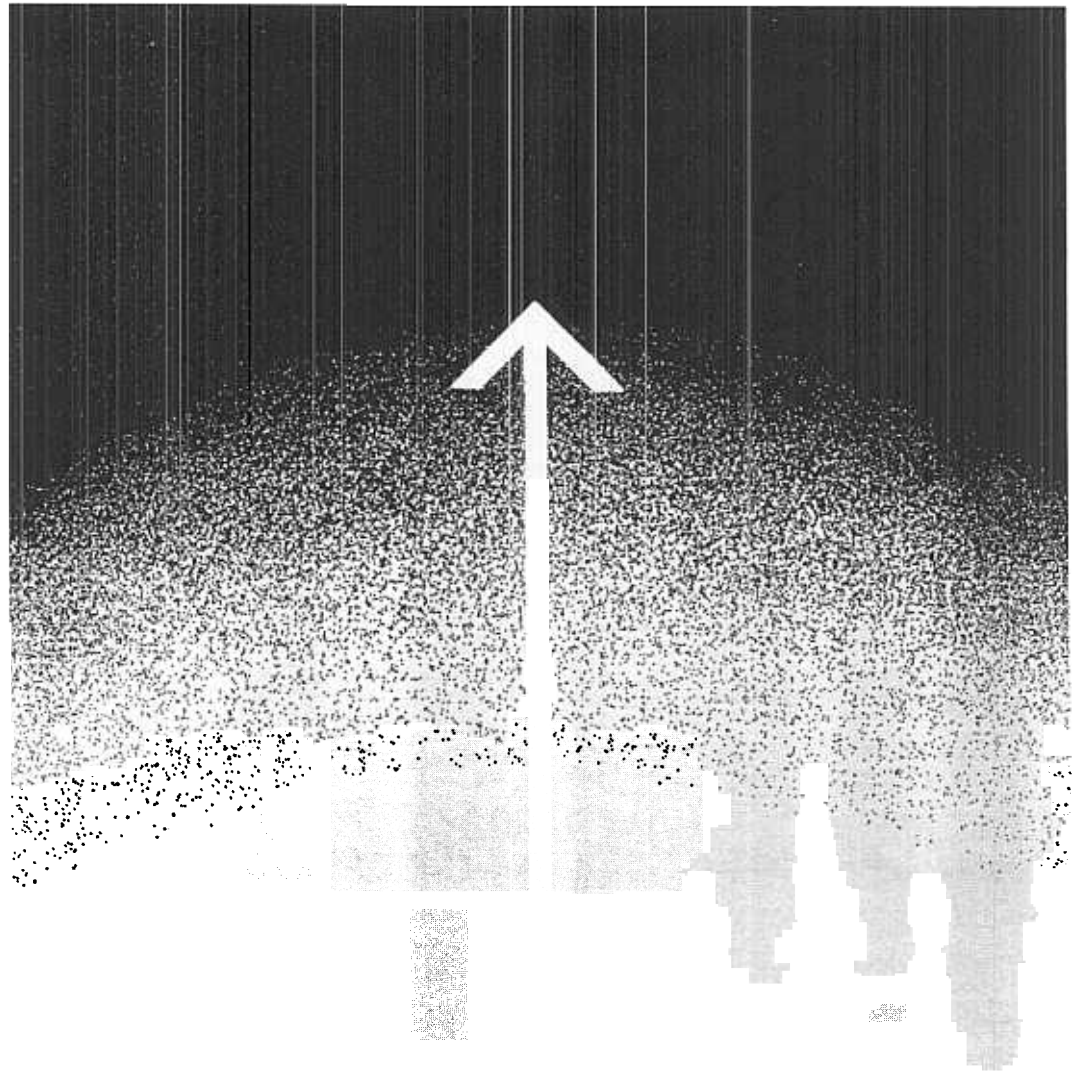


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Measurements and Modelling of Aerosol Single-Scattering Albedo: Progress, Problems and Prospects¹

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Abstract

The net effect of atmospheric aerosols in the radiation balance is determined by both their scattering and absorption of solar radiation. The combined optical effect is expressed in the single scatter albedo, ω , of the particles. Currently available data on ω are insufficient for definitive use in climate models because most of them are not corrected for the method-dependent effect of the scattering portion of the aerosol on the measured absorption, most refer to the dry state of the aerosol, and the coverage of the globe is far from being complete. Standardisation and calibration of the measurements is needed. Modelling exercises using currently available data on ω should clearly state that corrections are required. The purpose of this review is not to suggest a particular range of values for single scatter albedo. Rather, it is to illustrate that the uncertainties are currently imbedded in various data sets because of the lack of calibration, the possibility that many of the extant methods systematically overestimate light absorption coefficients, and the necessity of including the influence of humidity in models.

1 Rationale: The Importance of Single Scattering Albedo of Aerosols to the Problem of Radiative Forcing

Aerosol particles in the atmosphere both scatter and absorb sunlight, and may absorb infrared radiation as well. Aerosols deriving from human activities cause radiative *forcing*, that is they impose a change in heat balance (Wm^{-2}) above and beyond effects of natural aerosols by directly reflecting sunlight backward into space, or by absorbing sunlight or infrared radiation and heating the atmosphere. The relative magnitudes of these two processes – scattering and absorption – are embodied in the concept of the single scattering albedo, ω , which is defined as the dimensionless ratio of the scattering coefficient of the aerosol particles,

σ_{sp} (m^{-1}), to the extinction coefficient, σ_{ep} (m^{-1}). In turn, $\sigma_{ep} = \sigma_{sp} + \sigma_{ap}$ where σ_{ap} is the absorption coefficient². The *co-albedo of single scatter* ($1 - \omega$) is often used in radiative transfer calculations and, at typical values of ω close to unity, it is a sensitive indicator of the importance of light absorption. All of the above optical aerosol parameters are functions of wavelength, λ . Indirect radiative forcing by the same anthropogenic aerosols may be caused by their influence on cloud albedo, cloud area or cloud lifetime; however these will not be considered further here.

¹ This reviewed editorial article summarizes the outcome of an international workshop held in March 1997 at the Institute for Tropospheric Research (IfT) Leipzig

² As we only deal with the optical effects of aerosol particles index p is left out in the rest of this text.

