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## Comparisons of techniques for measuring shortwave absorption and black carbon content of aerosols from biomass burning in Brazil

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**Abstract.** Six methods for measuring the shortwave absorption and/or black carbon (BC) content of aerosols from biomass burning were compared during the Smoke, Clouds, and Radiation-Brazil (SCAR-B) experiment. The methods were the optical extinction cell (OEC), integrating plate (IP), optical reflectance (OR), particle soot/absorption photometer (PSAP), thermal evolution (TE), and remote sensing (RS). Comparisons were made for individual smoke plumes and for regional hazes dominated by smoke. Taking the OEC as a primary standard, measurements of the absorption coefficient ( $\sigma_a$ ) showed that the OR method had the lowest uncertainty (17%) in  $\sigma_a$ . The other optical methods had uncertainties ranging from 20 to 40%. However, with sufficient sample size, the values of  $\sigma_a$  derived from the optical methods converged to within 20% of each other. For biomass burning aerosols in regional hazes over Brazil, this led to systematic differences of  $\pm 0.02$  in the values of the single-scattering albedo derived from the various in situ techniques. It was found also that the BC content of the aerosol and  $\sigma_a$  were poorly correlated. This is likely due to a large uncertainty in the BC content of the aerosol measured by TE, and/or a high variability in the mass absorption efficiency of BC in biomass burning aerosol. Hence there is a high uncertainty in inferring  $\sigma_a$  from the BC content of smoke aerosol.

### 1. Introduction

It has been hypothesized that anthropogenic aerosols have a net cooling effect on the atmosphere by reflecting shortwave solar radiation back to space [e.g., Charlson *et al.*, 1991]. One of the most important parameters required to adequately model the direct radiative forcing of aerosols is their single-scattering albedo ( $\omega_0$ ), which depends on the absorption of solar radiation by the aerosol and therefore on their black carbon content.

Aerosols from biomass burning contribute to direct radiative forcing [Penner *et al.*, 1992; Hobbs *et al.*, 1997]. However, because the single-scattering albedo of biomass burning aerosol lies in the range 0.7 to 0.9 [Radke *et al.*, 1988, 1991; Hobbs *et al.*, 1996, 1997; Reid and Hobbs, this issue; Reid *et al.*, this issue], much of the potential cooling effect of these aerosols is mitigated [Hobbs *et al.*, 1997]. For highly reflective underlying surfaces ( $R_s > 0.2$ ) such as deserts, some grasslands, and clouds, aerosols with single-scattering

albedos in the range 0.7 to 0.9 can even change the sign of the direct radiative forcing, so the surface temperature perturbation shifts from cooling to heating [Chylek and Coakley, 1974; Hansen *et al.*, 1997]. Therefore it is important to quantify the precision with which the single-scattering albedo of aerosols from biomass burning can be presently measured.

While there are many methods currently in use to measure the absorption coefficient and black carbon content of aerosol particles, there is much debate as to which methods, if any, yield accurate values. Furthermore, intercomparison studies have shown that aerosols containing significant organic components, such as smoke particles from biomass burning, are among the most difficult to characterize [Shah and Rau, 1991].

During the Smoke, Clouds, and Radiation-Brazil (SCAR-B) field project, which was carried out in Brazil from August 17 to September 20, 1995, we employed six of the most commonly used methods to determine the light absorption and/or black carbon content of aerosols dominated by smoke from biomass burning in the cerrado and (Amazon) forest regions of Brazil. These methods were as follows: an optical extinction cell (OEC), the integrating plate (IP) technique, optical reflectance (OR), particle/soot absorption photometry (PSAP), thermal evolution (TE), and remote sensing (RS). Except for RS, data from all of these techniques were obtained from in situ instrumentation or filters collected aboard the University of Washington (UW) Convair C-131A research aircraft [Hobbs, 1996]. The purpose of the study described here was to determine the uncertainty in the measurements of the black carbon content, absorption coefficient, and single-scattering albedo of the biomass burning aerosol derived from these various techniques.

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## 2. Terminology

It is generally assumed that almost all of the atmospheric absorption of shortwave radiation by particles in polluted air is due to black carbon (BC), which is composed of a variety of soot-based species. Black carbon was once believed to be primarily graphitic (elemental) carbon and is frequently modeled as such. However, it is now realized that there are a variety of soot-based species mixed with nonabsorbing compounds that absorb light with various degrees of efficiency [Lioussse *et al.*, 1993]. The methodologies employed to determine the BC content and the absorption properties of an aerosol are based on the optical and chemical characteristics of BC.

It is frequently assumed [e.g., *Intergovernmental Panel on Climate Change (IPCC)*, 1994] that the mass concentration of BC ( $c_{\text{mbc}}$ ) in the fine-mode aerosol size range (diameter  $<2.5$   $\mu\text{m}$ ) and the absorption coefficient of the aerosol ( $\sigma_a$ ) are related through a mass absorption efficiency ( $\alpha_{\text{abc}}$ ):

$$\sigma_a \equiv \alpha_{\text{abc}} c_{\text{mbc}} \quad (1)$$

Two methodologies are commonly employed to measure  $\sigma_a$ . The most direct method involves determining the optical characteristics of the aerosol, usually by measuring the attenuation of a light beam through a sample of the aerosol. The aerosol sample can be collected on a filter or be in the free atmosphere. Alternatively, the BC content of the aerosol can be measured and  $\sigma_a$  determined from (1) by estimating or measuring a value for  $\alpha_{\text{abc}}$ .

