

LETTER TO THE EDITOR

Reply

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We cannot accept the proposition that, just because measurements may be difficult and their interpretation complicated by problems such as (in the present instance) imperfect collection of some sizes, we should abandon measurements and trust to “extensive model calculations” to draw conclusions about the real atmosphere.

It is not entirely clear why our measurements have been categorized as “poor”. The method which we used for measuring absorption (once the samples have been collected) has been compared with two other methods found in the literature, with surprisingly good agreement (Ramsey-Bell and Couture, 1985). Collection extended over several years (late fall, winter and early spring); some 250 samples were obtained and over 100 processed, so the sample overall could not be called small. We must presume that the acknowledged fact that some sizes of particle escaped collection, or were collected at very low efficiency, is the basis for slighting our results. In our original paper, we attempted to adjust our data for imperfect collection, used an upper limit rather than a trend line through the data, and finally demonstrated that increasing that absorption tenfold produced little change in our conclusions, as Fig. 6a of the original paper showed. Even if our underestimation was as high at 90% (rather than 30%), the necessary eightfold increase from our median (or mean) was more than accounted for in the steps leading to the dashed line in Fig. 6a.

Rather than indulge in endless modelling, it is more instructive to approach the problem from another perspective. The “optical thickness effect,”

as we computed it, gave a roughly 50% increase in cloud albedo (averaged over a distribution of cloud thickness) for a tenfold increase in nucleus concentration. Now we ask: how much absorption would give a compensating 30% decrease in cloud albedo *under the most favorable circumstances*? Infinitely deep, initially conservative clouds will be the most sensitive to absorption darkening, hence the question:

For  $\tau = \infty$ ,  $\bar{\omega} = 0$ , initially, what value for  $\bar{\omega}_0$  will reduce spherical albedo from 1 to 0.7?

For isotropic scattering, the question can immediately be answered. Table XI of Chandrasekhar (1950) provides the data needed to obtain plane albedo at solar zenith angle  $\cos^{-1} \mu_0$  using the well-known formula

$$(\text{plane albedo}) = 1 - \sqrt{1 - \bar{\omega}_0} H(\mu_0),$$

and, integrating over an illuminated sphere to obtain spherical albedo,

$$\text{spherical albedo} = 1 - 2\sqrt{1 - \bar{\omega}_0} \int_0^1 \mu_0 H(\mu_0) d\mu_0.$$

The results are tabulated below in Table 1 (and are easily verified).

Table 1. *Dependence of albedo (spherical) on  $\bar{\omega}_0$  in isotropic scattering*

$\bar{\omega}_0$	Albedo
1.0	1.00
0.975	0.70
0.95	0.60
0.925	0.53
0.9	0.48

\*Now with SM Systems & Research Corp., Landover, MD, USA.

A 30% decrease in spherical albedo is seen to require a single-scattering albedo of about 0.975. Cloud drops, of course, are not isotropic scatterers, but similarity relationships (Van de Hulst, 1980, pp. 477–479) show that an infinite isotropic layer with single-scattering albedo,  $\omega_{\text{iso}}$ , is similar to a non-isotropically scattering layer with single-scattering albedo,  $\bar{\omega}_c$ , provided  $\bar{\omega}_{\text{iso}} = (1 - g)\bar{\omega}_c / (1 - g\bar{\omega}_c)$ , which for typical cloud droplet asymmetry parameters gives  $\bar{\omega}_c \approx 0.995$  for  $\bar{\omega}_{\text{iso}} \approx 0.975$ . This means that *in the cloud* absorption should amount to 0.005 times extinction.  $0.05 \text{ m}^{-1}$  may be taken as a typical value for the latter in, say, marine stratus, hence the required absorption is about  $2.5 \times 10^{-4} \text{ m}^{-1}$ .

An absorption of  $2.5 \times 10^{-4} \text{ m}^{-1}$  is roughly 20 times larger than *scattering* coefficients measured by us at Mt. Lemmon, by Waggoner et al. (1981) in Arizona, or at GMCC monitoring stations (Bodhaine, 1983). If collected by aspiration through a  $\frac{1}{4}$ " diameter area on a  $1 \mu\text{m}$  Nuclepore disk, unit absorption optical thickness (absorption  $= 1 - e^{-1}$ , very easily observed) would accrue in 7 min at 100% collection efficiency, in 70 min at 10% collection efficiency, and in about half a day even at 1% collection efficiency. (We typically obtained unmeasurable absorption in such aspirated samples after several hours sampling; samples extending for 2–3 days consistently gave absorption of only a few %).

We can also examine the implications of an absorption of  $2.5 \times 10^{-4} \text{ m}^{-1}$  from the viewpoint of the required carbon mass and/or particle concentration. The refractive index of carbon approximates  $2 - i$  across the visible and near infrared (Janzen, 1979). Mie computations for that refractive index show a volume-weighted absorption which is relatively constant at  $7.5 \times 10^4 \text{ cm}^{-1}$  across a wide range of sizes, with a maximum of  $1.2 \times 10^5 \text{ cm}^{-1}$ ; hence  $2.5 \times 10^{-4} \text{ m}^{-1}$  atmospheric absorption would mean at least  $2 \times 10^{-11}$  (volume mixing ratio) of carbon aerosol—which

Table 2

Data source	Absorption ( $10^{-7} \text{ m}^{-1}$ )
Measured, Mauna Loa*	0.45
Measured, arctic summer**	0.4
Measured, arctic winter**	8
Measured, northern Arizona*	7–17
Formula, $N = 10 \text{ cm}^{-3}$	0.5
Formula, $N = 100 \text{ cm}^{-3}$	2
Formula, $N = 1000 \text{ cm}^{-3}$	10

\* Waggoner et al. (1981); \*\* Heintzenberg (1982).

exceeds most estimates of *total* aerosol mixing ratio outside of heavily polluted urban-industrial surface locations.

Perusal of the several references offered in rebuttal of our conclusions has not mitigated our disagreement with the "dirty cloud" proposition. Firstly, none of these references cited, even in urban-industrial samples, atmospheric absorptions as high as  $2.5 \times 10^{-4} \text{ m}^{-1}$ ; secondly, Heintzenberg (1982) and Heintzenberg and Covert (1984) consistently showed distributions of carbon with size in which at least half of the carbon lay in the particle radius range  $>0.1 \mu\text{m}$ ; furthermore carbon was spread widely across about two decades in radius, not concentrated in a narrow interval which might have escaped collection; thirdly, as Table 2 summarizes, there is no serious discrepancy in magnitude between the absorption values given by our formula  $A = 10^{-8} N^{2/3}$  and values reported in the papers of Waggoner et al. (1981) and Heintzenberg (1982).

We see no contradiction in our use of a formula which gave a range of  $0.5$  to  $10 \times 10^{-7} \text{ m}^{-1}$  over the present-day range of probable value for  $N$  in the light of measurements by others in other places which ranged from  $0.4$  to  $17 \times 10^{-7}$ . If anything, this bolsters our confidence in the use of data from a single site in the computations which we made.

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