Prior to ca. 1990, most estimates of the effects of atmospheric aerosols on heat balance considered the aerosol as a whole (natural plus anthropogenic) (Coakley Jr. et al., 1983; Pollack et al., 1976). The single scattering albedo of the aerosols figured prominently in these papers, and was usually estimated by assuming an imaginary part of the refractive index, n_{im} , of the aerosol material. Typical values of n_{im} of 0.004 to 0.2 yielded single scattering albedos in the range 0.97 - 0.5. The issue of heating versus cooling by aerosols formed a substantial part of the efforts in the 1970's and 1980's, with a consensus existing that cooling dominated, except perhaps close to sources of strong light absorbers like black carbon (BC). The critical influence of the ratio of aerosol scattering to absorption was already pointed out by Yamamoto and Tanaka (1972). Non-light-absorbing aerosols (primarily sulfates and organic matter) reflect sunlight and have a cooling effect that is greatest over low albedo surfaces (e.g. oceans) and smallest over high albedos (e.g. clouds or snow). Light absorbing aerosols (probably mainly BC but possibly also soil dust) also scatter light and their net effect depends on the relative magnitudes of scattering and absorption (e.g. single scattering albedo) as well as the albedo of the underlying surface. Because emissions of these aerosols depend on latitude and season the optical effects also have this dependence.

The advent of three dimensional models of the atmospheric cycle of sulphur (e.g., Langner and Rodhe, 1991; Erickson et al., 1991) made it possible to quantify separately the forcing by sulphate compounds as one key type of aerosol. Simultaneously, the consideration of sulphates alone momentarily obviated the problem of quantifying the single scattering albedo because these compounds do not absorb visible light.

However, the numerous papers suggesting the existence of a significant negative radiative forcing by anthropogenic aerosols also resulted in a variety of attempts to use the cooling effect of sulphates to reconcile the difference between climate model simulations of temperature and observations. They assumed that the totality of aerosol effects could be represented by the direct effect of sulphates alone. While it is perfectly reasonable to conduct calculations of the sensitivity of forcing to individual constituents like sulphates, failure to include other aerosol components that might absorb light prior to conducting response studies amounts to an unstated assumption that $\omega = 1$, which is obviously unrealistic because of the known existence of light absorbing materials in aerosols. The values of ω that should be used, where

and when significant departures of ω from unity occur, what the variability of ω is and what aerosols cause this are the questions to be discussed below.

Meanwhile, over the past 20 to 30 years, a variety of estimates of the magnitude of ω have been published based on different measurement or modelling approaches. Rekindled interest in radiative forcing in general and aerosol forcing in particular have resulted in a number of recent efforts to evaluate the role of light absorption by anthropogenic aerosols (e.g., Haywood and Shine, 1995; Liousse et al., 1996; Hansen et al., 1997).

The range of measured or modelled regional-mean values of ω at low *RH* range from ca. 0.7 (Malm et al., 1996) to 0.95 and higher (Liousse et al., 1996) for non-urban areas over the populated continents to 0.98 or higher for remote locations (Clarke and Charlson, 1985). Figures 1 and 2 compare measured versus modelled values of absorption in the form of co-albedo ($1 - \omega$) illustrating the existence of a wide range of values of ω around 0.9 and higher, the existence of one unrealistic outlier and a possibly slight overestimate of modelled values of co-albedo in remote regions. The underlying data are listed in Table 1 which is a modified version of Table 8 in Liousse et al. (1996) plus the data of Malm et al. (1996). There are several other data sets on ω in the literature, and still others have not yet been published. For the reasons given in Sections 4 and 5 below, we do not think it appropriate to include more data before the calibration problems have been rectified. In addition, the uncertainties in the data in Figures 1 and 2 require that these be viewed as examples for discussion rather than data to be used in climate models except in sensitivity studies.

These graphs exemplify that *both* modelling and measurement approaches are necessary and serve different and complementary purposes. It is impossible to acquire enough measurements to provide a meaningful three-dimensional map of the needed aerosol properties (e.g. comparable to weather maps) such that models are needed to provide the spatial extrapolation between and among the measurements and to calibrate the models. In addition, models are helpful in separating anthropogenic from natural aerosol effects. Clearly, neither approach alone is sufficient and comparing the two may reveal the degree to which the modeling needs refinement.

In climate response studies, significant errors result either from assuming unrealistically high or low values of ω . However, errors caused by using low values of ω , resulting from overestimates of absorption,

Table 1: Comparison of observed (ω_{obs}) and modelled (Liousse et al., 1996) (ω_{mod}) single scattering albedo values at different locations. # = reference number in Figures 1 and 2.

Locations (Lat.; Long.)	#	Time Period	ω_{obs}	ω_{mod}	References
Barrow (71.2; -156.3)	1	Annual	0.96	0.92	Bodhaine, 1995
Amundsen Scott (-90)	2	Annual	0.965	0.965	Bodhaine, 1995
Mauna Loa (19.3; -155.4)		June	0.95	0.94	Bodhaine, 1995
Mauna Loa (19.3; -155.4)	3,3'	July	0.96	0.955	Bodhaine, 1995
Ny Ålesund (79; 12)	4	Annual	0.948	0.94	Heintzenberg, 1982b
Kamchatka (56; 160)	5	May	0.88	0.92	Clarke, 1989
Equator (0; 100)	6	May	0.998	0.95	Clarke, 1989
Wellington (-41.17; 174.47)	7	June	0.88	0.90	Clarke, 1989
West Australia (-34.19; 115.09)	8	June	0.998	0.95	Clarke, 1989
Tropical Atlantic Ocean	9	August	0.8	0.89	Clarke, 1989
Arctic	10	Annual	0.96	0.91	Heintzenberg, 1982b
Allegheny Mts. (38.3; -80)	11	Summer	0.935	0.97	Japar et al., 1986
Shenandoah (38; -78)	12	Summer	0.949	0.97	Ferman et al., 1981
Mesa Verde (37.1; -108.3)	13	Annual	0.91	0.93	Waggoner et al., 1981
Abastumani (41.4; 42.5)	14	Annual	0.89	0.87	Waggoner et al., 1981
Anderson Mesa (35.12; -111.38)	15	Annual	0.94	0.935	Waggoner et al., 1981

could easily be as large as those caused by assuming $\omega = 1$.