We will assume, for comparison purposes only, that  $\alpha_{\text{abc}}$  is independent of wavelength and has a mean value of  $15 \text{ m}^2 \text{ g}^{-1}$  [Lioussse *et al.*, 1993]. However, many investigators [e.g., Waggoner *et al.*, 1981; Rosen and Hansen, 1984; Lioussse *et al.*, 1993; Chylek *et al.*, 1995; Martins *et al.*, this issue (a)] have demonstrated that the mass absorption efficiency and wavelength dependence of BC is variable and is strongly dependent on the way it is distributed in the aerosol (e.g., internally or externally mixed). Model calculations indicate that nonabsorbing coatings on a BC particle may produce  $\alpha_{\text{abc}}$  values from 7 to  $30 \text{ m}^2 \text{ g}^{-1}$ , as well as introduce a wavelength dependence [Chylek *et al.*, 1995; Martins *et al.*, this issue]. For example, it has been shown that as smoke ages, changes in the physical and chemical properties of the aerosol can vary  $\alpha_{\text{abc}}$  in the range  $15\text{--}25 \text{ m}^2 \text{ g}^{-1}$  [Lioussse *et al.*, 1995]. For a discussion of the absorption properties of the biomass burning particles measured in SCAR-B, the reader is referred to Martins *et al.* [this issue].

When a value for  $\sigma_a$  is established, and the atmospheric light-scattering coefficient  $\sigma_s$  is measured with a nephelometer,  $\omega_o$  is given by

$$\omega_o \equiv \frac{\text{light scattering}}{\text{light extinction}} = \frac{\sigma_s}{\sigma_s + \sigma_a} \quad (2)$$

Typically, errors in measured values of  $\sigma_a$  are large. It should be noted that  $\omega_o$  goes to unity as  $\sigma_a$  goes to zero, and  $\omega_o$  asymptotically approaches zero as  $\sigma_a$  becomes very large. Hence if the single-scattering albedo is 0.98, an error of 50% in  $\sigma_a$  produces a negligible effect ( $<0.01$ ) in the value of  $\omega_o$ . However, if the single-scattering albedo is 0.85 (a reasonable value for biomass burning aerosol), an error of 50% in  $\sigma_a$  has a substantial effect (0.06) on the value  $\omega_o$ . Hansen *et al.* [1997]

point out that when  $\omega_o$  reaches a value between 0.85–0.9, the radiative forcing of an aerosol changes from cooling to warming. Therefore an error in  $\omega_o$  of 0.06 could prevent determination of even the sign of the direct radiative forcing. Because  $\omega_o$  is sensitive to errors in  $\sigma_a$  in the range of  $\omega_o$  values most critical for assessing the sign of the radiative forcing, it is important to quantify the uncertainties in current measurements of  $\sigma_a$ .

Errors in the nephelometric measurement of  $\sigma_s$  derive from two principle sources: truncation errors and non-Lambertian light sources. Truncation errors arise since the detectors used in most nephelometers view roughly only  $7^\circ\text{--}170^\circ$  of the scattering. Thus scattering in the far forward and backward directions are not measured. For example, for an aerosol with a volume median diameter of 0.30 and a geometric standard deviation of 1.8 (typical for the aerosols being considered here [Reid *et al.*, this issue], the truncation error is about 4%. This error is accounted for, in part, in the calibration of a nephelometer (e.g., calibration with a Rayleigh scatter retrieves  $\sim 2\%$  (or half) of the truncation error). Fortunately, scattering by biomass burning aerosol is essentially limited to particles less than 0.7  $\mu\text{m}$  [Reid and Hobbs, this issue], and the calculated truncation error based on Reid *et al.* measured aerosol size distributions is less than 2%.

An ideal nephelometer would have a Lambertian light source; that is, it would emit light with equal intensity in all directions. In most nephelometer designs a milk-glass window is placed in front of the light source in an attempt to achieve this ideal condition. However, recent measurements have shown that a milk-glass window emits more light at  $90^\circ$  and less light at low angles than in other directions [Anderson and Ogren, 1998]. This can cause underestimates of scattering by submicron and supermicron aerosols of almost 8% and 60%, respectively. However, the principal nephelometer aboard the C-131A in SCAR-B was custom built for us by M. E. Electron to diminish this effect. Because of these modifications and because 95% of the light scattering by biomass burning aerosol is by fine particles, the sum of the truncation and non-Lambertian errors in our light-scattering measurements was  $<5\%$ . For an average value of  $\omega_o$  of 0.85 an error in  $\sigma_s$  of 5% results in an error in the derived single-scattering albedo of only 0.005. Because the errors in our measurement of light scattering were low relative to those of absorption, and the same light-scattering values are applied to all of the absorption measurements, in the remainder of this paper we will ignore the uncertainty in the scattering measurements.

## 3. Measurement Techniques

In this section we describe the techniques used for measuring the absorption coefficient and BC content of the aerosol from biomass burning in Brazil under the following headings: direct optical, chemical speciation, and remote sensing.

### 3.1. Direct Optical Methods

Direct optical methods are a group of techniques in which light absorption is derived from measurements of the optical properties of aerosol. The aerosol may be suspended in the free atmosphere or collected on a filter. In the present study, four direct optical methods were employed, which are described below.

