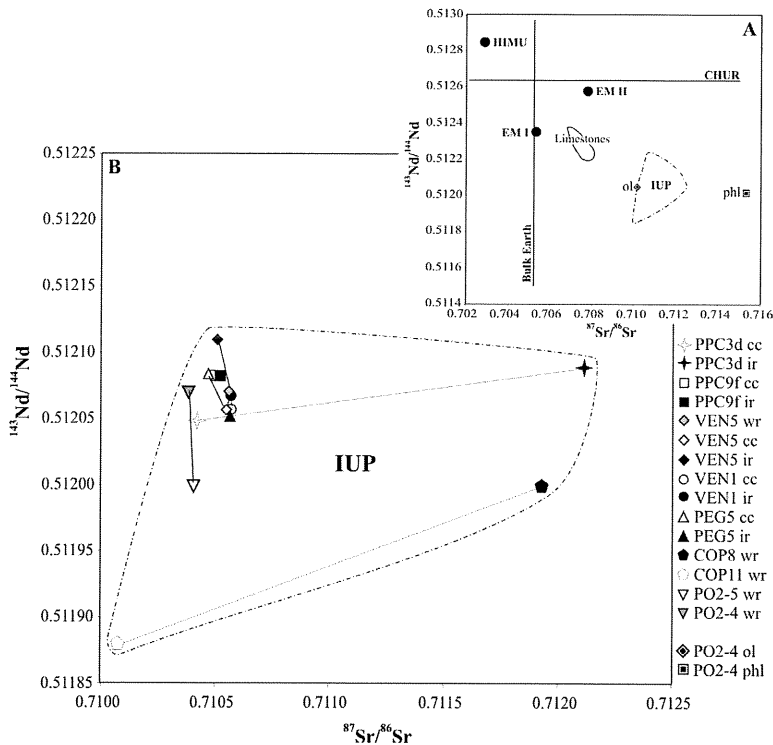


Fig. 2 – Plot of $^{87}\text{Sr}/^{86}\text{Sr}$ vs $^{143}\text{Nd}/^{144}\text{Nd}$ ratios of the IUP alkaline silicate and carbonatite rocks (data from Castorina *et al.*, 2000). A. Graph showing the distribution of Jurassic-Miocene Italian limestones (data courtesy of K. Bell), IUP rocks, olivine and phlogopite from IUP rocks with respect to the common mantle reservoir end members (Zindler and Hart, 1986). B. Plot of a range of samples from San Venanzo and Polino. Tie-lines connect the carbonate and residue fractions from the same sample. PPC = San Venanzo carbonatitic tuffs; VEN = San Venanzo kamafugite lava; PEG = San Venanzo melilitolite. PO2-4, Polino carbonatite tuffsite; PO2-5, Polino massive carbonatite; COP 8, Cupaello, kalsilitite; COP11, Cupaello, carbonatite. wr, whole rock; cc, HCl leached carbonate fraction; ir, insoluble residue after HCl leaching (Castorina *et al.*, 2000).

volcanic rocks are depleted in several trace and major elements relative to the kamafugitic lavas *from the same volcano*, but the element depletion is proportional to their carbonate content.” He is implying, presumably, that the carbonate-rich tuffs have been diluted with barren limestone. In fact, Stoppa and Woolley (1997) give analyses of the REE of leachates from these two rocks which indicate that the soluble fractions (essentially carbonate) are rich, in comparison with limestone, in these elements.

At Oricola the REE data of Stoppa *et al.* (in press, Fig. 4) for the co-existing carbonate lapilli and silicate glass fragments demonstrate that the higher REE values are in the carbonate. We illustrate this point by reproducing here (Fig. 4) part of Stoppa *et al.*'s Fig. 4. This relationship conforms to Peccerillo's carbonatite model so, on the basis of his own favoured criterion, he would,

presumably, accept this occurrence as comprising genuine carbonatite.

2. The Vulture (Monticchio) occurrence and direct evidence of mantle carbonate

The Monticchio carbonatite, within the Vulture volcanic complex, lies about 200 km southeast of the IUP but is on the same spatial trend lying to the east of the Roman Volcanic Region. Although Vulture is chemically rather different from the IUP occurrences, it is of approximately the same age and includes melilitites and melilitolites, that suggest a link with the melilite-bearing IUP rocks. Calcite carbonatite has also been collected in ejecta enclosed within the Vulture volcanic pile (Rosatelli *et al.*, 2000). At Monticchio the carbonatites are pyroclastic deposits that contain mantle xenocrysts and xenoliths which originate at depths of about 70 km (Jones *et al.*, 2000). Abundant accretionary

