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## Diffuse reflectance of foams

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### ABSTRACT

There have been few studies of the optical properties of foams but the results of these investigations are of particular relevance to current discussions about climate restoration by means of oceanic foam because they indicate the influence of foam raft number on albedo. The diffuse reflectance of foams prepared from eight surfactants was measured in a  $0^\circ/45^\circ$  geometry with a collimated, solar simulation light source and a photodiode by reference to barium sulphate white standards. Reflectance, or albedo, in the range of 0.5 to 0.59 was recorded for foams prepared with sodium dodecyl sulphate (SDS), methyl cellulose, and TWEEN 20<sup>TM</sup>. Perfluoro-surfactants produced more stable foams with reflectance over 60%. The apparatus was adjusted to the  $45^\circ/45^\circ$  configuration (specular reflection) and used to test the suggestion that a foam can be treated, for purpose of approximation, as a series of horizontal planar reflective layers in which reflectance reaches a limiting value. The resulting geometric series, when summed over an infinite number of layers yields an albedo of 0.62 to 0.65 when the reflectance of individual layers of which the model is comprised was varied over the range 0.01 to 0.1 respectively. This was tested experimentally using eight sheets of 500  $\mu\text{m}$  thick glass separated by 18  $\mu\text{m}$  thick glass cover slides in  $45^\circ/45^\circ$  mode and the results agreed to within less than 9% with the model calculation. The insensitivity to layer reflectance and the fact that limiting reflectance for the assembly is reached after about 20 layers provides a simple working model to assess the dependence of reflectance on thickness and cell diameter.

### 1. Introduction

There are socio-economic impediments to reducing carbon dioxide emissions by the three principle interventions of demand reduction, conversion to non-fossil fuels, and carbon capture and it is estimated that in the next 30 years energy demand will nearly double (Stern, 2007). The 2009 United Nations Climate Change Conference (Blesl et al., 2010) encouraged non-fossil energy resources such as wind energy, geothermal energy, biomass, and photovoltaics but these, as with structural changes to the energy landscape such as hydrogen economy, have technical lead times and market penetration rates that prevent them making significant short-term contributions. Similarly, carbon capture technologies

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are linked to power station replacement rates. Concerns about the health (Costello et al., 2009) and security (Sullivan et al., 2007) issues of climate change have been expertly analyzed and raise concerns in terms of water stress, food shortage, migration, disease, and security. A fourth class of intervention, climate engineering, more appropriately named climate restoration, therefore appears, albeit somewhat uncomfortably, on the agenda. The proposal by both US and UK scientists to use oceanic foams (Evans et al., 2010; Seitz, 2011) to increase global albedo has the advantage that it enhances a pre-existing natural processes: ocean whitecaps and ice sheets both participate in the planetary energy budget and are influenced by the 70% of the Earth that is covered by oceans. This intervention is easily arrested, potentially uses simple engineering strategies (compared to proposals for space parasols, for example), and can avoid large-scale deployment of sulfur suggested for stratospheric aerosols (Keith, 2000; Shepherd, 2009). Indeed, as climate change progresses, the deployment of foams is also proposed for conservation of freshwater by reducing peak and average water temperature (Seitz, 2011).<sup>3</sup>

Oceanic surface albedo (OSA) is defined as ‘the ratio of upward to downward irradiance at the air-sea boundary’ and it is estimated to be approximately 5% (Payne, 1972). It depends on solar zenith angle (SZA), aerosol optical depth, wind speed, and chlorophyll concentration (Jin et al., 2004). The SZA (angle between the sun’s rays and the local zenith) affects the OSA due to the large optical path that is seen by photons incident at large SZA, increasing ocean albedo at sunrise and sunset (Jin et al., 2004).

An increase in the wind speed results in the formation of whitecaps on the ocean surface. Maul and Gordon (1975) using measurements from LANDSAT-1, observed that whitecaps on the surface of the ocean at the Gulf of Mexico resulted in an increase of the outgoing radiation that escapes the upper level of the atmosphere. This was the first indication that the OSA could actually be affected by the presence of whitecaps.