**3.1.1. Optical extinction cell (OEC).** The OEC aboard the UW aircraft is, in effect, a 6.4 m long transmissometer [Weiss and Hobbs, 1992]. The OEC provides direct measurements of the extinction coefficient of the aerosol ( $\sigma_e = \sigma_s + \sigma_a$ ) by measuring the attenuation of a light beam (at a wavelength of 538 nm) which passes through a 6.4 m long tube containing a sample of the aerosol. A nephelometer simultaneously measures  $\sigma_s$ . Subtraction of  $\sigma_s$  from  $\sigma_e$  yields  $\sigma_a$ , and  $\omega_0$  is derived from (2).

When measurements were available from the OEC, it was considered the primary standard for determining aerosol absorption. With a path length of 6.4 m the OEC can be used to measure  $\sigma_e$  only for an extremely high mass concentration of aerosol, such as is present in smoke plumes. Also, a large concentration gradient of aerosols is needed to obtain a high value for the in-plume/clean air ratio, which is needed to correct for detector drift. The reported single-scattering albedo is an average across the plume (six to ten 1 s data points). For thick smoke plumes, such as those observed in SCAR-B, errors in the measurement of  $\sigma_e$  by the OEC were  $\leq 2\%$ . Since  $\sigma_a$  is obtained from the difference in two large numbers ( $\sigma_e$  and  $\sigma_s$ ), small errors in the measurement of either of these two quantities can result in large errors in  $\sigma_a$ . For example, for a single-scattering albedo of 0.85, a 2% uncertainty in  $\sigma_e$  yields an uncertainty of 10% in  $\sigma_a$  and a 2% ( $0.85 \pm 0.02$ ) error in  $\omega_0$ .

**3.1.2. Integrating plate (IP).** In the IP technique, the aerosol is collected on a filter, and  $\sigma_a$  is derived by measuring the attenuation of a light beam through the filter [Lin et al., 1973]. Calibration factors, based on aerosol loading and the filter substrate, are applied. Because the particles are removed from the atmosphere, and measurements are made in the laboratory, the absorption coefficient measured is for dried aerosol.

The value of  $\sigma_a$  is obtained from

$$\sigma_a = \left(\frac{c_s}{R}\right) \left(\frac{A}{V}\right) \ln\left(\frac{I_0}{I_f}\right) \quad (3)$$

where  $I_0$  and  $I_f$  are, respectively, the intensity of light transmitted through the filter before and after sampling the aerosol,  $A$  is the area of the aerosol deposit on the filter,  $V$  is the volume of air sampled,  $R$  is an empirical correction factor that varies with the amount of aerosol on the filter, and  $c_s$  is an empirical factor that corrects for multiple scattering and depends on the nature of the filter and the apparatus used.

Because  $I_0$  and  $I_f$  involve both the aerosol and the substrate filter, it is advisable to use only substrates that have relatively low optical depths (polycarbonate and stretched Teflon filters are commonly used). For stretched Teflon filters (which were used in this study), few calibration studies have been performed. However, the recommended values of  $c_s$  range from 0.5 to 0.97 [Campbell et al., 1995, 1996; Clarke et al., 1996]. This range of values leads to an uncertainty in  $\sigma_a$  of 100%. For the aerosols observed in SCAR-B, this uncertainty in  $c_s$  can result in a perturbation in the derived value of  $\omega_0$  of 0.1. The values of  $c_s$  used for this study was 0.5.

For the factor  $R$ , which allows for "shadowing" on heavily loaded filters, we used the expression recommended for the IMPROVE network standard operating procedures [Campbell et al., 1989]:

$$R = 0.36 \exp\left[-\frac{\rho}{22}\right] + 0.64 \exp\left[-\frac{\rho}{415}\right] \quad (4)$$

where  $\rho$  is the aerial density (aerosol mass/A) of the sampled aerosol.

The integrating plate used in this study was manufactured by Radiance Research and was operated at a wavelength of 550 nm. The Teflon filters were also weighed on a microbalance in a humidity-controlled chamber to an accuracy of  $\pm 6 \mu\text{g}$  ( $\pm 1 \mu\text{g}$  precision) to determine the mass concentration of the dried aerosol. The total filter loadings ranged from 50 to 150  $\mu\text{g}$ .

**3.1.3. Optical reflectance (OR).** This is a reflectance analog of the IP technique [Delumea et al., 1980]. Aerosol particles are collected on a smooth polycarbonate filter. Subsequently, the filter is illuminated from above and the amount of light it reflects is measured. The amount of reflected light is used to deduce the absorption by aerosols on the filter by comparing it with a calibration curve. This curve was developed from measuring the light reflected from a series of filters with various loadings of graphitic carbon spheres. The absorption coefficient  $\sigma_a$  is then deduced from equation (1) using a value for  $\alpha_{abc}$  that is specific to the standard employed. It should be emphasized that this value of  $\alpha_{abc}$  is used to obtain the correct value of  $\sigma_a$ ; it is not necessarily applicable to a specific aerosol sample.

The OR apparatus used in this study was manufactured by Diffusion Systems and was operated in a broadband visible mode. The BC standard used was the Monarch 71, with a mean geometric radius of 0.15  $\mu\text{m}$  and a geometric standard deviation of 1.69. The soot standard Monarch 71 was introduced by Heintzenberg [1982] as a reference material for light absorption measurements and is commercially available through the Cabot Corp. Black carbon loadings on each filter standard were determined gravimetrically. The value of  $\alpha_{abc}$  of 6.8  $\text{m}^2 \text{g}^{-1}$  was used for this standard and was determined through Mie theory calculations based on the Monarch 71 size distribution and an index of refraction of 2.0-1i. This value of  $\alpha_{abc}$  is in agreement with calibration studies using the Monarch 71 by Clarke et al. [1987].