There are demanding requirements on the accuracy and precision of single scattering albedo due to the high sensitivity of radiative forcings and responses to small changes in ω . The crossover between positive and negative forcing in various models of the direct aerosol effect occurs at values of ω of ca. 0.85, higher values yielding negative forcing. General circulation model calculations of global mean temperature response with fixed clouds yields a transition from warming to cooling at about the same value. However, if a feedback of the cloud cover on boundary layer heating by aerosols is included, the transition from warming to cooling occurs at a higher value of ω (Hansen et al., 1997). So, it appears to be necessary to resolve values of ω to ca. two significant figures in the observed range of values of ca. $0.8 < \omega < 1.0$. To complicate matters still further, these values of model inputs are supposed to represent the single scattering albedo at ambient relative humidity (RH), while virtually all of the determinations of ω are made at low RH . Thus, a means to determine the magnitude of the expected increase in ω due to the hygroscopic growth of the particles and the subsequent increase in σ_s is required.

The key objective of this paper is to show pathways towards obtaining realistic bounds on ω ; and a second objective is to indicate what needs to be done to further constrain and reduce the uncertainties and systematic errors that are inherent in the existing measurement and modelling approaches. The purpose is not to focus on errors; rather, it is to carefully define what is known and, most especially,

not known regarding the light absorption by anthropogenic aerosol particles. Because of the essential role of observational data, particular attention will be paid to measurement methods and their intercalibration.

2 Factors Controlling the Single Scattering Albedo of Atmospheric Aerosols

All substances contained in atmospheric aerosols scatter visible light, but only a small fraction of them have the property of light absorption. The amount of light absorption, that is the absorption coefficient, σ_a , is controlled by the amount of absorbing material and the elemental and molecular forms that are present, the particle sizes in which the absorbers occur, the state of mixing and morphology of the absorbers within individual particles. As mentioned above, relative humidity and the molecular forms of the aerosol substances control the amount of water in or on the particles, thereby further influencing the relative amounts of scattering and absorption.

Far from being a constant or conserved property, single scattering albedo is highly variable and depends on the relative source strengths of the various aerosol substances, aerosol transformations (e.g., coagulation) and in situ chemical reactions that produce aerosol mass (e.g. the oxidation of SO_2) and removal processes (e.g., in rain). No single value of ω can represent the entire globe, nor can a single value be assumed for a given location as a function of time. Seasonality is known to occur due to changes in source strength (e.g. of black carbon from biomass burning).

On the decade to century time scale, changes in ω must occur because of changes in industrial practices, agronomy, volcanism, forest fire etc.

Widely different values of ω are possible because the chemical composition and microphysics of the atmospheric aerosol are highly variable. Both the real and imaginary parts of the refractive index are controlled by the molecular form and morphology of the aerosol substances as discussed above. Therefore, connecting the physical effects of light absorption to the sources of the light absorbing material requires a means to quantitatively describe the sources, transformations and removal of the material that realistically portrays the role of the molecular form(s) and morphologies that are involved. Recognising and including all of the important controlling variables itself becomes a difficult task. This has not yet been achieved but the process has been started (e.g., Cooke and Wilson, 1996; Liou et al., 1996; Penner et al., 1992, 1993). *In fact, most of the data on or calculation of ω that are available today were acquired for purposes other than application to climate models and certainly were not optimised for that purpose. Similarly, almost none of the data acquired for the source strength of light absorbing materials were intended for such purposes and as a result, at least some of them are clearly inappropriate for that task* (Bond et al., 1997).

3 The Role of Single Scattering Albedo in Climate Models

An assessment of the climatic impact of aerosols requires knowledge of the geographical and vertical distribution of the aerosols, their composition, their seasonal variation and longer-term trends. From this distribution it is necessary to calculate the optical depth, single scattering albedo and some measure of the angular distribution of the scatter, which requires knowledge of, or assumptions about, the size distribution and even the shape of the particles. To determine anthropogenic forcing, it is necessary to know the fraction of each component of the aerosol which is due to human activity.

Because existing aerosol-climate studies are limited by current observations and uncertainties in chemical transport models (to be discussed below), these studies to be regarded only as "sensitivity studies" which should motivate improved measurements and modelling of the distribution and characteristics of aerosols. This situation can be contrasted with attempts to clarify the climatic forcing due to changes

in carbon dioxide (and other well-mixed greenhouse gas) concentration which are, relatively, much more straightforward.

The dependence of the sign of the aerosol climatic effect on single scattering albedo has long been recognised (e.g., Ackerman and Toon, 1981; Coakley Jr. et al., 1983; Hansen and Lacis, 1990; Grassl, 1988). During the early 1990's the rekindled interest in the climatic effects of aerosols focused first on the effect of anthropogenic sulphate aerosols which, because of $\omega = 1$ at visible wavelengths, would, on their own, undoubtedly act to cool climate (Charlson et al., 1992).

Even though BC constitutes only a few percent of the aerosol mass, it can have a significant positive forcing effect because it decreases the planetary albedo (Chylek and Wong, 1995; Chylek et al., 1995; Schult et al., 1996; Haywood and Shine, 1995; Haywood and Shine, 1997; Haywood et al., 1997b; Haywood and Ramaswamy, 1997; Ramaswamy and Chen, 1997). BC forcing is enhanced several-fold if the soot is above or within the cloud, and it may be enhanced if it is internally-mixed with sulphates or other components. The role of BC in clouds could be enhanced considerably if is located within cloud droplets rather than remaining interstitial (Chylek et al., 1984; Chylek et al., 1996; Heintzenberg and Wendisch, 1996). The global mean forcing due to anthropogenic BC could be up to the order of several tenths of a $W m^{-2}$ (Haywood and Shine, 1995). Thus, it might be of comparable size, and opposite in sign, to the effect of anthropogenic sulphate aerosols but probably occurs at different altitudes, at different locations and has a different trend over time.

Whilst the role of absorbing aerosol is enhanced at higher altitudes, particularly if above clouds, the role of sulphate decreases because at lower relative humidity the particles are smaller and scatter less. This makes the determination of the vertical profile and hygroscopicity of aerosol of particular significance. Horizontal variation in relative humidity, on scales smaller than those typically used in General Circulation Models, can impact the overall forcing (Haywood et al., 1997a). The size of this dependence will be determined by the degree to which the aerosol is influenced by humidity changes.

BC is not the only aerosol component that absorbs at visible wavelengths. Organic and mineral aerosols, though much less absorbing than BC, absorb radiation, especially in the near-UV. (Tegen et al., 1996; Sokolik and Toon, 1996). Partly because of lack of

data, and partly because the large mineral dust particles may have a significant "greenhouse effect", even the sign of the mineral dust forcing is uncertain.