A few studies have been conducted to determine foam reflectivity. In 1982, Whitlock and colleagues (1982) reported a laboratory experiment to measure the optical properties of foam using distilled water in a tank and adding high pressure air to produce foam patches. Depending on the adjustments, they produced bubble monolayers and one multiple layer of 0.1 m thickness. Spectral measurements resulted in a value of whitecap albedo of 50% with the use of a polynomial expression of the reflectance. Foam reflectance is a function of the wavelength and is affected by the thickness of the foam patch, by the salinity, if any, of the water which influences the whitecap bubble spectrum, and potentially by surfactant monolayers on the whitecap bubbles. For wavelengths above 0.9  $\mu\text{m}$ , a decrease in reflection of 10% was observed due to absorption by water while the reflectance of the bubble monolayer was found to be 20% of that of the multiple layers.

In 1972, Payne (1972) used pyranometers situated on a seagoing platform to measure upward and downward irradiance over a four-month period resulting also in a value of

3. The authors are most grateful to a reviewer who reflects that the optical benefits of foams were proposed in a humorous but illuminating column by David Jones in *Nature* (1990) 348, 396.

50% for the whitecap albedo. In 1996, Frouin and colleagues (1996) conducted another experiment taking in situ measurements at the Scripps Institution of Oceanography Pier, La Jolla, California, which resulted in the same value, approximately 50% for the albedo of spilling wave crests. Apart from this value, they also observed that there is a 40% reduction in the reflectance for wavelengths greater than 0.9  $\mu\text{m}$ , which is somewhat different to Whitlock's findings possibly attributable to the use of seawater instead of distilled water and the use of surfactant.

In 1984, Koepke (1984) reported photographic analysis of 13 whitecaps at 0.5 s intervals to take into account the increase in area of a whitecap as it aged and a corresponding decrease in reflectance. An efficiency factor based on area and reflectance was then applied to Whitlock's reflectance of 55% to give an effective reflectance of  $(22 \pm 11)\%$  in the visible spectral range averaged over whitecap lifetime.

Ocean foam may also release two types of seawater droplet into the atmosphere. Film drops typically about 100 nm in diameter result from rupture of the bubble film while jet drops are propelled by the closure of the bowl under surface tension. Some of these drops loft, dry, and act as cloud condensation nuclei (Jaenicke, 1993) increasing the albedo of marine stratocumulus clouds. In this way ocean foams could have a double effect on the radiation balance addressing both overcast and cloudless conditions.

In general, foams consist of at least 95% gas and 5% liquid and the cells are spherical at low gas content (Pugh, 1996) developing into polyhedra at low water contents depending on the type of surfactant and its concentration in solution. It is not possible to produce stable foam in pure liquid (Kitchener, 1959) but in seawater, dissolved organic species of biological origin act as foaming agents. Biodegradable additives might be used in the low chlorophyll ocean regions where there is limited marine life and hence where the effect on air-sea gas transfer and aquatic life would be minimal. A foam starts to decay after formation due to thinning of the film, liquid drainage, and gas diffusion between bubbles of different sizes (Garrett, 1993); foam persistence is strongly influenced by the size and size distribution of bubbles.

This work examines the optical properties of foam prepared from different surfactants, most of which are obviously not suitable for deployment in oceans but which provide sufficient stability for reliable measurements in order to assess the range of reflectance that is likely to be obtainable from aqueous foams. Clearly the eventual selection of biodegradable surfactants will require studies in seawater where, even without added surfactants, bubble size distributions can differ from those in fresh-water. It also attempts to describe both mathematically and experimentally the conjecture of Stabeno and Monahan (1986) that the reflectance of a foam depends on the effective number of rafts and not on the size of the bubbles contained in it. Thus if a breaking wave entrains a given volume of air, the resulting whitecap will have fewer bubble rafts if the bubbles are larger and will present lower reflectance. This convenient simplification provides a criterion for the thickness of foam based on the number of bubble rafts needed to achieve a given reflectance. It is of course possible that the Stabeno-Monahan rule begins to break down at very low bubble

Table 1. Surfactants and concentrations of active species used for the preparation of foam samples.

Surfactant	Conc <sup>n</sup> /gl <sup>-1</sup>	Active constituent
TWEEN 20	20	
SDS	10	
Methocel	2	
Chemguard S-559-100	2	100%
Chemguard FS-221	2	40%
Chemguard FS-818-6	2	36%
Chemguard FP-5103	2	38%
Chemguard FS-909-7	2	40%

diameters ( $\sim 100 \mu\text{m}$ ) so there is considerable scope for refining the optical studies of foams.

