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BACKSCATTER AND TRAMSMISSION OF AEROSOL AT UV

THROUGH MIDDLE IR WAVELENGTHS



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S.G. JENNINGS

(Principal Investigator) University College Galway.

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5th Interim Report

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Backscatter and Transmission of Aerosol at UV through middle IR wavelengths

This 5th interim report describes:

- (i) Backscatter and transmission of obscuring aerosol of Asbury carbon graphite tlakes at UV through middle IR wavelengths.
- (ii) Measurement of biological aerosol (pollen and spores) using a continuous Burkard spore sampler at the Mace Head field station, on the west coast of Ireland.
- (iii) Measurement of biological aerosol (pollen and spores) using an array of passive samplers, at seven sites in western Ireland.

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#### Backscatter and transmission of obscuring aerosol of Asbury carbon graphite flakes at UV through middle IR wavelengths.

Preliminary experiments were carried out to investigate the aerosol of carbon graphite flakes (Asbury M260 # 4676) generated in the aerosol chamber. A continuous power He-Ne laser (632.8 nm wavelength) was directed through the 1 m length aerosol chamber shown in Figure 1. Filtered air jets were blown at an angle of 45° across the  $3/8^{\circ}$  diameter entrance and exit holes for the laser in order to contain the aerosol within the chamber. Another filtered air jet blew in the carbon graphite aerosol horizontally a distance 3 cm below the top of the chamber which then was allowed to fall 21 cm under gravity to the laser beam path. The aerosol was collected in a  $1.3 \text{ m}^3$  air-tight Velostat conducting bag. The carbon graphite aerosol was investigated for different aerosol clouds by measuring the extinction of the He-Ne laser as a function of time using a photodiode detector and readout meter. The decay in extinction coefficient,  $\sigma_{e}$ , with time for carbon graphite aerosol clouds is shown in Figure 2. The initial decay time constant was between 2.7 and 4.7 minutes followed by a slower decay. This is consistent with the given deposition velocities of 0.120 cm s<sup>-1</sup> and 0.069 cm s<sup>-1</sup> which yield deposition times of 2.9 and 5.1 minutes respectively in the present set-up.

Laboratory measurements of simultaneous backscatter and transmission for obscuring aerosol of carbon graphite flakes (Asbury M260 #4676) were made using the experimental arrangement shown in Figure 3. A continuum Surelite Nd: YAG pulsed laser was used at its fundamental 1064 nm wavelength and harmonic wavelengths of 532, 355 and 266 nm. The same 1 m aerosol chamber was used as in the preliminary experiment apart from the requirement of larger entrance and exit holes (3.2 and 2.5 cm diameter respectively) for the laser, the holes being plugged when not in use. The larger holes are required for the following reasons:

(i) because of the larger diameter of the Nd: YAG laser beam (6 mm diameter)

(ii) in order to eliminate edge effects from the 8 mm diameter hole in the mirror

(iii) in order to obtain all the backscattered signal from the aerosol over the solid angle reflected by the mirror onto the detector (see Figure 4).

The laser beam passed through a hole at 45° through an appropriate wavelength matched high reflectance mirror (> 99.5% reflectance). The backscattered signal was reflected onto the detector from immediately below the hole in the mirror so that the backscattered signal was as close to 180° as possible to the main beam, in this case between 0.3 and 2° away from 180°. The extinction and backscattered signals were measured simultaneously by Molectron pyroelectric detectors J50 + JBX and J4-09 respectively. All measurements were an average of 10 pulses and made during cloud decay conditions. The cloud dissipated in 3 or 4 minutes as before, but the longer aerosol decay was not observed due to the aerosol being blown away by incoming air jets. The values for volume extinction coefficient,  $\sigma_e$ , and volume backscattere coefficient  $\sigma_h$ , were derived from the extinction and backscattered signals.

Great care was taken to reduce the background signal as much as possible because of the low values of backscattered signal. When the J4-09 detector was displaced, the background noise was not measurable ( $< 5 \times 10^{-9}$ J). When the J4-09 detector was aligned to measure the backscattered signal a significant signal ( $\sim 1 \times 10^{-7}$ J depending on wavelength) was observed due to reflection from the far end of the chamber and detector. This background signal, I<sub>b</sub>, was assumed to be attenuated by the aerosol by an amount given by

$$I_{b} = I_{bo} \exp\left(-2\sigma_{e}L\right) \tag{1}$$

where  $I_{bo}$  is the background signal with no aerosol and L is the chamber length.