The climate response from a given radiative forcing due to an absorbing aerosol, especially when present in large amounts, may differ from the climate response due to an identical value of radiative forcing due to some other mechanism, such as changes in greenhouse gas concentration (Cess et al., 1985); which complicates direct comparison of radiative forcing values. Recently Hansen et al. (1997) used a simplified GCM to explore its sensitivity to single scattering albedo. The critical single scattering albedo ω_c , for which the global mean (but not necessarily the local) temperature change was zero was found, if cloudiness was kept fixed, to be 0.86; when certain cloud feedbacks were allowed in the model, ω_c increased to 0.91, significantly closer to current observational estimates of ω . Hansen et al. attributed the increase in critical ω to a "semi-direct" aerosol feedback effect whereby the heating due to the absorbing aerosol caused a decrease in low cloud amount.

Hansen et al.'s result was for a homogeneously-distributed aerosol restricted to the lowest 3 kilometres; the value of critical ω was shown to depend on the geographical and vertical distribution of the aerosol and would alter if the aerosol within clouds was incorporated in the droplets. Coupled with the fact that cloud feedbacks are notoriously different from model to model, this means that Hansen et al.'s value of the critical ω should not be taken too literally. The importance of the study is that it draws attention to a previously neglected feedback mechanism which appears to make absorbing aerosols of greater climatic significance than is indicated by their contribution to radiative forcing.

Changes brought about in the radiative fluxes at the surface, due to introduction of an absorbing aerosol versus a purely scattering aerosol, could perturb the surface energy and water balance over land surfaces (Chen and Ramaswamy, 1995; Coakley Jr. and Cess, 1985) and possibly alter the manner of changes in the soil moisture (Chen and Ramaswamy, 1996).

Another effect of the introduction of absorbing aerosols in the atmosphere is the increased heating (Coakley Jr. and Cess, 1985). This effect is enhanced if the absorbing aerosol is above a high albedo surface such as snow or low clouds. If the surface and the troposphere are well-coupled (e.g., through convection/turbulent mixing) then this heat would tend

to be communicated throughout the tropospheric column, effecting a radiative warming of the surface-troposphere system. If the surface and troposphere are not coupled in an efficient manner as in the case of large aerosol amounts (Cess et al., 1985; Ramaswamy and Kiehl, 1985), there would be a preferential heating of the atmosphere and cooling of the surface, with accompanying impacts on the static stability of the atmosphere, and possible inhibition of convection.

Absorbing aerosols reduce reflection of sunlight at the top of the atmosphere, thus affecting the direct aerosol radiative forcing. In particular, model-based calculations of NH aerosols forcing by anthropogenic sulphate versus a mixture of sulphate and BC reveal areas of negative and positive aerosol forcing, displaying a fine spatial structure (Haywood et al., 1997b; Schult et al., 1996). At the same time, the equator-to-pole gradient and hemispheric difference of aerosol radiative forcing is altered from the sulphate-only case. This presumably would impact the NH meridional gradient of the surface temperature response. A major inference is that recent findings (Haywood et al., 1997c; Santer et al., 1996; Tett et al., 1996) of good agreement between temperature changes simulated by transient climate models and those measured from pre-industrial to present must be tempered with the realisation that they did not include the significant positive forcing associated with light absorbing aerosols.

4 Progress: The Capability for Quantifying ω at Low RH

Interpretation of any potential radiative forcing associated with light absorbing aerosol in the atmosphere must ultimately be consistent with measurements and their uncertainties. Uncertainties in measurements of albedo reflect uncertainties in both the measured absorption coefficient (σ_a) and scattering coefficient (σ_s) of the aerosol. These uncertainties have diverse origins that are associated both with peculiarities of the techniques employed and the environment or conditions under which the measurements were made. Both parameters underlying ω are strong functions of

- the size of the scattering and absorbing particles,
- the state of mixture of absorbing and scattering particulate matter
- the wavelength of light, and

- the relative humidity.

These facts make the generalisation of optical effects from aerosol mass burdens (or vice versa) subject to large and as yet undefined uncertainties. Reducing this uncertainty requires ancillary information that is often unavailable. Moreover, although most instruments make an optical measurement of absorbing species, some of the instruments are calibrated in terms of σ_a while some are calibrated in terms of BC (e.g., Aethalometer, Magee Scientific). As a result the former are most directly applicable for measurement of aerosol optical properties while the latter are intended to provide data on BC mass estimated from *assumed constant optical properties* (Gundel et al., 1984).

4.1 Methodological Issues

Many methods exist for measuring the components of ω , i.e. σ_a , σ_s , or σ_e each of which has both advantages and disadvantages. Because of the small magnitudes of σ_a (typically 10^{-8} to 10^{-5} m^{-1} at $\lambda = 550$ nm) most of the experimental techniques quantify aerosol absorption after preconcentration on a substrate. A description of and intercompared performance of many of these techniques can be found in Gerber and Hindman (1982a). The more common instrumental and modelling approaches are summarised here.

A. Light Scattering

In most atmospheric settings the scattering process dominates the aerosol extinction of light. Direct measurements of σ_s are often used as a measure of total aerosol extinction. They are also used in combination with measurements of σ_a to derive ω . The most common instrument for this purpose has been the integrating nephelometer. Its design features have been summarised in a recent review (Heintzenberg and Charlson, 1996) while the system accuracy of this instrument was characterised by Anderson et al. (1996) at ± 7 %.

B. Integrating Plate Method (IPM) and Variations

Probably the most widely used method for measuring σ_a (not mass of BC) is based upon the IPM (Lin et al., 1973). In this approach, a sample is collected on Nuclepore^R polycarbonate membrane filters and

placed on a Lambertian opal glass diffuser and the normal emergent transmission measured before and after sampling. It is assumed that the Lambertian incident light on the sample is scattered equally into and out of the sensor direction so that the detector is only sensitive to the absorption term. A first error analysis is included in the original publication. Subsequent testing and analysis (Clarke, 1982a) found that perturbation of the optical properties of the Nuclepore^R media caused by the deposited particles generally resulted in overestimating absorption by about 20 – 30 %. This result was later confirmed by long path extinction cell measurements (Weiss and Waggoner, 1984) and others (Hitzenberger, 1993). Amelioration of this enhancement in absorption was shown to be possible by the inclusion of a neutral density filter above the opal glass diffuser, at the cost of poorer characterisation of very low σ_a (Clarke, 1982a). Uncertainties due to substrate inhomogeneities in Nuclepore^R filters were reduced in a multi-sample IPM technique described in Heintzenberg (1988).