## 2. Experimental details

For diffuse reflectance measurements, white standard samples of  $\text{BaSO}_4$  99% (Sigma Aldrich, Gillingham, Dorset, UK) and polytetrafluoroethylene (ptfe) in the form of a 12 mm thick plate abraded on 600 SiC paper were used for calibration. Barium sulphate was also used in Whitlock's research on foams (1982) and the absolute  $\text{BaSO}_4$  reflectance of 98.7% as determined by Grum and Wightman (1977) was used as the multiplier. Two disc samples of  $\text{BaSO}_4$  each of diameter 80 mm were prepared by slip casting from aqueous suspension.

Laser methods are not ideal for measuring the reflectivity of foams because of the need to average over a large number of bubbles. The integrating sphere needs modification for liquids leaving the  $0^\circ/45^\circ$  arrangement as an ideal and accepted geometry for diffuse reflectance measurement (Springsteen, 1999). A 63 mm diameter collimator fitted with a 12V, 55W natural daylight MR16 Halogen Display Lamp (EiKO UK, Leixlip, Co. Kildare, Ireland) and planoconvex lens was used to provide an illuminated field to even out the effect of large bubbles. The collimator was set vertically and a tube containing a type BPW 41 N photodiode shunted with a  $470 \Omega$  resistor was arranged at an angle of  $45^\circ$  in such a way that the ray diagram constructed for the rays entering the detector tube originated from an ellipse well within the 63 mm diameter illuminated disc. The photodiode was connected to a Thurlby Thandar 1705 multimeter and measurements were made in a dark room.

The surfactants used were hydroxy propyl methylcellulose (Methocel; Dow, E50FG, E464), polyoxyethylene (20) sorbitan monolaurate (TWEEN 20; Sigma Aldrich, CAS 9005-64-5), sodium dodecyl sulphate (SDS; Sigma-Aldrich, CAS 151-21-3), Chemguard<sup>TM</sup> S-559-100 an ethoxylated non-ionic fluorosurfactant, Chemguard<sup>TM</sup> FS-221 and Chemguard<sup>TM</sup> FS-818-6 both non-ionic fluorosurfactants, Chemguard<sup>TM</sup> FP-5103 which is an anionic poly-perfluoroalkylated polyamino acid, and Chemguard<sup>TM</sup> FS-909-7. The concentrations are given in Table 1.

Foam samples were prepared by vigorously stirring 300 ml solution of surfactant in distilled water for 30 s. The foam was transferred in a glass Petri dish of diameter 90 mm, painted matte black providing a foam depth of 10.5 mm. Foam cell diameters, as deduced from photographs, were in the range of 0.5 to 0.7 mm ensuring at least 15 layers at the start of the experiment but keeping foam depth sufficiently low that the optical consequences of coarsening could be observed in reasonable timescales.

Stabeno and Monahan (1986) suggest a very convenient way of assessing foam reflectance which proposes that the optical reflectance is independent of the bubble size but depends solely on the number of bubble rafts. Their argument is that if the foam is modeled as an assembly of rectangular units, the ray diagram indicates that vertical walls have little effect on reflectance and only horizontal components contribute. After a specific number of bubble rafts, the reflectance should remain the same, regardless of the number added. This simplification is of considerable relevance to the implementation of oceanic foams, indicating the minimum thickness that needs to be obtained in terms of bubble diameter. They point out that a similar situation prevails with snow: the albedo increases up to a snow depth of 127 mm but further depth produces no increase in albedo.

In the experiment, glass slides were used to represent the rafts and a theoretical model based on a geometrical series was derived for comparison with the experimentally measured reflectance. A  $45^\circ/45^\circ$  geometry was used for measurement of specular reflectance. The detector tube was of diameter 36 mm and length 120 mm with a photodiode attached at the center and was internally blackened. Square glass plates of  $505\ \mu\text{m}$  thickness were used, the spacers inserted at corners were  $18\ \mu\text{m}$  thick glass cover slides and the top glass plate was kept in the optical plane as the number of plates was increased. The lower surface was matte black.

