

REVEALING HISTORIES OF EXPOSURE USING *IN SITU* PRODUCED ^{26}Al AND ^{10}Be IN LIBYAN DESERT GLASS

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ABSTRACT. We present the results of measurements of ^{26}Al and ^{10}Be produced *in situ* in 12 samples of Libyan Desert Glass by cosmic rays during the last ten million years. Based on the variability of the concentrations of ^{10}Be and of the $^{26}\text{Al}/^{10}\text{Be}$ ratios we measured, we conclude that individual fragments of glass have experienced different exposure histories, implying several major redistributions of the glass within the past 10^6 years. The ^{26}Al and ^{10}Be concentrations are inconsistent with the theoretical estimates of the rates of *in situ* production. We estimate minimum production rates of $70 \text{ atoms g}^{-1} \text{ yr}^{-1}$ and $10 \text{ atoms g}^{-1} \text{ yr}^{-1}$ for ^{26}Al and ^{10}Be , respectively, produced in quartz at sea level between $60\text{--}90^\circ$ latitude. Despite the present uncertainty in the rates of production, we feel that these results show clearly the effectiveness of *in situ* produced ^{26}Al and ^{10}Be in studying earth-surface processes.

INTRODUCTION

The measurement of ^{26}Al ($\tau_{1/2} = 0.7 \text{ Ma}$) and ^{10}Be ($\tau_{1/2} = 1.5 \text{ Ma}$) produced by the interaction of cosmic rays with oxygen and silicon in rocks may well provide a chronometer for studying weathering/erosion and exposure/burial histories occurring within the top few meters of the earth's surface (Hampel *et al*, 1975; Lal & Arnold, 1985). ^{26}Al is produced primarily from neutron reactions on ^{28}Si and ^{27}Al and to a lesser extent (ca 10% at the earth's surface) by muon capture in ^{28}Si . ^{10}Be is produced by neutron-induced spallation of O; and in silicates, not at all by muons. Within the top few meters of the earth's surface, the neutron-induced reactions predominate and the $^{26}\text{Al}/^{10}\text{Be}$ ratio is virtually independent of depth. In this region, the concentrations of ^{26}Al and ^{10}Be decrease by a factor of two for every 122 g cm^{-2} and 120 g cm^{-2} of shielding respectively (Nishiizumi *et al*, 1984a; Nishiizumi *et al*, 1984b; Lal & Arnold, 1985). At greater depths, a larger and larger proportion of the ^{26}Al is due to muon capture (muons are more penetrating than neutrons), so that the $^{26}\text{Al}/^{10}\text{Be}$ ratio increases, and the rate at which the ^{26}Al concentration decreases with depth decreases. A more detailed description of these dependences and their consequences for measuring rates of erosion will be dealt with elsewhere (Klein *et al*, ms in preparation).

In suitable materials such as quartz and high-silica glasses where the concentration of ^{27}Al is low, it is possible to determine exposure histories of 5 to 10g samples over the range of 10^4 to 10^7 years despite the rather low production rates, 10 to 100 $\text{atoms g}^{-1} \text{ yr}^{-1}$, of these isotopes (Nishiizumi *et al*, in press). This is possible because of the recent development of accelerator mass spectrometry which allows measurements on 10^6 atoms, at isotopic abundances as low as 10^{-15} (Brown, 1984; Wölfli, Polach & Andersen, 1984).

Although much can be learned from the buildup of a single cosmo-

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genic radioisotope in a sample, there are many advantages that accrue from being able to measure two or more. In particular, it is possible to 1) distinguish between short exposures to cosmic rays and long exposures followed by periods of burial; 2) place upper and lower limits on "burial" times within the past 10^7 years; 3) place an upper limit on the rate of erosion; and 4) estimate cumulative times of exposure on the surface of the earth.