**3.1.4. Particle soot/absorption photometer (PSAP).** This technique is an extension of the IP technique to continuous sampling and measurement. Aerosols are continuously collected on an illuminated filter substrate, and measurements of the attenuation of a light beam through the filter yields  $\sigma_a$ . It is subject to many of the same errors as the IP technique.

The instrument used in this study was the particle soot/absorption photometer (PSAP), manufactured by Radiance Research. The PSAP operates at a wavelength of 550 nm and uses a quartz filter substrate with a  $c_s$  value of 0.5 and an  $R$  value of  $0.355 + 0.5398 \text{ Tr}$ , where  $\text{Tr}$  is the transmittance relative to a clean filter. The integration time for each data point was 1 min.

## 3.2. Chemical Speciation Methods

If the BC content ( $c_{\text{mbc}}$ ) is measured chemically,  $\sigma_a$  (and hence  $\omega_0$ ) can be estimated from equation (1) using an assumed value of  $\alpha_{abc}$ .

The primary procedure used to determine the BC content of an aerosol sample is through the use of various thermal evolution (TE) methods. All of the TE techniques involve the thermal separation of the carbonaceous aerosols into BC and organic carbon (OC) [Novakov, 1982]. Aerosols that are to be subjected to TE are generally collected on quartz filters. A thermal pretreatment is performed on the quartz filter sample to remove organic matter. The remaining carbon content

(presumed to be BC and soil carbonates) is determined by coulometric titration. There are many thermal methods in use each with unique procedural modifications [e.g., *Cadle et al.*, 1980; *Novakov*, 1982; *Tanner et al.*, 1982; *Huntzicker et al.*, 1982; *Ellis et al.*, 1984; *Ohta and Okita*, 1984; *Cachier et al.*, 1989]. Intercomparison studies [*Shah and Rau*, 1991] show that if the aerosol has a high organic carbon content (such as biomass burning aerosol), minor procedural differences can result in >50% differences in the derived value of the ratio of BC to OC.

During collection, volatile organic compounds can adsorb onto the quartz fibers causing, on average, a positive 15 to 20% artifact [*Appel et al.*, 1989]. Conversely, the collected aerosols may volatilize from the filter. During the sampling of biomass burning aerosols, the former artifact can be so large that if a substantial amount of aerosol is not collected (less than 50  $\mu\text{g C}$  per filter), the derived value of the apparent organic carbon content can be larger than the total aerosol mass [*Martins et al.*, 1997]. Although this artifact does not influence the collection of nonvolatile BC, it can interfere with the thermal analysis.

The TE technique is subject to several other artifacts, including pyrolysis of organic matter (charring) and its conversion into BC [*Cadle et al.*, 1983; *Tanner et al.*, 1982], and the premature release of BC at low temperatures. These two artifacts can lead, respectively, to overestimates and underestimates of  $c_{\text{mbc}}$ . The key parameters that determine the importance of these artifacts are temperature, the duration of combustion, and the nature of the carrier gas. The higher the temperature, the less important is charring and the more pronounced is the premature release of BC. For an oxidizing gas, BC removal is more significant [*Cadle et al.*, 1983], although a low gas flow may reduce this effect [*Cachier et al.*, 1989]. With oxygen as the carrier gas, charring is very sensitive to the way in which the temperature is increased. A "flash heating" method (i.e., a few seconds under high temperature) can be used to avoid charring [*Cadle et al.*, 1983]. The optimum temperature for a balance between charring and the premature release of BC is about 340°C [*Cachier et al.*, 1989].

The chemical composition of the aerosol sample can also influence the temperatures at which carbon evolves [*Lin and Friedlander*, 1988]. The presence of potassium and sodium in aerosols may oxidize BC particles, decreasing their temperature of combustion [*Novakov and Corrigan*, 1995].

The specific TE method used in this study was that of *Cachier et al.* [1989]. To evolve the organic carbon, the filter samples were submitted to a temperature of 340°C for 2 hours, although the sample temperatures reach 340°C after 5 min of combustion. The carrier gas was pure oxygen with a low flow rate of 0.6 L/h.

Carbonates (such as contained in soils) also evolve at high temperatures. Without effective carbonate removal or estimation, the value of  $c_{\text{mbc}}$  may be greatly overestimated. Therefore prior to TE, soil carbonates were extracted from our samples through exposure of the filters to HCl vapor for 24 hours. The carbon remaining on the filters after the decarbonation treatment is referred to as atmospheric particulate total carbon (TC). Both TC and BC measurements were performed on filter aliquots by coulometric titration with a Ströhlein Coulomat@702C. The BC fraction was analyzed after the thermal removal of the organic compound. The difference between the TC and the BC was assumed to be OC.

The main uncertainty in the reproducibility of this method is associated with the separation into BC and OC. The published uncertainty in the BC content is of the order of 10% when BC/TC ratio is in the 10-35% range and there is at least 100  $\mu\text{g}$  of carbon per filter [*Cachier et al.*, 1989]. When the BC/TC ratio is >35% or <10%,  $c_{\text{mbc}}$  has been found to be underestimated and overestimated, respectively. To allow for these effects, we used the calibration curves given by *Cachier et al.* [1989].

### 3.3. Remote Sensing (RS) Techniques

Remote sensing measurements from satellites or Sun photometers combined with irradiance data can be used to obtain estimates of the columnar-averaged, single-scattering albedo using radiative transfer theory. In this study, two spectral irradiance models were employed: SPCTRAL2 [*Bird and Riordan*, 1986] and the second simulation of the satellite signal in the solar spectrum, or 6S model [*Vermote et al.*, 1996, 1997].