According to this approach, as the number of layers of foam increases, so does the reflectance, until the foam thickness reaches a value above which further increases in thickness make no appreciable difference to the reflectance. As more glass plates are inserted, reflectance measured by the detector is expected to increase due to the fact that light is reflected from each surface of glass and the rate of increase is expected to decrease. Since this is a specular reflectance measurement, the 'white' standard was a mirror and a value of reflectance of 0.9 was used (Twidle, 1957).

### 3. Results and discussion

#### a. Bulk foam reflectance

Three photodiodes were used to measure the reflected intensity in terms of voltage from two  $\text{BaSO}_4$  discs and gave means of 35.50 mV and 34.91 mV with coefficients of variation between the three diodes of 1.2% and 0.8% respectively. The PTFE sample gave 35.70 mV with coefficient of variation of 1%. Diffuse reflectance measurements were referenced to  $\text{BaSO}_4$  and multiplied by the absolute  $\text{BaSO}_4$  reflectance of 98.7% determined by Grum and Wightman (1977).

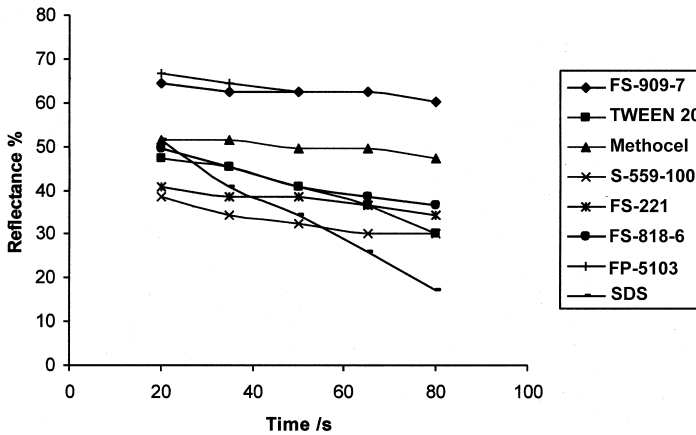


Figure 1. Reflectance of foams as a function of time.

Table 2. Reflectance of foams at formation (by extrapolation) and after 20 s (measured).

Foam type	Reflectance %	
	0 s	20 s
SDS	59*	51.6
TWEEN 20	50	47.3
Methocel	53	47.3
S-559-100	43	38.7
FS-221	43	40.9
FS-818-6	55	49.5
FP-5103	68	66.7
FS-909-7	66	64.5

\* Neglecting the upturn of the curve at 20 s.

The reflectance of foamed solutions for those compositions in Table 1 was measured over a period of 80 s. The first measurement was taken at 20 s, this being the time needed to set the foam in position and obtain a reliable reading. The reflectance decay associated with foam coarsening and drainage over this period is plotted in Figure 1 and extrapolation is used to obtain the value at  $t = 0$  (Table 2).

The reflectance values obtained for the SDS, TWEEN 20 and Methocel foams are very close to those recorded in previous laboratory work by Whitlock (1982) and in ocean studies by Frouin and colleagues (1996) and by Payne (1972). Two of the fluorinated surfactants FP 5103 and FS 9097 produce higher extrapolated initial reflectance and the foam is retained for long periods.

*b. Simulation of foam reflectance*

If the influence of incidence angle on reflectivity is taken into account, the reflectance of multiple parallel surfaces can be found exactly using the methods described by Heavens (1955) but when the reflectivity of the surface under consideration is low (in this case approximately 0.1) the overall reflectance turns out to be remarkably insensitive to reflectivity. The refractive index of air is 1.0008 and the refractive index of the glass (263TDesag, UQG Optics, Cambridge, UK) was 1.535. Absorption in the 500  $\mu\text{m}$  thick glass plates was neglected based on data for transmittance (Kaye and Laby, 1995). The value of  $r$  for a single surface was obtained by averaging the s- and p- polarized reflection coefficients from Fresnel's equation ( $r = 0.0550$ ) and then finding  $R$  for both surfaces from  $R = 2r/(1+r)$  which accounts for the reflections within the film, to give  $R = 0.104$  for each glass plate.