For several years, in spite of the ability to measure naturally occurring ^{26}Al on the accelerator, (Thomas *et al*, 1983, Raisbeck *et al*, 1983, Nishizumi *et al*, 1984) the measurement of terrestrial ^{26}Al was hampered by a fixation on the idea that the ^{26}Al we wanted to measure was that produced by cosmic-ray-induced spallation occurring in the atmosphere (Raisbeck *et al*, 1983). In most samples this ^{26}Al is very difficult to detect, since systems which accumulate ^{26}Al from the atmosphere usually also collect ^{27}Al from other sources. The consequence is that the $^{26}\text{Al}/^{27}\text{Al}$ ratio is rarely high enough to measure. It was on the inspiration of one of our coworkers at the time, F Yiou, that we made the first measurements of ^{10}Be and ^{26}Al on a piece of Libyan Desert Glass (LDG) in order to see if we could detect the *in situ* production of these isotopes. LDG was attractive because 1) it is low in ^{27}Al , $<0.5\%$, making an ^{26}Al measurement feasible, 2) it is reported to be older than 28 Ma, and 3) it occurs on the surface of the Western Desert of Egypt; thus it seemed likely that measurable quantities of ^{26}Al and ^{10}Be would be present. It was with some surprise and a great deal of gratification that the first experiments on LDG were successful (Yiou *et al*, 1984). However, it took nearly two years to recognize the full significance of these measurements.

The research reported here represents the continuation of those original measurements in an effort to discover the systematics in the exposure/burial histories of LDG, and a short discussion of the significance of these measurements with respect to exploring weathering/erosion and exposure/burial histories in other more general geologic settings.

LIBYAN DESERT GLASS

Libyan Desert Glass is a unique, enigmatic, 98% SiO_2 glass found strewn between NNW-SSE trending linear dunes in the Sand Sea at 25° N Lat on the western border of Egypt. Since its "discovery" in 1932, though there are reports of its existence as early as 1846, it has been the object of at least 10 expeditions, and a vast quantity of speculation as to its origin. Fission-track dating places its age at 28.5×10^6 yr (Gentner, Storzer & Wagner, 1969). Recent estimates (Weeks, Underwood & Giegengack, 1984) place its present mass at 1.4×10^9 g, and infer that its original mass may have been 10,000 times greater. No glass fragment has been found that represents the edge of the original glass or a transition texture with its precursor material. Since pieces of glass are invariably found in the same geologic context as hydraulic equivalents, except when found as collections of artifacts, it has been concluded that all the glass was transported from its site of formation by earth-surface processes, primarily fluvial.

The origin of the glass is obscure. Chemically, it is similar to the Nubia Formation sandstone on which it is found. Gas inclusions have isotopic

compositions which are typically atmospheric, but may be the result of atmospheric diffusion into the glass during the 28 Ma since its formation. The low concentrations of water and volatiles and the relative paucity of bubbles suggest that the fusion process responsible for the formation of LDG involved high temperatures, or long times at moderate temperatures. The existence of flow lines, and the oxidation states and distribution of iron are also indicative of a sustained period at high temperature. Despite its glassy nature, cooling was probably slow.

Chemically, LDG is apparently of terrestrial origin, but no known physical process can account for its formation. There is no evidence of a meteorite-impact structure of sufficient magnitude near the strewn field; but more important in arguing against meteoritic origin is the fact that no other impact glasses resemble LDG in its high degree of internal equilibrium, its flow lines, or its high purity of silica. Formation by lightning seems improbable in light of fulgurites which display too short a history of high temperature, and volcanic processes seem unlikely, as they do not produce temperatures high enough. For the interested reader, the literature on Libyan Desert Glass and its origins can provide many a fine hour of diversion (Giegengack & Issawi, 1975; Barnes & Barnes, 1973; O'Keefe, 1963, 1976; Weeks, Underwood & Giegengack, 1984).

EXPERIMENTAL DETAILS

Twelve samples of LDG were analyzed for their ^{26}Al and ^{10}Be concentrations. Six samples were selected from a group collected from a small geographic region because they were suspected of having experienced similar recent exposure histories. Five samples were chosen from those collected during the 1981 field season to represent as wide a geographic area as possible. One piece of glass was sampled twice; one sample was taken from the "top," the other from the "bottom," where "top" and "bottom" were determined from the pattern of erosional etching and clinging sand.