A technique related to the IPM technique replaces the Nuclepore^R filter (surface collector) with fibrous filters (e.g., quartz, glass, paper) which results in particle penetration into the filter media (Hansen et al., 1982; Rosen and Novakov, 1983). This introduces the particles into a multiple scattering optical environment that amplifies apparent absorption, up to a factor of two or more (Clarke, 1982b; Edwards et al., 1983) and must be empirically calibrated and corrected. A first attempt at this calibration was reported by Ruoss et al. (1992).

Other effects have been observed for the IPM when used with stretched Teflon filters (e.g., IMPROVE program, Malm et al., 1996) and for BC collected in conjunction with other scattering aerosol. In this configuration and with BC mixed with a scattering aerosol of larger particles, the apparent absorption also has been observed to artificially increase by a factor of two or more (Bennett and Patty, 1982). Because the effect increases with increased deposition of scattering aerosol, an empirical correction must be determined that requires concurrent assessment of the scattering aerosol component that is co-deposited on the filter.

C. Integrating Sphere (IS)

The authentic IS technique involves collecting a sample on a substrate and placing it the centre of an integrating sphere (Fischer, 1970; Patterson and Marshall, 1982). In a later version, the sample is dis-

solved in a solvent that does not dissolve the BC before placing it in the sphere (Heintzenberg, 1982b). The change in the detected diffuse light relative to the incident light can be related to the absorption and complex refractive index of the sample and there is no perturbation due to aerosol scattering. Apart from the fact that the dissolution technique destroys the sample, the drawbacks for routine use are the labour intensive sample preparation and minor corrections for putting the absorbing aerosol into a liquid medium for analysis.

An error on the same order of magnitude as for the IPM is reported in (Heintzenberg, 1982b). It is mainly due to optical variabilities in dissolved filter blanks and in the mechanical stability of the optical arrangement.

More recently others have used the IS as an integral detector of diffuse light (Hitzenberger and Puxbaum, 1993; Campbell et al., 1995) for variations of the IPM method. This application does not utilise the properties of a true integrating sphere measurement because it is sensitive to scattering, and spurious absorption has been described using it (Hitzenberger and Puxbaum, 1993).

D. Integrating Sandwich (ISW)

The ISW (Clarke, 1982b) is a cross between the IPM and IS techniques. The samples are collected on Nuclepore^R filters, "sandwiched" between two translucent, diffusely reflecting surfaces, and the change in transmission through the sandwich is measured. It is simpler than the IS procedure, requires less sample and has little sensitivity to scattering i.e. measured ω for non-absorbing salts > 0.999 (Clarke and Waggoner, 1982). It has the advantage that it non-destructive and results in an amplified absorption signal that is sensitive to low absorption values even in the most remote parts of the troposphere but the disadvantage is that response is non-linear and requires carefully calibrated reflective surfaces.

E. Photo Acoustic Detectors (PAD)

The PAD technique places an aerosol-laden filter into a sealed cavity where it is pulsed with light (usually laser) at an acoustic frequency. This results in pulsed heating of the sample that generates small pressure fluctuations detected by a sensitive acoustic microphone (Szkarlát and Japar, 1981). An improved design with sensitivities down to a few micrograms per

cubic meter has been described by Petzold and Niessner (1995). It is non-destructive and has the advantage of avoiding some of the optical uncertainties present in some other methods but generally requires much higher absorber concentrations, making it of limited use for sites beyond polluted urban areas.

F. Combustion and Other Techniques

Another class of techniques employs thermal heating of the sample and detecting of evolved CO₂ to determine total carbon and, in conjunction with some optical device, also the light absorbing fraction. The process destroys the sample and often involves the pyrolysis (charring) of organic carbon into artefact BC in the process. The appropriate interpretation of the optical signal and evolved CO₂ in this process as an absorption coefficient is complicated and under question (Huffmann, 1996).

Other techniques exist for measurement of σ_a (Gerber and Hindman, 1982b) but are generally limited to laboratory or special activities due to their difficulty for field implementation or their lack of sensitivity under clean conditions. A very promising Raman spectroscopic technique was introduced by Rosen et al. (1978) to identify graphitic carbon as the major light absorbing component of anthropogenic aerosols. Recently this technique has been developed further to quantify graphitic carbon on the same Nuclepore^R substrate that is used for light absorption measurements (Keller and Heintzenberg, 1997).

G. Inverse Methods for Deriving the Complex Refractive Index

Inverse methods utilise various multivariate optical data along with assumptions and Mie computations to fit the optical data to a value of ω . In general, inverse methods are ill-posed and the solutions are non-unique (e.g., Fymat and Zuev, 1978). They are ill-posed because the measured quantities are dependent on other (assumed) parameters such as the form of the particle size distribution. In principle, the use of a refractive index implies a homogeneous material interacting with the electromagnetic waves which often is not given in the atmospheric aerosol. Furthermore, the retrieved values of the complex refractive index

or ω are columnar quantities (integrated over varying relative humidities) and thus are not directly comparable to the results from in situ methods. Therefore, the complex refractive index obtained from inverse methods is often called an "effective", "optically equivalent" or "apparent" quantity. The results do represent a measure of the optical properties of the atmospheric column but only for the case in which the assumptions made in the retrieval procedure are valid. Also, in general, inverse methods have not been tested against direct measurements of σ_a , ω or complex refractive index.

One group of inverse method utilises solar broad band (Herman and Browning (1975) or spectral (Tanaka et al., 1990) irradiance data, sometimes in combination with LIDAR (Reagan et al., 1980). The different techniques mainly differ in the assumptions made about the aerosol size distribution, the real part of refractive index and the surface albedo.

Another group of methods inverts measurements of the angular distribution of the scattered radiation to estimate the complex refractive index. Qia and Xiuji (1986) consider specific scattering angles to retrieve separately the real and imaginary part of refractive index. However, their approach is questionable, because the specified scattering angles strongly depend on aerosol size distribution and on the assumption of homogeneous spheres. Tanaka et al. (1982) show that using polarisation data are particularly sensitive to the imaginary part of refractive index. The backwards hemisphere of scattering is very sensitive to the shape of the particles but the forward hemisphere is not (von Hoyningen-Huene and Posse, 1997). Thus, restriction to the aureole region of scattering angles avoids problems due to any nonsphericity of the aerosol particles. Combining scattering measurements with simultaneous irradiance data reduces inherent uncertainties in this technique (Nakajima et al., 1996).