The unmodified ocean surface also has a refractive index of approximately 0.05 and by examining the geometric series for multiple reflections one term at a time, we can see how higher albedo can be developed even by modest foams. This makes it easier to judge how the reflectance of a whitecap changes as it both coarsens and thins out in terms of the number of reflective layers remaining intact. Let  $A$  be the intensity of the incident beam on the first glass plate. Then, the light that passes through the plate to fall on the second glass is  $A(1-R)$ , remembering that  $R$  here includes light reflected from *both* sides of the glass. At each plate transmission, a factor  $(1-R)$  is applied and at each plate reflection a factor  $R$  is applied to the ray diagram in Figure 2. For a total of  $N$  plates where  $n$  is an individual plate numbered from the top, the light escaping from the upper surface as a result of primary reflections produces the series shown in Figure 2a which has the sum:

$${}^1R_n = R \sum_{n=1}^{n=N} (1-R)^{2(n-1)} \quad (1)$$

The secondary reflections are transmitted downward but the tertiary reflections also emerge upward and a similar ray diagram produces the following power series:

$${}^3R_n = R^3 \left[ (1-R)^2 + 3(1-R)^4 + 6(1-R)^6 + \dots + \frac{n(n+1)}{2} (1-R)^{2n} \right] \quad (2)$$

It soon becomes clear that since the fifth reflections have a coefficient of  $10^{-5}$  only the primary and tertiary reflections make significant contributions to reflectance.

The summation to infinity for the primary reflections is  $\frac{1}{2-R}$  giving, for an infinite number of plates  ${}^1R_{n=\infty} = 0.53$ . This value is insensitive to the individual plate reflectance  $R$ . Thus for  $R = 0.01$  to  $0.1$ ,  ${}^1R_{n=\infty}$  varies from only 0.50 to 0.53. The tertiary reflections sum to infinity with  ${}^3R_{n=\infty} = \frac{(R-1)^2}{(2-R)^3}$  and so  ${}^3R_{n=\infty} = 0.12$  making a maximum of 0.65 at  $R = 0.104$ , very close to the value of approximately 0.62 estimated by Stabeno and Monahan (Fig. 2, 1986) which of course was based on the refractive index of water at 1.33.

Clearly the reflectance of a soap film is likely to be different from that of a glass plate and excellent work has been done in relating the thickness of a soap film to its reflectance by



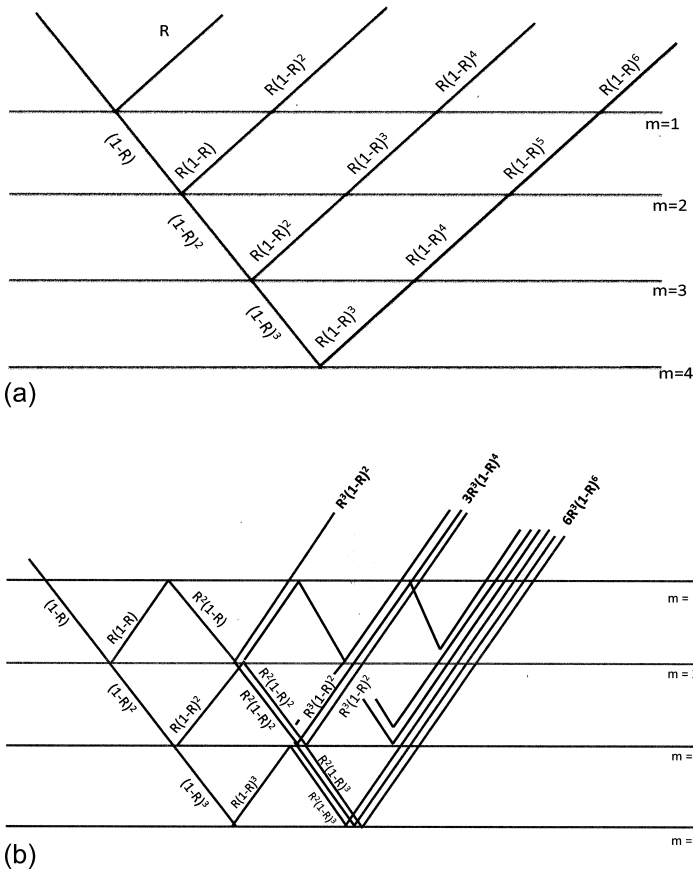


Figure 2. Ray diagram for (a) primary reflections and (b) tertiary reflections. (Note that the last ray bundle includes rays reflected from levels 2,1,4; 3,1,3; 4,1,2; 3,2,4; 4,2,3; and 4,3,4).