Samples of 9 to 33g were cleaned mechanically in an ultrasonic cleaner and chemically by boiling in a mixture of concentrated aqua regia and dilute HF. They were dissolved in HF in the presence of 1.98mg of ^9Be carrier and converted to chloride using HCl. The final volume of each solution was brought to 100ml. Twenty ml were removed for ^{27}Al analysis by flame atomic-absorption (AA) spectrometry. Al and Be were extracted from the remaining solution by precipitation with NH_4OH and separated by cation exchange in 1.1N HCl. The Al was further purified by anion exchange. The Al and Be were converted to hydroxides and finally to oxides by baking at 950°C in a furnace. The ^{26}Al and ^{10}Be measurements on the accelerator were made using techniques described in Middleton *et al* (1983) and Klein, Middleton and Tang (1982). All chemistry and accelerator measurements were performed at the University of Pennsylvania.

RESULTS AND DISCUSSION

A summary of our results is given in Table 1. The uncertainty in the measurement of ^{26}Al and ^{10}Be are estimated to be 10% and 7%, respectively, unless otherwise noted. The uncertainties for the ^{10}Be and ^{26}Al

TABLE 1
Summary of measurements on Libyan Desert Glass

Sample	Al %	²⁶ Al (10 ⁶ atom/g)	¹⁰ Be (10 ⁶ atom/g)	²⁶ Al/ ¹⁰ Be	Exposure* (Ma)	Burial** (Ma)
N27 37m 1 (top)	0.350	5.62± 10%	2.30± 7%	2.44± 12%	1.4	1.4
N27 37m 1 (bottom)	0.260	4.60± 10%	2.40± 7%	1.86± 12%	1.9	1.8
N7W 28m 1	0.198	6.64± 10%	1.20± 7%	5.53± 12%	0.3	0.3
N29W 37m 1	0.691	8.59± 10%	1.30± 30%	6.61± 32%	0.3	~ 0.0
N85W 33m 1	0.284	9.64± 10%	1.60 ± 7%	6.02± 12%	0.4	0.09
S37E 50mA 1	0.578	10.7 ± 10%	1.90± 7%	5.64± 12%	0.5	0.2
S37E 50mB 1	0.224	7.80± 10%	2.00± 7%	3.90± 12%	0.8	0.8
4435	0.205	1.48± 25%	0.90± 7%	1.64± 27%	0.8	2.4
4415	0.184	2.69± 22%	2.60± 7%	1.03± 23%	4.3	2.4
4405	0.241	1.45± 20%	1.70± 7%	0.85± 21%	2.9	3.0
4560	0.290	11.7 ± 10%	6.0 ± 7%	1.95± 12%	> 7.5	1.0
4100.5	0.354	8.71± 10%	4.5 ± 7%	1.94± 12%	4.2	1.3
Yiou <i>et al</i> (1984)	0.420	9.2 ± 15%	3.1 ± 15%	2.70± 21%	1.8	1.1

Notes:

* The exposure time is the cumulative exposure experienced by a piece of glass during the past 10 Ma. This time is independent of the sequence of exposures and burials constituting the sample's history, but is modeled on exposures at the surface, and burials deep enough to reduce production substantially (>1.5m). These calculations use the production rates of ²⁶Al and ¹⁰Be determined in this paper, and consequently represent maximal times since the estimated rates are minima.

** Burial times are the shortest burials consistent with the ²⁶Al/¹⁰Be ratio and the ¹⁰Be concentration of the sample. Burial times, unlike exposure times, are not independent of the exposure-burial sequence. The minimum in the burial time results from a single exposure, followed by a single burial.

measurements were determined from long-time averages of replicate measurements, and do not reflect "counting statistics" unless the number of particles detected was small enough to make this the dominant uncertainty. Consequently, we use a rather conservative estimate of the precision of these measurements. For a summary of our long-term experience with ¹⁰Be, see Tera *et al* (in press).

The reproducibility and accuracy of the ²⁷Al measurements¹ were determined by preparing standards which ranged from 1 to 1000ppm, with solutions prepared from an NBS standard rock, BCR-1, and by replicating the AA measurements on several samples. Replicate measurements agreed to within 1%, as did the measurements on the standard solutions. Agreement between the AA results for the NBS standard and the accepted value were within 2.5%. Two blanks were measured, each yielding 0.2ppm. The overall accuracy of the ²⁷Al measurements was therefore taken to be 3%.

Implications for Libyan Desert Glass

The results in Table 1 have several implications with regard to the history of LDG.

¹ The measurements were done by Hinderberger, University of Missouri, Environmental Trace Substances Research Center, Columbia, Missouri.