In all measurements  $I_b$  was subtracted from the measured backscattered signal to give the true signal due to the aerosol.

The distribution of the carbon graphite flakes (Asbury M260 # 4676) was analysed using the given data for mass distribution as a function of diameter. The number distribution n(r) was obtained assuming the particles were spherical. Plots of  $dn(r)/d \log_{10} r$  against radius, r, and of mass distribution  $dm(r)/d\log_{10} r$  against r are shown in Figures 5 and 6 respectively. A lognormal distribution with geometric mean radius,  $r_g$ , of 1.25  $\mu$ m and geometric volume radius,  $r_v$ , of 2.62  $\mu$ m was obtained. Figure 7 shows that radius against cumulative mass percentage on log probability paper is linear up to 90% mass and gives a value of  $r_v$  of 2.65  $\mu$ m at the 50% cumulative mass reading.

The standard Mie values for extinction,  $Q_e$  and backscatter efficiency, G, (assuming spheres) for the carbon graphite flakes (Asbury M260 # 4676) at 1064, 532, 355 and 266 nm wavelengths,  $\lambda$ , as a function of particle size parameter x (where  $x = 2\pi r/\lambda$ ) are shown in Figures 8 and 9 respectively. The values of refractive index for carbon graphite flakes for each wavelength are given in Table 1.

The carbon graphite aerosol consisted of a polydispersion of particles with a lognormal distribution with sizes ranging from 0.75 to over 10  $\mu$ m radius with a numerical mean radius of 1.25  $\mu$ m. In each case, the size parameter was such that the backscatter and extinction values fall in the relatively constant region leading to asymptotic values of G and Q<sub>e</sub>.

In this case the backscatter gain,  $G_{\infty}$  is given by

$$G_{-} = \frac{(n-1)^2 + k^2}{(n+1)^2 + k^2}$$
(2)

where n and k and the real and imaginary indices of refraction.

The asymptotic values of  $Q_e$  and G lead to the following appealingly simple theoretical form independent of size distribution given by

$$\frac{\sigma_{e}}{\sigma_{b}} = \frac{4\pi Q_{e}}{G_{-}} \tag{3}$$

The theoretical Mie values for  $\sigma_e/\sigma_b$  for spheres shown in Table 1 were computed for the geometric mean radius of 1.2  $\mu$ m and over the radius range 0.75 to 10  $\mu$ m.

The experimental extinction and backscatter coefficient results obtained for obscuring carbon graphite flakes aerosol (Asbury M260 # 4676) are shown in Figures 10, 11, 12 and 13 for 1064, 532, 355 and 266 nm respectively. The experimental values for  $\sigma_e/\sigma_b$  (and standard error) shown in Table 1 are also shown on these figures as are the Mie theoretical values for  $\sigma_e/\sigma_b$  (at radius 1.2  $\mu$ m).

The values for  $\sigma_e < 0.1 \text{ m}^{-1}$  (corresponding to an aerosol mass density of  $< 0.056 \text{ g/m}^3$ ) have large errors due to the low levels of backscattered signal observed. The values for  $\sigma_e > 1 \text{ m}^{-1}$  (aerosol mass density  $> 0.56 \text{ g/m}^3$ ) include multiple scattering effects whereas the theoretical values assume single scattering. Reasonably good agreement was obtained between the experimental and theoretical values for  $\sigma_e/\sigma_b$  for obscuring aerosol of carbon graphite flakes (Asbury M260 # 4676) at 1064, 532, 355 and 266 nm.

Some consideration has been given to the effect of the shape of the carbon graphite flakes. The absorption of an infinite but thin slab with parallel sides in the Rayleigh limit (ie. for small particles whose diameter is much less than wavelength  $\lambda$ ) has been determined by Faxvog and Roessler (1981). This slab can also be considered as a large thin disc. The ratio R of the absorption cross section per unit volume for slabs to the absorption cross section per unit volume for spheres (Mie) is given by  $|m^2 + 2|^2/9$  where refractive index m = n - ik. This ratio R is given in Table 1 for carbon graphite flakes (Asbury M260 # 4676) at 1064, 532, 355 and 266 nm wavelengths and is calculated to be 4.3, 2.6, 2.4 and 1.6 respectively. Incident light on a carbon graphite flake is absorbed, transmitted or scattered. An increase in absorption for a disc shaped particle results in a decrease in scattering for that same aerosol particle. This in return will result in a reduced value of backscatter. This effect is expected to be more dominant at larger wavelengths for a given aerosol since proportionally more Rayleigh scatterers will be present. The size distribution for carbon graphite flakes (Asbury M260 # 4676) is shown in Figure 5 and 6. The sizes were only measured down to 0.75  $\mu$ m radius although some smaller Rayleigh particles would be expected. For absorbing aerosols Faxvog and Roessler (1981) have shown that maximum Mie scattering for spheres occurs in the range  $0.15 < 2r < 0.5\lambda$  ie. for particle radius ranging from 0.16 to 0.53  $\mu$ m at 1064 nm and from 0.040 to 0.13  $\mu$ m at 266 nm wavelength.