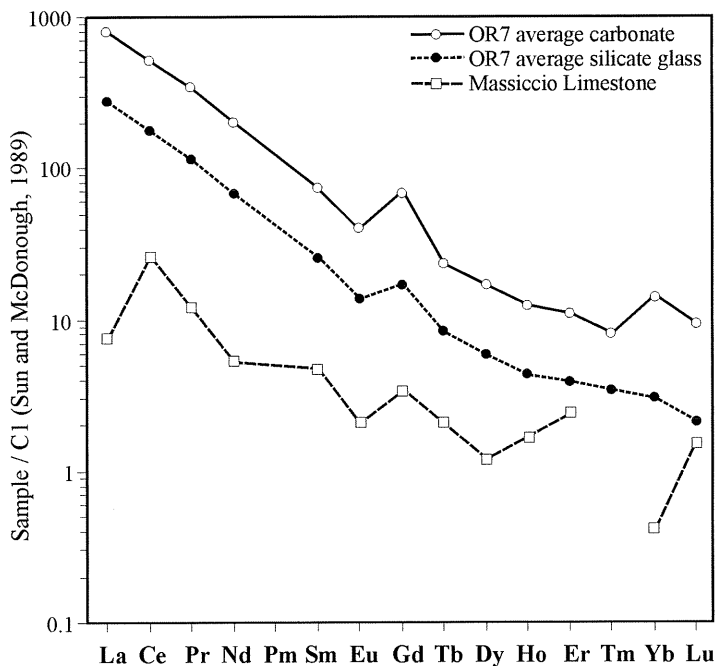


Fig. 4 – Average contents of REE (chondrite normalised, Sun and McDonough, 1989) in carbonate and foidite glass of the Oricola carbonatitic tuff (Stoppa *et al.*, in press), compared with “Massiccio” limestone. Mixing of the sedimentary “Massiccio” limestone with the silicate glass cannot produce the REE concentration measured in the carbonate fraction of the Oricola tuff deposit.

lapilli, up to a centimetre across, are cored by mantle minerals surrounded by successive rims of melilitite, carbonate-rich melilitite and, in some samples, carbonatite. The tuff matrix and thin, individual layers are carbonatitic. Calcite is found enclosed within individual crystals in mantle xenoliths, and as individual crystals and veinlets, and so provides direct evidence of the presence of mantle carbonate (Rosatelli, 2001). Thus, at this locality there is direct evidence that carbonate is present in the mantle and not a product of high-level assimilation of sedimentary limestone.

CONCLUSIONS

In this paper we counter the objections made by Peccerillo (2004) to our interpretation of the carbonate-rich volcanic rocks of a number of localities in central and southern Italy as carbonatites. The principal reasons are as follows:

1. The sedimentary limestone model proposed by Peccerillo (2004) is untenable, as demonstrated by the mixing model calculations that are the subject of the paper by Bailey, who shows clearly that the carbonatites cannot be produced by mixing of the silicate magmas with sedimentary limestone.

2. Sr isotope ratios point directly towards primary mantle melts produced by a strongly metasomatized and isotopically zoned source. Accumulation of high Rb/Sr minerals, such as phlogopite, account for the within outcrop isotope variation. Any hypothesis of assimilation of limestone cannot explain this isotope variation.

3. The $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ stable isotope data indicate that the Italian carbonatites are similar to a range of extrusive carbonatites from elsewhere. They also demonstrate that sedimentary limestones did not comprise one component in a mixing model.

4. Peccerillo (2004) objects to the fact that we have pointed out that the LILE, and notably the REE, the trace element contents of the calcite, and the presence of certain minerals are compatible with these rocks being carbonatites. He cites occurrences of rocks other than carbonatites in which these features can be found. Obviously, in isolation these features are not definitive, but they are characteristic and must be taken in geological/volcanological context. The observed chemistry is perfectly compatible with carbonatites.

5. The REE data for co-existing carbonate lapilli and silicate glass fragments in the Oricola carbonatites, which are much fresher than those of any of the other occurrences, indicate higher values in the carbonate fraction. This criterion is one that Peccerillo (2004) says he would expect in a carbonatite.

6. Calcite is present, as both individual grains and veinlets, within xenoliths originating at depths of about 70 km, in the Vulture carbonatitic tuffs. This points directly to the carbonatite originating in the mantle.

There is one more argument that we find persuasive, and this is the extraordinary similarity of the rock suite of the IUP province of Italy to that of western Uganda. These two localities comprise the principal 'kamafugite' igneous provinces on Earth. There are very few kamafugites elsewhere. Kamafugites are characterised by the presence of kalsilite, melilite and leucite and in both provinces are associated with carbonatite. The absence of any limestone in the basement of western Uganda, rules out any limestone involvement in the genesis of these rocks. It seems improbable, in the extreme, that the exceptionally rare and mineralogically exotic kamafugitic rocks were generated in two completely different ways, one involving mantle-derived carbonatites and the other high-level sedimentary limestones.

The presence of carbonatitic volcanism at San Venanzo, Cupaello, Polino, Oricola and Vulture must be considered an integral part of magmatic activity in Italy, and must therefore be taken into consideration in any model involving the petrogenesis of the Roman Province, and the overall geodynamic evolution of Italy.

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