1) Based on the fission-track determined age of the LDG of 28.5 Ma, none of the ^{10}Be or ^{26}Al measured in these samples could possibly have been “inherited” from the precursor material from which the glass was formed, or from an impacting body (meteorite) which may have been responsible for its formation.

2) Consequently, if we accept the fission-track age of the glass (or any age >10 Ma), the ^{26}Al and ^{10}Be must have been produced *in situ* in the glass after formation for the following reasons:

a) The $^{26}\text{Al}/^{10}\text{Be}$ ratio is >1 in every sample. This precludes a significant atmospheric contribution since the $^{26}\text{Al}/^{10}\text{Be}$ ratio from cosmogenic production in the atmosphere is $1/260$ (Raisbeck *et al.*, 1983) at production and can only decrease with decay. Thus, any contributions to these isotopes from the atmosphere have been virtually excluded for more than 10^7 years.

b) The great variation in the ratio of $^{26}\text{Al}/^{10}\text{Be}$ and in the concentrations of ^{26}Al and ^{10}Be between samples, even those found near one another, makes it unlikely that the production of these radioisotopes occurred before the individual pieces of glass were formed. The variation between pieces is better explained as resulting from the idiosyncratic histories of burial and exposure experienced by individual pieces.

c) Since the variations among pieces found near one another is considerable, size sorting in a fluvial environment is inferred to have occurred relatively recently, within the last 100,000 years. This is consistent with what is known of the climatic history of this area.

Other Implications

As already mentioned, there is a great deal of variation in the radioisotope concentrations measured in these samples. This is undoubtedly due to differences in the histories of burial and exposure experienced by individual pieces of glass, not surprising if we consider the fate of a small object adrift in a sea of sand for 28 Ma.

These differences have affected not only the amount of ^{26}Al and ^{10}Be contained in these samples, but also the ratio of $^{26}\text{Al}/^{10}\text{Be}$. The maximum $^{26}\text{Al}/^{10}\text{Be}$ ratio occurs during production and is highest at the inception of *in situ* production. For exposures $>ca\ 10^5$ yr, the ratio measurably decreases due to the longer lifetime of ^{10}Be . At saturation, *ie*, when the decay rate just equals the production rate, the ratio is about half (the ratio of the half-lives) the value at the inception of production. During burial, the ratio further decreases because of the faster rate of decay of ^{26}Al .

A plot of the $^{26}\text{Al}/^{10}\text{Be}$ ratio *vs* the concentration of either of the isotopes provides a useful aid to understanding these effects, (see Fig 1). The upper curve in the figure shows the $^{26}\text{Al}/^{10}\text{Be}$ ratio for a sample that starts with a zero concentration of ^{26}Al and ^{10}Be , as a function of its ^{10}Be concentration during exposure. It represents the maximum possible $^{26}\text{Al}/^{10}\text{Be}$ ratio for a particular ^{10}Be concentration for any combination of exposures and burials, and at a given set of production rates for ^{26}Al and ^{10}Be —in this case, those of Lal and Arnold (1985). The bottom curve shows how the $^{26}\text{Al}/^{10}\text{Be}$ ratio decreases as the ^{10}Be concentration decreases for a buried