The experimental values for  $\sigma_e/\sigma_b$  are greater than the theoretical values for spheres for carbon graphite flakes at the longer wavelengths 1064 and 532 nm as shown in Table 1. At the shorter wavelengths 355 and 266 nm the effect would not be expected as there are fewer Rayleigh scatterers. These results are consistent with the assumption that the carbon graphite flakes have a shape between that of spheres and infinite thin parallel slabs or large thin discs.

#### References

F.R. Faxvog and D.M. Roessler, "Optical absorption in thin slabs and spherical particles", Appl. Opt. 20, 729 - 731 (1981).

F.R. Faxvog and D.M. Roessler, "Carbon aerosol visibility vs particle size distribution, "Appl. Opt. <u>17</u>, 2612 - 2616 (1978).

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M260
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Flakes
Graphite
Carbon (
from
Backscatter
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Extinction
Table 1

Wavelength	Refractive (a) Real n	: Index (b) Iniaginary k	2	σ <sub>e</sub> /σ <sub>b</sub> sr (theoretical (a) radius range (b 0.75 to 10 μm	) ) Geometric mean radi $r_{g} = 1.2 \ \mu m$	σ <sub>6</sub> /σ <sub>6</sub> sr (experimental) ius	Number of measurement
266	1.39	0.99	1.6	1/1 ~ 091	166	126(± 17)	74
355	1.57	0.76	2.4	216 - 235	225	173(± 15)	137
532	1.64	0.84	2.6	185 - 215	204	228(± 24)	81
1064	1.89	1.27	4.3	109 - 141	133	243(± i3)	80







Experimental decay in extinction coefficient with time for aerosols of carbon graphite flakes (Asbury M260 # 4676)





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# Figure 4 Schematic diagram of experimental arrangement for transmission and backscatter measurements.



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## Mass distribution $dm(r)/d \log_{10} r$ as a function of radius r for carbon graphic flakes (Asbury M260 # 4676) Figure 6



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flakes (Asbury M260 # 4676)





Mie backscatter gain G (m,x) as a function of particle size parameter x for carbon graphite flakes (Asbury M260 #4676) at different wavelengths  $\lambda$  and index of refraction m.



Figure 10

Measured values of backscatter and extinction coefficient for carbon graphite flakes (Asbury M260 # 4676) at wavelength  $\lambda$  of 1064 nm. The solid line represents the experimental value of  $\sigma_e/\sigma_b = 243$  sr and the dashed line represents the Mie theoretical value of  $\sigma_e/\sigma_b = 133$  sr.



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Figure 11

Measured values of backscatter and extinction coefficient for carbon graphite flakes (Asbury M260 # 4676) at wavelength  $\lambda$  of 532 nm. The solid line represents the experimental value of  $\sigma_e/\sigma_b = 228$  sr and the dashed line represents the Mie theoretical value of  $\sigma_e/\sigma_b = 204$  sr.



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Figure 12

Measured values of backscatter and extinction coefficient for carbon graphite flakes (Asbury M260 # 4676) at wavelength  $\lambda$  of 355 nm. The solid line represents the experimental value of  $\sigma_{\rm e}/\sigma_{\rm b} = 173$  sr and the dashed line represents the Mie theoretical value of  $\sigma_{\rm e}/\sigma_{\rm b} = 225$  sr.



Figure 13

Measured values of backscatter and extinction coefficient for carbon graphite flakes (Asbury M260 # 4676) at wavelength  $\lambda$  of 266 nm. The solid line represents the experimental value of  $\sigma_e/\sigma_b = 126$  sr and the dashed line represents the Mie theoretical value of  $\sigma_e/\sigma_b = 166$  sr.