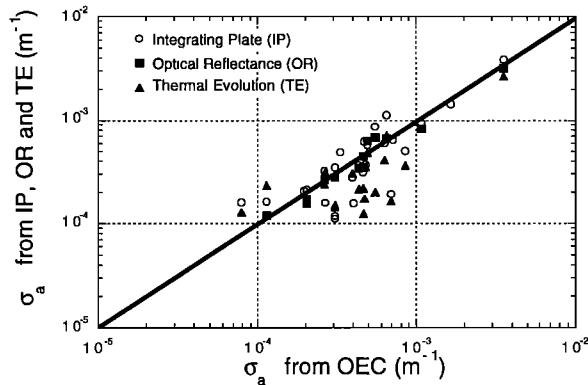
The SPCTRAL2 model is a clear sky parameterization model, which can provide both narrowband (2 nm) and broadband (400-700 nm) irradiance estimates. This model requires as inputs the optical depths at the measured wavelengths  $\tau(\lambda)$ , the aerosol asymmetry factor ( $g$ ), inferred from a Sun-sky scanning radiometer, and the single-scattering albedo ( $\omega_0$ ). Of the three inputs, only  $\omega_0$  is not measured by a Sun photometer. Using the measured values of  $\tau(\lambda)$  and  $g$ ,  $\omega_0$  is varied until agreement is optimized between the irradiances derived from model and the irradiances measured at the surface. For results presented here a broadband photosynthetically active radiation (PAR) sensor (400-700 nm) was utilized to measure irradiance.

The 6S model is a radiative transfer model that relies on the "successive-order-of-scattering" method [*Vermote et al.*, 1996, 1997]. The model includes 13 aerosol layers (2 km scale height) and a spectral resolution of 2.5 nm. This model requires as input the aerosol optical depth at 550 nm,  $\tau(550)$ , the aerosol size distribution, and the refractive index of the aerosol. To derive the aerosol single-scattering albedo,  $\tau(550)$  is measured and a particle size distribution and real refractive index are assumed. The imaginary part of the complex index of refraction is then varied until the modeled and measured irradiances agree. A more detailed discussion of  $\omega_0$  retrieval from measurements of irradiance and aerosol optical properties is given by *Eck et al.* [this issue].

## 4. Results From SCAR-B

In SCAR-B, smoke samples were collected aboard the UW C-131A aircraft in three main locations: Cuiabá (in the state of Mato Grosso), Porto Velho (Rondonia), and Marabá (Pará). The airborne smoke samples can be grouped into three categories. (1) Those obtained in individual smoke plumes within 1 km or so from the fire, where the concentrations and gradients of smoke were high ( $c_m$  from 300 to 2000  $\mu\text{g m}^{-3}$ ); (2) regional haze samples dominated by aged smoke ( $c_m < 100 \mu\text{g m}^{-3}$ ); and (3) samples of regional haze (dominated by smoke) obtained during vertical profiles above ground-based Sun photometers.

When individual smoke plumes were sampled, the PSAP could not be used because of its long time constant. Also, the OEC and TE methods could not be used in regional hazes



**Figure 1.** Comparison of measurements of the absorption coefficient of the aerosol ( $\sigma_a$ ) in regional hazes measured by the integrating plate (IP), optical reflectance (OR), and thermal evolution (TE) techniques versus measurements from the optical extinction cell (OEC).

because the concentrations and gradients of the aerosol were not large enough for accurate measurements.

#### 4.1. Individual Smoke Plumes

**4.1.1. Determination of  $\sigma_a$ .** During SCAR-B the UW C-131A aircraft obtained measurements in 20 individual smoke plumes, in which 27 measurements were obtained with the OEC, 27 with the IP, 13 with the OR, and 20 with the TE. The average quartz filter loading for smoke samples was 550  $\mu\text{g}$  (over 5 times the necessary amount). The derived values of  $\sigma_a$  ranged from  $8 \times 10^{-5}$  to  $5 \times 10^{-3} \text{ m}^{-1}$ . Coarse particles (mostly soils) accounted for less than 15% of the aerosol volume and less than 5% of the aerosol extinction [Reid and Hobbs, this issue]. Therefore their presence does not substantially effect the results.

Figure 1 shows a comparison of all of the measurements of  $\sigma_a$  derived from the IP, OR, and TE techniques compared to the corresponding OEC measurements of  $\sigma_a$ . The IP and OR methods produced data that scatter around the 1:1 line with the OEC; the mean residual from the 1:1 line for the IP and OR techniques were -3% and -0.5%, respectively. Excluding the largest  $\sigma_a$  value (because of its large leverage on the distribution), the linear correlation coefficients ( $r$ ) of the OR and IP with the OEC were 0.9 and 0.7, respectively. The standard deviations of the residuals from the IP and OR techniques (i.e., the uncertainties in the methods) were  $\pm 40\%$  and  $\pm 17\%$ , respectively.

An investigation was conducted to determine why the IP method had such a large uncertainty. It was found that filters with higher loadings had considerably fewer errors associated with them. For example, filters with aerial densities greater than 20  $\mu\text{g cm}^{-2}$  had an uncertainty of  $\pm 30\%$  (10 filters total). Furthermore, if we excluded one outlying data point, the uncertainty improved to  $\pm 20\%$ . Thus increased filter loading had a significant effect in reducing the uncertainty in the IP method.