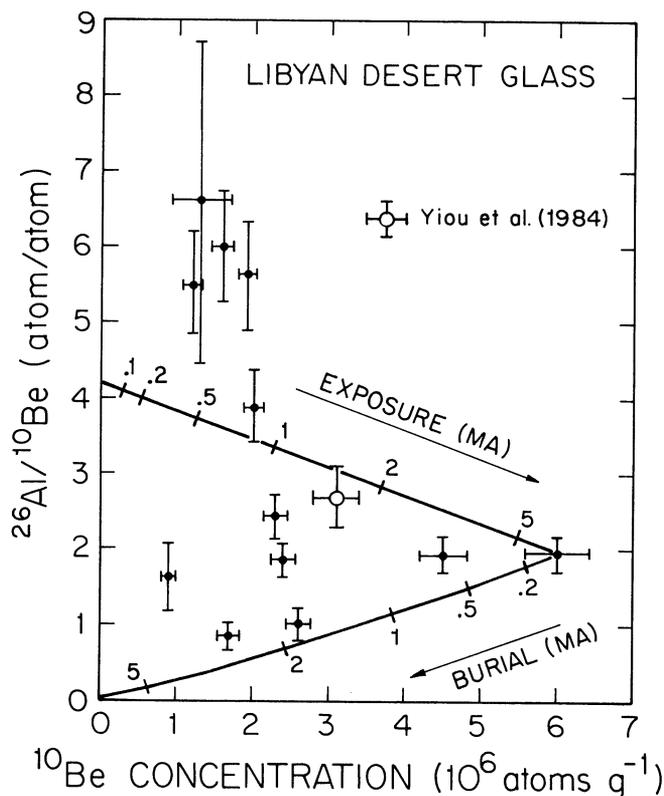


Fig 1. Measurements of Libyan Desert Glass plotted against the "exposure-burial triangle" based on the production rates of Lal and Arnold (1985). The region within the "triangle" represents the "allowed" values for the ratio of $^{26}\text{Al}/^{10}\text{Be}$ as a function of ^{10}Be concentration. The fact that several data points fall outside of this region is a strong indication that the rates of production for ^{26}Al and ^{10}Be predicted by Lal and Arnold are not correct. Note also that the time scale for exposure and burial are indicated on the figure.

sample that previously had been exposed long enough to achieve saturation. It defines the minimum $^{26}\text{Al}/^{10}\text{Be}$ concentration for a given ^{10}Be concentration. Only the area within these curves represents combinations of $^{26}\text{Al}/^{10}\text{Be}$ ratios and ^{10}Be concentrations possible for a given set of production rates. With appropriate sequences of exposures and burials, any value of the $^{26}\text{Al}/^{10}\text{Be}$ ratio and ^{10}Be concentration within the "triangle" of Figure 1 is possible. A detailed examination of these two parameters (Klein & Middleton, ms in preparation) allows for many comments about this history, including the cumulative exposure over the past 10 Ma and the minimum period of time the sample has been buried. By way of illustration, these properties are tabulated in Table 1.

What is immediately apparent from Figure 1 is that the production figures of Lal and Arnold (1985) are inconsistent with the values measured in the LDG. Figure 2 shows that the values calculated by Yokoyama, Reys and Guichard (1977) are also inconsistent with these data. To reconcile the pro-

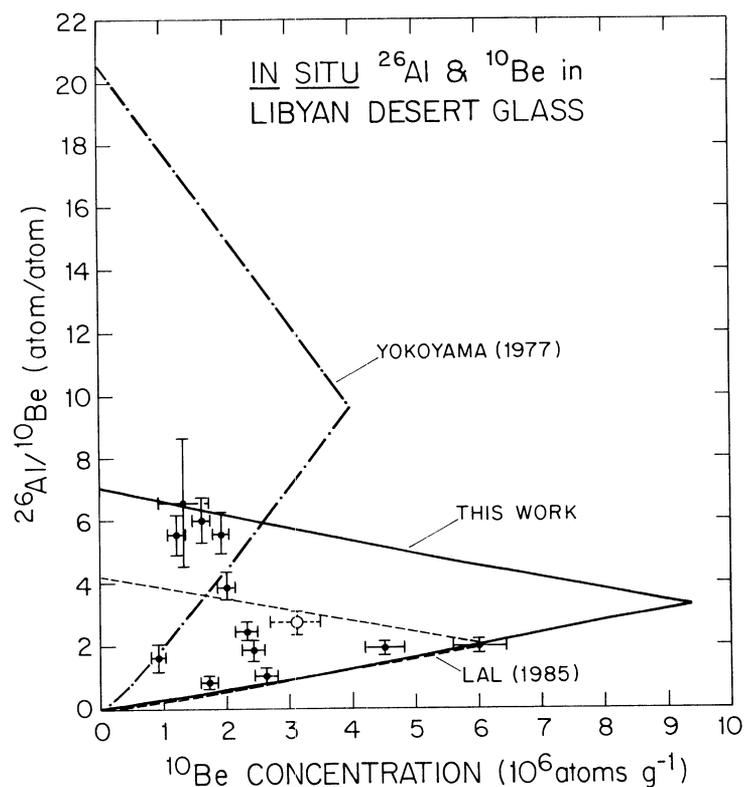


Fig 2. Similar to Figure 1, but including the "exposure-burial triangle" based on the predicated production rates of Yokoyama, Reys and Guichard (1977) as well. The solid triangle is based on the minimum rates of production for ^{26}Al and ^{10}Be consistent with the data presented in this paper. These rates are shown in Table 2.