Two level flux measurements have been used to derive the average aerosol absorption in the intermediate layer (Pilewskie and Valero, 1992; Valero et al., 1984). As with the inverse methods discussed above this approach involves assumptions about the atmosphere which make it difficult to assess the uncertainty of the result.

For climate considerations global approaches are needed, therefore satellite retrievals have received increasing attention. Kaufman (1987) applies an iterative method to AVHRR signals for inversion to aerosol single scattering albedo. Uncertainties in the aerosol particle size distribution strongly limit the accuracy of this method. A sensitivity study has yielded

an uncertainty of ± 0.05 in the retrieved single scattering albedo within the assumptions of size distribution and homogeneous spheres. The author states that validation of this value is very complicated. (Hsu et al., 1996) derive a new aerosol absorption index from TOMS reflectivity data. However, this index is only used to detect absorbing material, it does not yield quantitative optical results, and there is no stated direct link to the imaginary part of refractive index, σ_a or ω .

H. Calculation of ω from 3-d Chemical / Transport Models

Besides the instrumental methods that exist, it is also possible to estimate single scattering albedo from models that are used to calculate the mass concentration of atmospheric aerosol substances. One such effort was based on 3-d models of carbonaceous aerosols (including sub-models for organic and black carbon constituents) and sulfate aerosols, with the assumption that no other aerosols were involved (Liousse et al., 1996). Smoke particles from biomass burning and fossil fuel combustion are the assumed main input of organic and black carbon aerosols. A 3-dimensional map of the concentration of both sulfate and organic materials in the aerosols is calculated. From this map and an assumed, empirical scattering efficiency (in $\text{m}^2 \text{g}^{-1}$) the scattering coefficient also is calculated. The calculated 3-dimensional map of light absorbing or black carbon is then used with an absorption efficiency to calculate the map of absorption coefficient.

With this approach, it has been possible to estimate the global geographical distribution of ω , which cannot be done with limited numbers of in situ observations. Data from the maps thus obtained are compared to the in-situ data in Figures 1 and 2 (see discussion in Section 1 above). We note though that present 3-d chemical modelling excludes the possibility of simulating adequate aerosol mixtures, hence does not offer realistic ways of calculating single scattering albedo.

Uncertainties in this calculation arise from uncertainties in all of the input quantities, especially from the amount of biomass that is burned (factor of 3). Additional sources of uncertainty exist, e.g. for fossil fuel the emission factor for BC particles (at least a factor of five), and the ratio of organic to black carbonaceous matter (perhaps on the order of $\pm 50\%$), the height of injection into the atmosphere ($\pm 30\%$), and the size-dependent deposition rate of the aerosol. The overall uncertainty has not yet been estimated.

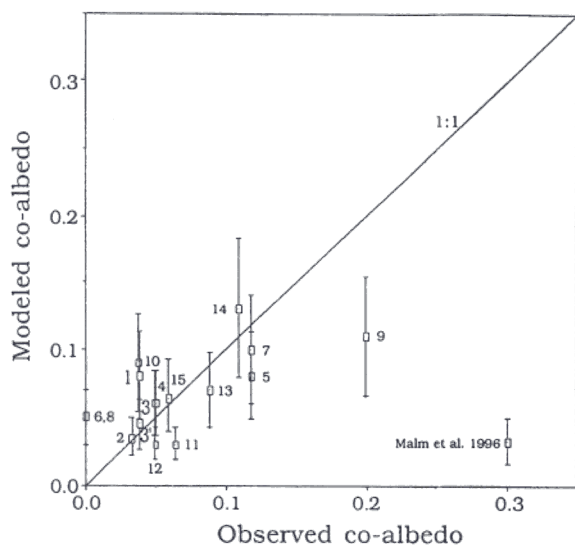


Figure 1: Measured vs. modelled aerosol co-albedo. The data are taken from Table 1 and Malm et al. (1996). The 1:1 relationship is marked with a thin line.

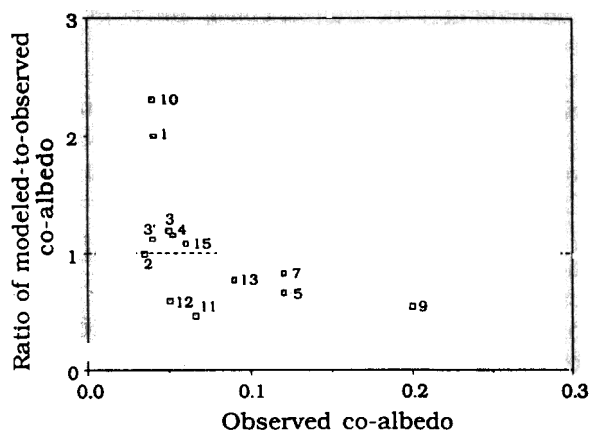


Figure 2: Ratio of modelled-to-measured dry co-albedo vs. measured co-albedo. The data are taken from Table 1, # 6 and 8 removed.

One factor that has not been included is the calculation of ω for the “preindustrial” case for comparison to the current picture. Ultimately, the method would involve calculating the preindustrial aerosol loadings and properties and the forcing it caused, then calculating the current forcing with both natural and anthropogenic aerosols present; the difference is the anthropogenic climate forcing, of course with an overall uncertainty that results from all of the constituent uncertainties.

I. Extinction and Determination of σ_a as the Difference of σ_e and σ_s

Perhaps the most direct and absolute method for determining light absorption coefficient, and the necessary inputs to the determination of ω consists of separate measurements of the extinction coefficient via long path extinction and the scattering coefficient with an integrating or polar nephelometer (Waldram, 1945). The difference between the two is the absorption coefficient. The current state of the art in nephelometry allows such measurements to be made literally anywhere in the troposphere with an accuracy of perhaps 7%; the main problem is measuring σ_e because the small magnitude of σ_e in most of the atmosphere precludes its direct measurement with available instruments. Typical values of σ_e in polluted air may approach 10^{-3} m^{-1} , while industrial regions often have values of 10^{-4} , and remote areas typically have aerosol components of extinction that are less than that due to Rayleigh scattering by air molecules (ca. 10^{-5} m^{-1} at $\lambda = 550 \text{ nm}$ and at sea level). Measuring extinction in such locations for regional or continental situations requires path lengths that yield optical transmissions that are significantly less than unity, e.g. σ_e times path length is of order unity. Industrial regions would require 10 km, and cleaner areas 100 km or more, which clearly is not practical. Thus, the measurement of extinction is limited to the places of highest aerosol content (e.g., in the densest of industrial and biomass burning regions) and to laboratory settings where high aerosol contents can be generated.