It was found also that there was a minor systematic difference between the OEC and the OR for samples with low  $s_a$  values. In these cases the OR underestimated  $\sigma_a$  by as much as 15%. This difference could arise from many sources, including differences between the wavelength used in the OEC and the OR (green versus broadband) or a minor systematic error in the

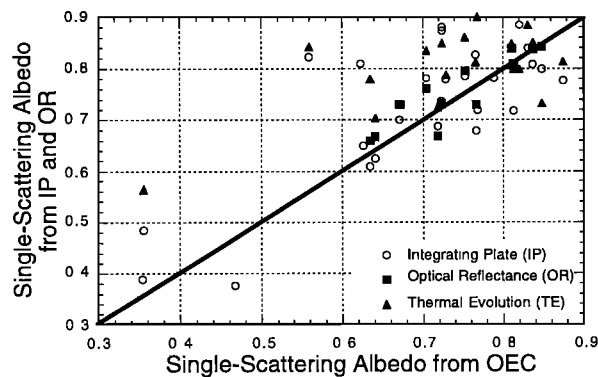
calibration curve for the OR. Furthermore, because there were so few OR samples, we cannot rule out chance.

The TE method performed less well than the IP and OR methods, with most of the data points lying well below the 1:1 line with the OEC ( $r=0.45$ ). The mean residual from the OEC 1:1 line was -49%, indicating a substantial systematic error. The standard deviation of the residual for the TE was  $\pm 31\%$ , which is better than the IP but worse than the OR method.

While the errors in the various methods we used for determining  $\sigma_a$  are mainly random, the TE method has major systematic errors. These errors can come from two principle sources: (1) errors in the determination of the BC content of the aerosol and (2) variations in the appropriate value of  $\alpha_{abc}$  for smoke aerosols from different sources, with different ages, etc. The method of Cachier *et al.* [1989] has been employed with much success. As discussed in section 3.2, Cachier *et al.* performed sensitivity tests and attempted to compensate for artifacts (e.g., precombustion of BC, possible artifacts from soil carbonates and decarbonation procedures, etc.). However, several variations of the TE method are in use, and in cases where there is a low BC to OC ratio (such as biomass burning aerosol), these methods can produce systematic differences in the derived BC (and hence  $\sigma_a$ ) which are in excess of 50% [Shah and Rau, 1991]. Therefore while the Cachier *et al.* study is reassuring as to the reproducibility of the BC data used in this study, there is uncertainty because of possible systematic errors in the BC data due to the specific method used. For example, there is a possibility that in the thermal analysis the BC was combusted during the thermal pretreatment of the sample, because of catalytic reactions with alkali Earth materials [Novakov and Corrigan, 1995]. This would produce an underestimate of BC from the TE method.

Compounding possible systematic errors in the measurement of BC is the uncertainty in the value of  $\alpha_{abc}$ . As discussed in section 1, the mass absorption efficiency of BC is highly dependent on the chemical mixing and size of the particles. In this paper we employed a value of 15  $\text{m}^2 \text{g}^{-1}$  for  $\alpha_{abc}$ . However, from the analysis by Martins *et al.* [this issue (a)] on the absorption properties of smoke particles in SCAR-B, the value of  $\alpha_{abc}$  could be as high as 20  $\text{m}^2 \text{g}^{-1}$ . If we assume that all of the systematic differences between the OEC and the TE methods were due to an underestimate of the value of  $\alpha_{abc}$ , then  $\alpha_{abc}$  for BC must be on the order of 25  $\text{m}^2 \text{g}^{-1}$ . However, this would be an unusually high value for  $\alpha_{abc}$  for a viable aerosol size distribution. Therefore it is likely that the systematic difference between the OEC and the TE methods is due to a combination of measurement error of the BC content of the sample and an underestimation of  $\alpha_{abc}$ .

**4.1.2. Determination of  $\omega_o$ .** Combination of the derived values of  $\sigma_a$  with simultaneous measurements of  $\sigma_s$  from the nephelometer aboard the aircraft yields  $\omega_o$ . The OEC produced  $\omega_o$  values for smoke particles that varied from 0.35 (for vigorous flaming grass fires) to 0.9 (for smoldering slash). As discussed in section 2,  $\omega_o$  is more sensitive to errors in  $\sigma_a$  for lower  $\omega_o$  values. Although the values of  $\sigma_a$  derived from the OR technique are well correlated with the OEC measurements, even the comparatively good 20% variation is enough to significantly affect the accuracy of the derived values of  $\omega_o$  (Figure 2). Compared to the OEC, the OR technique performed best, with a mean residual of 0.01 and a standard deviation from the 1:1 line of  $\pm 0.04$ . The IP technique also yielded values for  $\omega_o$  which were not



**Figure 2.** Comparison of measurements of concentrations of aerosol black carbon ( $c_{\text{mbc}}$ ) in smoke plumes derived from the integrating plate (IP) and the optical reflectance (ORT) techniques versus the thermal evolution (TE) method. The line shows the 1:1 ideal correlation.