### (ii) Measurement of Biological aerosol (pollen and spores) using a continuous Buckard spore sampler at the Mace Head field station, on the west coast of Ireland

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Number concentration of pollen and spore species are obtained using the continuous Burkard volumetric spore sampler in place at the Mace Head Atmospheric Research station. The number concentration is given per unit volume  $(m^3)$ . A sample of the Burkard pollen and spore count for an eight day period in July 1993 is shown in Table 2. The total pollen concentration,  $m^{-3}$  at Mace Head for the months of June, July and August 1993 are shown in Figures 14, 15 and 16. The concentration of individual species for June 1993 is shown in Figure 17 (a) through (f). Further details of species concentration and analysis thereof will be presented in subsequent interim reports.

julian date	gramineae	uimus	plantago	salix	pleridium
195.33	3.8	0.0	0.0	0.0	0.0
195.67	0.0	0.0	0.0	0.0	3.8
195.83	3.8	0.0	0.0	3.8	3.8
195.92	22.7	0.0	0.0	0.0	0.0
196.08	83.3	0.0	75.8	0.0	0.0
196.17	45.5	0.0	0.0	0.0	0.0
196.25	87.1	0.0	0.0	0.0	0.0
196.33	11.4	0.0	0.0	0.0	0.0
198.50	3.8	0.0	3.8	0.0	0.0
196.75	7.8	0.0	3.8	0.0	0.0
197.00	7.6	0.0	3.8	0.0	0.0
199.92	3.8	0.0	0.0	0.0	0.0
200.08	3.8	0.0	0.0	0.0	0.0
201.35	3.8	0.0	0.0	0.0	0.0
201.85	11.4	0.0	0.0	0.0	0.0
201.94	7.6	0.0	0.0	0.0	0.0
202.02	22.7	0.0	0.0	0.0	0.0
202.10	3.8	0.0	0.0	0.0	0.0
202.27	7.8	0.0	0.0	0.0	0.0
202.44	3.8	0.0	0.0	0.0	0.0
202.52	3.8	0.0	0.0	3.8	0.0
202.94	3.8	0.0	0.0	0.0	0.0
203.02	3.8	7.6	0.0	0.0	0.0

A sample of the Burkard pollen and spore count per m<sup>3</sup> at Mace Head from the month of July 1993 Table 2





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Pollen count for July 1993



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### Figure 17(a) Pollen and spore count (m<sup>-3</sup>) from June 1993 at Mace Head



Figure 17(b) Pollen and spore count (m<sup>-3</sup>) from June 1993 at Mace Head





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# Figure 17(e) Pollen and spore count (m<sup>-3</sup>) from June 1993 at Mace Head





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# (iii) Measurement of Biological aerosol (pollen and spores) using an array of passive samplers, at seven sites in Western Ireland

Data for spore and pollen species using an array of passive samplers or so called Tauber traps is presented in this section. Seven such samplers were positioned in the field in the west of Ireland and are changed regularly once a month within a day or so of each other so as to permit intercomparison of the biological aerosol for the different sites. The Tauber trap changing log to date is shown in Table 3. A short description of the sample analysis and counting procedure is already given in Interim Report 4.

The percentage count for the range of pollen and spore species encountered at the seven sites is shown in an extended Table 4. This covers the period from 18 November 1992 through 14 September 1993. An extended data base for the same range of pollen and spore species is given in Table 5 for the same period. The concentration is given in units of 10 grains  $cm^{-2}$  (equivalent to a scale unit of 10 shown in the horizontal axis) over each month's sampling period. A scale unit labelled 10 on this set of plots represents 10 grains. Concentrations of less than 1 grain per day per cm<sup>2</sup> of trap orifice are represented by a black dot. This dot does not indicate the concentration of grains of that species for that period of time. It merely expresses that some grains were found and counted of that species for that period of time but that their number when calculated for concentration per day per cm<sup>2</sup> of trap orifice was reduced to less than one, which is a division too minute to be shown on this particular plot. On-going sampling counting and analysis of the biological aerosol is taking place and analysis of the main findings from the measurements will be presented in subsequent interim reports.