This approach is currently the method of choice in many situations for laboratory calibration of absorption methods that utilise filter substrates. The problem in such calibrations is the generation of aerosols in the laboratory that accurately simulate the microphysical characteristics and the optical characteristics of real atmospheric aerosols.

4.2 Calibrations and Intercomparisons

From the discussion above one might assume that the issue of instrument calibration and intercomparison would be a common and repeated effort. However, we are aware of only one formal effort to assemble and intercompare light absorption techniques with controlled aerosols. This was at the First International Workshop on Light Absorption by Aerosol Particles at Colorado State University in July and August of 1980. The major activities and experiments have been summarised in the above mentioned text

(Gerber and Hindman, 1982a) and selected papers from some participants were published in *Applied Optics* (February, 1982). Though incomplete in some ways, this first intercomparison did provide an opportunity to evaluate a number of techniques and identify their strengths, weaknesses and problems. Regrettably, there has been no follow-up to this workshop and many of the issues raised remain unresolved and persist in some of the measurements to this day.

Some individuals and small groups have worked to understand measurement differences and uncertainties. An intercomparison of the IPM, IS and ISW instrumentation demonstrated that these techniques provided similar measurement of the aerosol light absorption coefficient to within about $\pm 30\%$ and that the measured and modelled optical properties were consistent with behaviour expected from Mie theory for the size distributions and compositions that were studied (Clarke et al., 1987). Other individual efforts at calibration exist (Hitzenberger, 1993; Hitzenberger and Puxbaum, 1993). Notably, the whole family of inverse methods has never been systematically compared to the instrumental approaches. Hence, the uncertainties in such methodologies are totally undefined.

4.3 Atmospheric Data

A full discussion of existing global measurements of σ_a , σ_s and $(1 - \omega)$ is beyond the scope of this paper. Some of these measurements are incorporated into data sets as part of monitoring programs and often emphasise local or city pollution events e.g. are focused on health effects (WMO). Small amounts of data are available on the regional to global scales that are more appropriate for comparisons to models and the concerns of this paper. Even so, some general observations can be made for measurements in remote regions. It is important to recognise that for the measurement of σ_s we know that the role of increasing relative humidity is to increase σ_s and thereby lower co-albedo. In general, nephelometers are operated at temperatures above ambient conditions. Thus, σ_s measurements will be made at lower *RH* and therefore will result in lower than ambient values. Also, for remote regions, the loss of coarse but non- or weakly absorbing sea-salt and soil dust particles before entering the nephelometer will also lower σ_s and thereby lead to somewhat higher measured co-albedo values. However, under some conditions, this loss may partially compensate for the fact that sea-salt exists at generally higher concentrations

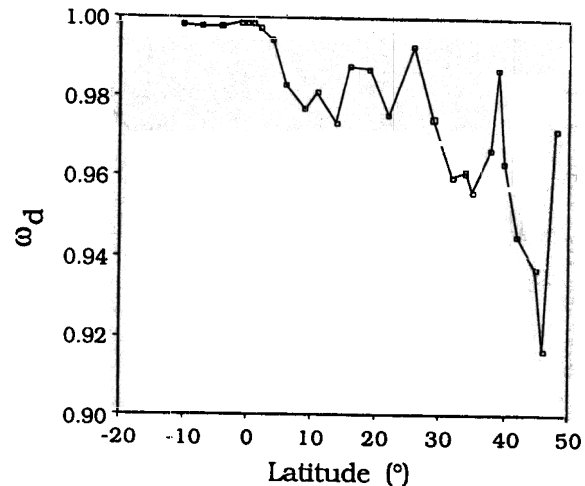


Figure 3: Dry particle single scattering albedo (ω_d) measured at sea level during the NOAA RITS-88 cruise down the central Pacific from Dutch Harbor (Alaska) to Samoa in March 1988.

near the sea-surface so these measurements may yet reflect column co-albedo values.

Observations of ω using the ISW and IS methods that minimise the artefact enhancement of σ_a mentioned above, and generally carried out for “dry” aerosol, have been reported for several remote regions over the globe (Clarke, 1989; Heintzenberg and Ogren, 1986; Lannefors et al., 1983). These data reveal that extended regions over the South Atlantic Ocean at times have low “dry” values $\omega_d = 0.8$ due to biomass burning and $\omega_d = 0.85$ for spring Arctic haze but also show extensive regions of the tropical and S. Pacific and Indian Oceans with $\omega_d = 0.99$ or higher. The appropriate ambient values of ω will, of course, be higher due to relatively higher σ_s values at ambient *RH*. The major sources of non-crustal light absorbing particulate matter often show considerable annual variations while light scattering aerosol components have different or less seasonality. In the northern hemisphere, the Arctic aerosol is one such example with $\omega_d = 0.94$ and 0.86 in summer and winter, respectively (Heintzenberg and Leck, 1994). Even in remote regions in the southern hemisphere, similar seasonal variations can be expected due to the seasonal variations of σ_a and σ_s (Heintzenberg and Bigg, 1990).

Figure 3 shows an example of ω_d for a transect down the central Pacific during March of 1988 aboard a NOAA-RITS (Radiatively Important Trace Species) cruise. This clearly illustrates the impact of BC, primarily from Asian sources over the North Pacific. Absorption diminishes upon approach to the inner-tropical convergence zone, where there is an abrupt

transition to a virtually non-absorbing aerosol in the Southern Hemisphere ($\omega_d \approx 0.99$). These measurements took place during the season when a maximum impact of BC on σ_a in the North Pacific has been identified (Clarke and Charlson, 1985). Hence, consideration of the ambient and annual average values can be expected to show significantly less impact. This figure also illustrates that σ_a and σ_s are independent variables.