modifying Airy’s formula (Rijnbout, 1970) distinguishing the two surfactant zones each of the order of 1.5 nm and the aqueous central region of 2 to 5 nm. This approach offers some explanation for the much higher reflectance of the perfluoro surfactants. Replacing hydrogen in the alkyl chain with fluorine increases the electron density and is likely to change the refractive index, which analogously contributes to PTFE being a strongly scattering material (Li et al., 2008) often used for reflectance standards (Kuszczynska and Pietrzykowski, 2001).

Table 3 indicates how the reflectance accumulates from successive plates and orders of reflection. Tertiary reflections contribute less to overall reflectance, each being always less than 0.7% with a sum to infinity of 12%. The experimental results for specular reflection are consistently lower than theoretical because of scattering effects at the glass plates despite careful cleaning. In addition, error is incurred from the photodiode measurement described

Table 3. Contributions to total reflectance from each layer (n) for primary and tertiary reflections, total calculated reflectance, and experimental (expt.) results.

n	R (primary)	R (tertiary)	Total R	Expt.	Deviation between expt. and theoretical prediction (%)
1	0.1040	0.0009	0.105	0.105	0
2	0.0835	0.0022	0.191	0.188	-1.6
3	0.0670	0.0035	0.261	0.251	-3.8
4	0.0538	0.0047	0.319	0.298	-6.6
5	0.0432	0.0056	0.368	0.346	-6.0
6	0.0347	0.0063	0.409	0.377	-7.8
7	0.0278	0.0067	0.444	0.404	-9.0
8	0.0224	0.0070	0.473	0.435	-8.0
9	0.0179	0.0070	0.498	-	-
10	0.0144	0.0069	0.519	-	-
11	0.0116	0.0066	0.538	-	-
12	0.0093	0.0063	0.554	-	-
13	0.0075	0.0059	0.567	-	-

above and from the inexactitude of angle setting. The insensitivity to the reflectivity of each surface and the fact that this simplified model produces both theoretical and experimental results that are close to measured foam reflectances under both laboratory and ocean conditions is both a tribute to the perception of Stabeno and Monahan (1986) and provides a simple tool to decide on the conditions, thickness, and bubble size for artificial foams for ocean albedo modification as well as an assessment of how albedo is expected to decay as a foam coarsens.

#### 4. Conclusions

Measurements of diffuse reflectance in the visible region obtained using a  $0^\circ/45^\circ$  optical arrangement for surfactant SDS in distilled water and referenced to a barium sulphate standard resulted in values of 52–59%, comparable to previous measurements conducted both in laboratories and in ocean conditions notably by Payne (1972), Whitlock (1982), and Frouin and colleagues (1996). Foams prepared with other surfactants had reflectances varying from 43% to 68%. The temporal deterioration of reflectance indicated that the SDS foam reflectance had fallen to 17% after one minute but the reflectances of two flourosurfactants had only decayed from above 60% to 58% in this same time interval. This of course confirms Koepke's suggestion that reflectance for ocean whitecaps be averaged over the duration of their lives. Even though foam stability can be modified, all aqueous foams have a limited lifetime. Stabeno and Monahan's proposal for estimating foam reflectance by considering the horizontal planes was tested with glass plates in  $45^\circ/45^\circ$  optical configuration for specular reflectance and the corresponding geometric series for multiple reflections was tabulated. This shows that 10 to 12 layers are needed to reach 50% reflectance and the

experimental results agreed with the progression within 0 to 9% error. Beyond this number of rafts, there is reduced contribution to the reflectance. The geometric series was summed to infinity to give an overall reflectance of 0.65. This theory treats the foam as a grid of bubble rafts and suggests that its reflectance depends mainly on the number of rafts. This considerably simplifies the technological assessment of interventions to increase average global albedo by the use of oceanic bubble rafts. Furthermore, it was shown that as the reflectance of an individual layer varies by a factor of 10 from 0.01 to 0.1, the reflectivity of an infinite assembly changes only from 0.62 to 0.65.

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