TABLE 3	TAUBER TRAP CHANGING RECORD AT THE SEVEN
	STTES IN THE WEST OF IRELAND

Location	set	1st change	synchronisation	second change	third change	fourth change	fifth change
Mace Head unroofed	18/11/92	25/1/93	18/2/93	15/3/93	14/4/93	19/5/93	16/6/93
Mace Head roofed	18/11/92	25/1/93	18/2/93	15/3/93	14/4/93	19/5/93	16/6/93
Letterfrack	4/12/92	25/1/93	19/2/93	19/3/93	4/93	19/5/93	16/6/93
Burren platform	17/12/93		17/2/93	16/3/93	19/4/93	18/5/93	17/6/93
Kylemore	9/2/93 .		9/2/93	15/3/93	14/4/93	19/5/93	16/6/93
Ballyconneely	9/2/93		9/2/93	15/3/93	14/4/93	19/5/93	16/6/93
Burren lake	17/2/93		17/2/93	16/3/93	19/4/93	18/5/93	17/6/93

Location	6th change	7th change	Sth change	9th change	10th change	11th change	12th change
Mace Head unroofed	12/7/93	18/8/93	16/9/93	18/10/93	13/12/93	16/2/94	15/3/94
Mace Head roofed	12/7/93	18/8/93	16/9/93	18/10/93	13/12/93	16/2/94	15/3/94
Letterfrack	19/7/93	16/8/93	13/9/93	18/10/93	13/12/93	17/2/94	15/3/94
Burren platform	14/7/93	17/8/93	14/9/93	19/10/93	15/12/93	15/2/94	16/3/94
Kylemore	19/7/93	16/8/93	13/9/93	18/10/93	13/12/93	16/2/94	15/3/94
Bailyconneely	19/7/93	16/8/93	13/9/93	18/10/93	13/12/93	raft stoien	raft replaced 15/3/94
Burren lake	14/7/93	17.'8/93	14/0/93	19/10/93	11/1/94	18/2/94	16/3/94

Location	13th change	14th change	15th change	16th change	17th change	18th change	19th change
Mace Head unroofed	18/4/94	16/5/94					
Mace Head roofed	18/4/94	16/5/94					
Letterfrack	19/4/94	16/5/94	-				
Burren platform	21/4/94	19/5/94					
Kylemore	19/4/94	16/5/94					
Bailyconneely	19/4/94	16/5/94					
Burren lake	21/4/94	19/5/94					

Contract funds to the amount of US\$106,924 have been used to date

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# TABLE 4(A)PERCENTAGE OCCURRENCE FOR THE INDICATED SPECIES ATTHE SEVEN SITES IN THE WEST OF IRELAND FROM18 NOVEMBER 1992 -14 SEPTEMBER 1993



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# Tauber traps: Percentages



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### **TABLE 5 CONCENTRATION OF POLLEN AND SPORE SPECIES IN UNITS OF 10** GRAINS PER cm<sup>3</sup> (CORRESPONDING TO A SCALE LENGTH OF 10 ON THE HORIZONTAL AXIS) AT THE SEVEN SITES.



1 horizontal scale unit = 10 grains  $cm^{-2}$ 



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# TABLE 5 CONCENTRATION OF POLLEN AND SPORE SPECIES IN UNITS OF 10 GRAINS PER cm<sup>3</sup> (CORRESPONDING TO A SCALE LENGTH OF 10 ON THE HORIZONTAL AXIS) AT THE SEVEN SITES.

Sphagnum Mace Head unroofed 18 nov - 25 jan 25 jan - 18 feb 18 feb - 15 march 15 march - 14 april 14 april - 19 may 19 may - 16 june 16 june - 12 july 12 july - 18 august 18 august - 16 september Mace Head roofed 18 nov - 25 jan 25 jan - 18 feb 18 feb - 15 march 15 march - 14 april 14 april - 19 may 19 may - 16 june 16 june - 12 july 12 july - 18 august 18 august - 16 september Lenerfrack 4 dec - 25 jan 25 jan - 19 feb 19 feb - 19 march 19 march - i4 april 14 april - 19 may 19 may - 16 june 16 june - 19 july 19 july - 16 august 16 august - 13 september Burren platform 17 dec - 17 feb 17 feb - 16 march 16 march - 19 april 19 april - 18 may 18 may - 17 june 17 june - 14 july 14 july - 17 august 17 august - 14 september Kylemore 9 feb - 15 march 15 march - 14 april 14 april - 19 may 19 may - 16 june 16 june - 19 july 19 july - 16 august 16 august - 13 september Ballyconneely 9 feb - 15 march 15 march - 14 april 14 april - 19 may 19 may - 16 june 16 june - 19 july 19 july - 16 august 16 august - 13 september Burren lake 17 feb - 16 march 16 march - 19 april 19 april - 18 may 18 may - 17 june 17 june - 14 july 14 july - 17 august 17 august - 14 september 1x1