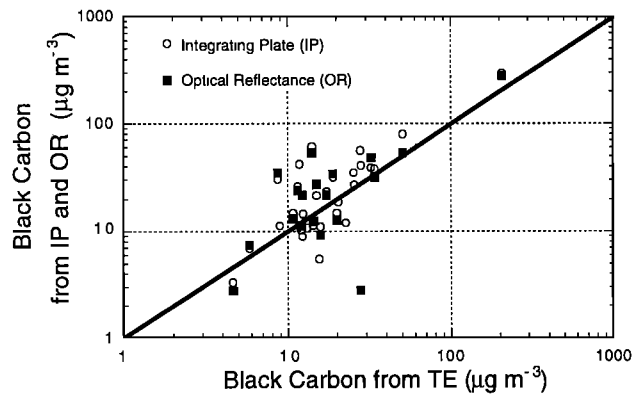
systematically different from the OEC (mean residual of  $<0.01$ ). However, for  $\omega_0$  values between 0.6 and 0.9 the IP technique showed considerable uncertainty, with a standard deviation of  $\pm 0.075$  from the 1:1 line with the OEC.

The large discrepancy between the TE method and the OEC method in the determination of  $\sigma_a$  leads to a large discrepancy in the value of  $\omega_0$ . Using a value of  $15 \text{ m}^2 \text{ g}^{-1}$  for  $\alpha_{\text{abc}}$ , the TE technique overpredicted the value of  $\omega_0$  by about 0.07. The standard deviation of the data points from the regression line is  $\pm 0.10$ , which indicates substantial random variability.

It is interesting to note that while the TE technique had less random uncertainty than the IP technique in the determination of  $\sigma_a$ , in the determination of  $\omega_0$  the TE technique had a higher uncertainty than the IP. This is simply a result of the data weighting. Errors in the determination of  $\sigma_a$  by the IP technique happened to be larger for samples with higher values of  $\omega_0$ . Thus these errors have a diminished influence on the derived value of  $\omega_0$ . From our data we cannot determine if this is by chance or is due to some systematic error for which we have not accounted.

**4.1.3. Determination of black carbon.** Black carbon concentrations ( $c_{\text{mbc}}$ ) are frequently inferred from optical methods, such as the IP and OR techniques, using equation (1). The validity of this approach was tested by comparing  $c_{\text{mbc}}$  values derived from the optical methods with those derived from the TE technique. Since the TE had significant systematic and random errors in comparison to the OEC, we would expect that methods such as the IP and OR would perform poorly in predicting the BC content of the aerosol. This is demonstrated in Figure 3, where the BC content of the aerosol, derived by the IP and OR techniques (from equation (1)), is plotted against the BC derived from the TE technique. Here we have again assumed a value of  $\alpha_{\text{abc}}$  of  $15 \text{ m}^2 \text{ g}^{-1}$ .

From Figure 3 we see a 35% and 40% systematic overestimate of BC from the IP and OR techniques, respectively. This is expected, since the TE consistently underestimated  $\sigma_a$  (see section 4.1.1). The standard deviation in the residuals for the IP and OR techniques were 93 and 100%, respectively. However, these large standard deviations were heavily influenced by two data points. If these data points are removed from the analysis, the standard deviation of the residuals from the IP and OR methods reduces to 65% and 55%, respectively. Thus allowing for systematic errors in the



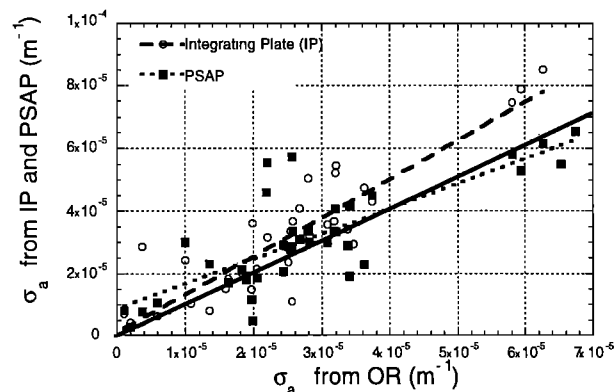
**Figure 3.** Comparison of the single-scattering albedo ( $\omega_0$ ) for smoke plumes derived from measurements with the integrating plate (IP), optical reflectance (OR), and thermal evolution (TE) techniques versus those derived from the optical extinction cell (OEC). The line shows the 1:1 ideal correlation.

measurement of  $\sigma_a$  and the BC,  $c_{\text{mbc}}$  can be estimated from  $\sigma_a$  to, at best, a 70% uncertainty.

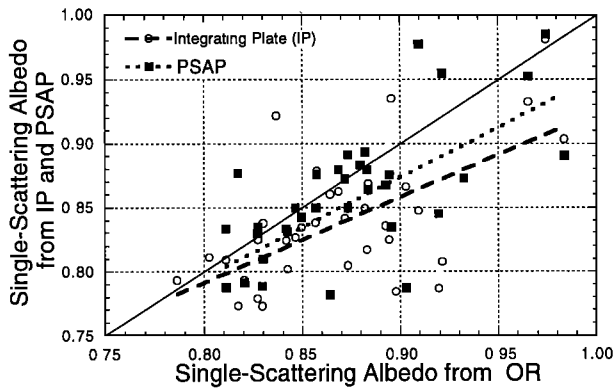
#### 4.2. Regional Hazes

Fifty regional haze samples, dominated by smoke, were collected aboard the UW C-131A aircraft in SCAR-B. In the regional hazes,  $\sigma_a$  was of the order of  $10^{-5} \text{ m}^{-1}$ . Because the OEC requires high particle concentrations and concentration gradients, it could not be used to measure  $\sigma_a$  for these samples. Also, the TE technique was not used because of inadequate amounts of aerosol mass collected on the filters. Since the OR technique compared best with the OEC for the individual plume samples (see section 4.1), it was used as a “secondary standard” in comparing the regional haze samples.