duction figures of Lal and Arnold with these data, it is clear that their production rate of ^{26}Al must be increased in order to increase the "initial" ratio of $^{26}\text{Al}/^{10}\text{Be}$. It is also necessary to increase the rate of ^{10}Be production. The minimum rates necessary are listed in Table 2, as are the rates predicted by Lal and Arnold (1985) and Yokoyama, Reys and Guichard (1977). The "triangles" resulting from these production rates are plotted in Figure 2. This is a rather unsatisfactory way of determining the rates of production of cosmogenic radionuclides produced *in situ*; consequently, we are planning to measure these rates directly within the next 18 months.

CONCLUSIONS

The idea of using ^{26}Al and ^{10}Be produced *in situ* for measuring exposure and burial histories at the surface of the earth is rather new and results from a departure in thinking: from considering cosmogenic production on earth occurring in the atmosphere, to its occurring in rocks. This change in perspective is significant for several reasons.

TABLE 2
Estimates of *in situ* production rates of ^{26}Al and ^{10}Be in quartz

	Production rate* (Atoms $\text{g}^{-1} \text{yr}^{-1}$)		$^{26}\text{Al}/^{10}\text{Be}$		^{10}Be at saturation in LDG † (10^6 atoms g^{-1})
	^{26}Al	^{10}Be	Production	Saturation**	
Yokoyama, Reyss and Guichard‡ (1977)	87.4	4.23	20.7	9.65	4.0
Lal and Arnold(1985)	27.5	6.5	4.23	1.97	6.1
This work§	70	10.	7.0	3.27	9.35

Notes:

* For sea level and 60–90° lat.

** Half-lives: $^{26}\text{Al} = 0.7 \text{ Ma}$, $^{10}\text{Be} = 1.5 \text{ Ma}$ † Adjusted for 500m elevation (increases production by $\sim 1.44\text{x}$), and 25° N Lat (decreases production by 0.3x). Elevation and latitude factors are those in Yokoyama. Effect of latitude may be less than assumed because of wandering geomagnetic poles.‡ Values are actually for granite (73% SiO_2) for which ^{26}Al production may be slightly underestimated.§ Based on Figure 2, these numbers represent minima for the rates of production and the $^{26}\text{Al}/^{10}\text{Be}$ ratio as described in the text.

1) The major problem in measuring ^{26}Al in terrestrial systems is the low $^{26}\text{Al}/^{27}\text{Al}$ ratio, which rarely is greater than 10^{-15} for ^{26}Al produced in the atmosphere (Raisbeck *et al*, 1983). This is a result of the high abundance of ^{27}Al and the relatively low rate of production of ^{26}Al , due to the lack of suitable spallation targets (only Ar is heavier than Al, and it constitutes only 1% of the atmosphere).

2) By looking at the ^{26}Al produced within a rock, it is possible to find systems which are low in ^{27}Al (<300ppm) but with sufficient target materials for reasonable production rates of ^{26}Al and ^{10}Be . In fact, nearly 80% of all ^{26}Al produced on earth is produced in rocks; and although less than 1% of all terrestrial ^{10}Be is produced at the earth's surface, the low abundance of ^{9}Be makes the measurement of ^{10}Be in rocks quite straightforward.

3) *In situ* production also avoids the problems associated with the transport of radioisotopes from the site of production to the system under study. Chemical differentiation often results in $^{26}\text{Al}/^{10}\text{Be}$ ratios being determined geochemically and not by rates of production or decay.

4) Finally, *in situ* produced radionuclides allow us to study systems completely different from those accessible through the measurement of atmospherically produced ^{26}Al and ^{10}Be . In particular, weathering/erosional and exposure/burial histories of rocks can be examined. The history of soils can be approached from a radiochronologic perspective independent of that provided by measuring the build-up of atmospheric ^{10}Be , *eg.* by studying the history of quartz grains contained within the soil. In short, measurement of ^{26}Al and ^{10}Be produced *in situ* offers a means of studying earth-surface processes not readily approachable using any other technique now available.

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