5 Problems

While progress has been made on measurements, calibration, models and field observations, and while the capability to make reasonably accurate measurements of ω exists, several serious problems have emerged recently, leading to very large uncertainties in some data sets, e.g., a factor of three or more in the magnitude of σ_a , as is evident in the distinct outlier in Figure 1. These difficulties can be summarised:

- Lack of calibration and the need for a primary standard. There are three methods which might be considered to be primary standards for σ_a : (1) The difference between σ_z determined with a transmissionmeter and σ_s determined with a nephelometer. This method is only useful with very high values of σ_a , as in chimneys, plumes of forest fires or laboratory settings. (2) Absorbance determination with an integrating sphere, operated as such (Heintzenberg, 1982b). (3) Photoacoustic determination (Roessler and Faxvog, 1980). While the latter two of these were used in the only large-scale intercomparison (Gerber and Hindman, 1982b) none of the three is today commonly available, and most of the recently published data have been acquired with methods that were not included in that intercomparison. Specifically, the aethalometer (Hansen et al., 1982) and the laser integrating plate method (LIPM, Campbell et al., 1995) used in the IMPROVE network have not been calibrated with one of the possible standard optical methods. In addition, both appear to yield values of σ_a that are high, perhaps by large amounts (again, cf. Figure 1). In the latter case, it is possible that the overestimate of light absorption by LIPM is itself the main cause of the unusually low value of ω around 0.7. Other causes of high light absorption coefficient cannot be ruled out, e.g., local contamination at the IMPROVE sites in the US National Parks that would not be captured by the modeling. It is important to recognise that the errors in the

aethalometer, LIPM and "raw" IPM that have been identified so far yield measured values that exaggerate light absorption; that is, the errors are all in the same direction. If a calibration study confirms this finding, it will be necessary to correct (or eliminate from use in climate models) large amounts of published data, yielding higher values of single scatter albedo. When improving experimental techniques, it must be kept in mind that radiative transfer calculations need ω as a function of wavelength.

- Uncertainties of similar or even larger magnitude (i.e. several fold) exist in estimates of the source strengths of the light absorbing materials (graphitic or organic light absorbing carbon, soil dust etc.). The commonly used anthropogenic source strength of "black carbon" Q(BC) of ca. 12 Tg a⁻¹ (Cooke and Wilson, 1996; Liousse et al., 1996) appears to be high relative to the SO₄²⁻-source strength when compared with field measurements of light absorption made with the calibrated light absorption methods and with chemical determinations of the carbon/sulphate ratio in rural areas of the industrialised continents and in remote regions. The observed BC/SO₄²⁻-ratio at the surface ranges from 0.03 in the summer Arctic (Heintzenberg and Leck, 1994) to 0.11 in urban Hamburg (Heintzenberg and Winkler, 1984), with 0.05 in Swedish rain (Ogren and Charlson, 1984) and 0.08 over a year in Hungary (Heintzenberg and Mészáros, 1985), with much lower values in broad areas over the Pacific Ocean (Clarke, 1989). Urban values are similar to the estimated ratio of source strengths of BC and BC/SO₄²⁻ of ca. 0.1 (including contemporal and fossil fuel combustion sources), but do not represent all of the globe or even the northern hemisphere. Also, cities and industrial sites emit SO₂ that will react to SO₄²⁻ over a few days time, decreasing the ratio of BC/SO₄²⁻ in more remote places. Some of the discrepancy may be due to a higher ratio of fossil/contemporal sources in these continental NH sites. But, it is apparent in Table 1 and Figures 1 and 2 (i.e., from the comparison of measured and modelled values of co-albedo) that the modelled values of co-albedo are high for sites that are well away from sources of BC, which could be caused by overestimates of the source strength of BC. We note that (while there are few data) such remote sites may represent most of the globe. To be sure, we do not have enough field data to define the ratio globally; in particular, we only have surface data, and most

of it is from sites that are close to industrial regions (where both fossil and biomass sources are present) or close to biomass burning regions. It is easy to explain qualitatively that the known errors in the magnitude of source strength of BC all tend to yield overestimates rather than underestimates (Bond et al., 1997). This is because at least some of the source strengths are estimated from total mass emissions, with assumptions that BC forms a large fraction of the total mass and that the particle sizes are all small enough to have significant atmospheric residence times.

- There is a serious lack of data with calibrated instruments in crucial regions of the globe, particularly in rural to remote regions of the NH. The data base for smoke from biomass combustion appears more extensive than that for industrial regions. Most of the data have been and are being taken at the surface while we need vertical distributions of aerosol-optical properties.
- There also is a lack of sensitivity tests of the chemical transport models of the geographical distribution of BC to the source measurement uncertainties, and the inherent variability of the absorption efficiency of BC.
- A potential exists for ambiguity in the measurements of the graphitic carbon or black carbon component of the aerosol due to the combustion analysis that is widely used. Artefact light-absorbing materials are generated by the heating that is used and the correction scheme with reflectance may produce errors that have not been evaluated. Further, differences in the rate of heating, the presence or absence of O₂, the duration of heating etc. are not standardised. Clearly, alternative methods of analysis and a calibration of this method against a primary standard of absorption are needed. In addition, other light absorbing matter, e.g. soil dust and/or humic substances may contribute light absorption in addition to BC, requiring still further improvements in the chemical analyses.

6 Prospects and Conclusions

While there is a clear need for coordination of modelling and measurement activities (both chemical and climate-type of models), and while there are plenty of opportunities within the context of large international

programs (e.g. IGAC³) only a very limited amount of effort appears to be in progress. In particular, there is a clear-cut need for standardisation and intercalibration of measurements of light absorption coefficient and of its use in calculating ω . This effort should be international in scope, and would be most effective if it occurred soon.

In the meantime, great caution should be used by those engaged in modelling of climate forcing and response; most especially to recognise the paucity of data from calibrated measurements, the need for inclusions of the influence of RH on σ , and therefore ω , and the requirement to identify data from uncalibrated systems as being of undefined uncertainty.

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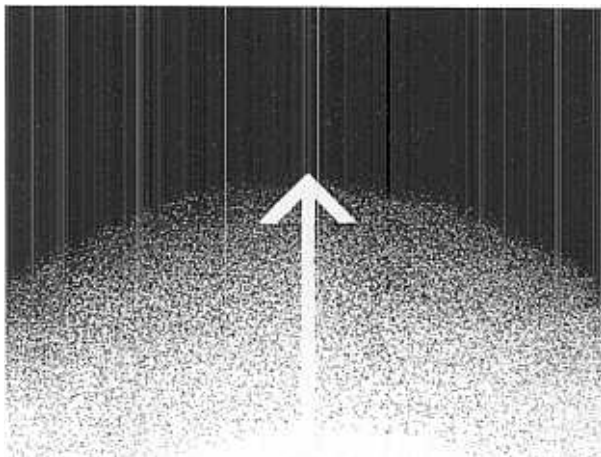
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³ IGAC = International Global Atmospheric Chemistry Programme

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