Values of  $\sigma_a$  derived from the IP and PSAP techniques are compared to those from the OR technique in Figure 4. As shown in the case of the smoke plume samples, the optical methods agree fairly well, with  $r$  values of 0.89 and 0.79 for the IP-OR and PSAP-OR comparisons, respectively. In view of the 20% variance in the OR-OEC comparisons discussed in section 4.1, roughly half of this variability could be due to the use of the OR technique as a secondary standard.



**Figure 4.** Comparison of measurements of the absorption coefficient of aerosol ( $\sigma_a$ ) in regional hazes measured by the integrating plate (IP) and the particle/soot absorption photometer (PSAP) techniques versus the optical reflection (OR) technique. The solid line shows the 1:1 ideal correlation.



**Figure 5.** Comparison of measurements of the single-scattering albedo ( $\omega_0$ ) in regional hazes derived from absorption measurements with the integrating plate (IP) and the particle/soot absorption photometer (PSAP) techniques versus the optical reflection (OR) technique. The solid line shows the 1:1 ideal correlation.

There are only slight systematic differences between the methods, with slopes of  $1.2 \pm 0.10$  and  $0.8 \pm 0.1$  for the IP and PSAP techniques, respectively. These differences can arise from a variety of sources, including chance, calibration factors, and the wavelength used in each technique. If one considers the uncertainty in the OR method, these regressions are not statistically different from the 1:1 line. Despite the good  $r$  values for the regression, there is still a fair amount of scatter around the regression line. The mean residuals from the 1:1 line were 28% and 14% for the IP and PSAP, respectively. Standard deviations of the residuals were  $\pm 45\%$  and  $\pm 60\%$  for the IP and PSAP techniques, respectively.

The derived values of  $\omega_0$  for the regional hazes varied from 0.75 to 0.95. Comparisons of values of  $\omega_0$  derived from the IP and PSAP techniques with those derived from the OR technique are shown in Figure 5. The optical methods compare only marginally well with each other, with  $r$  values of 0.64 and 0.58 for the IP-OR and PSAP-OR regressions, respectively. This results in a standard deviation of the residuals of  $\omega_0$  from the regression lines of roughly  $\pm 0.06$  and  $\pm 0.05$  for the IP and OR techniques. This uncertainty places constraints on the statistical significance of the individual absorption measurements. Recall that the change over between a warming and a cooling effect of an aerosol layer in the atmosphere occurs at a value of  $\omega_0$  roughly 0.85, and almost all of our  $\omega_0$  measurements lie between 0.8 and 0.9, with a best precision of  $\pm 0.04$  for the OR technique. To determine even the correct

sign of the direct radiative forcing due to biomass burning aerosol, the uncertainty in  $\omega_0$  must be  $\pm 0.02$  or less. In SCAR-B this accuracy was achieved by using four or more filters per data point.

**4.3. Comparison of in Situ and Ground-Based Remote Sensing Measurements**

On several occasions in SCAR-B the UW C-131 aircraft flew vertical profiles over ground-based Sun photometers and irradiance sensors [Ross *et al.*, this issue]. On three of these occasions, the ground-based remote sensing techniques were used to retrieve aerosol single-scattering albedo. However, on two of these flights (August 30 and September 7), clouds were present during the profile over the ground instruments. Therefore cloudless time periods that occurred within approximately 1.5 hours of the flights were utilized to retrieve the aerosol single-scattering albedo from the combined Sun photometer/irradiance data.

In the vertical profiles the number of flight levels at which intensive measurements were obtained varied from 5 to 10. Average columnar single-scattering albedos were derived from the OR, IP, and PSAP techniques using a weighting function based on the fraction of the total column optical depth apportioned to each flight level. A hygroscopic growth factor was applied to the in situ measurements to allow for the effect of the ambient relative humidity (RH) on the growth of the aerosol (Kotchenruther and Hobbs [this issue] showed that aerosols in regional hazes dominated by biomass smoke in Brazil have a relatively low hygroscopic growth factor, namely, 1.2 at RH=80%). A  $c_p$  value of 0.5 (from the IP-OEC regression line for the in-plume samples) was employed for the IP technique. The values for the single-scattering albedo obtained in this way were compared to those derived from the Sun photometer and irradiance data using the SPCTRAL2 and 6S models. To test how well the remote sensing techniques perform on their own,  $\omega_0$  values were derived without the benefit of data from the UW aircraft. Inputs for the 6S model included a remote-sensing-derived lognormal volume size distribution for smoke from Remer *et al.* [1996] and an assumed value for the real index of refraction of the aerosol of 1.53 [Lenoble, 1991; Anderson *et al.*, 1996]. The results of these comparisons are listed in Table 1.

The column-averaged values of the single-scattering albedo ( $\omega_0$ ) for the three in situ optical methods agree fairly well. With the exception of one data point from the OR technique over the Pantanal region of Brazil, the in situ methods yield values of  $\omega_0$  within  $\pm 0.02$  of each other (the outlying data point from the OR technique can be partially explained by the

**Table 1.** Column-Averaged Values for Single-Scattering Albedos in Regional Hazes Obtained From Airborne in Situ Measurements Using the Optical Reflection (OR), Integrating Plate (IP), and Particle Soot/Absorption Photometer (PSAP) Techniques Compared to Values Derived From Sun Photometer and PAR Irradiance Measurements Using Two Radiative Transfer Models

Brazil	1995	In Situ Optical Measurements			Sun Photometer Measurements	
		OR	IP	PSAP	SPCTRAL2 Model	6S Model
Cuiabá	August 27	N/A	0.83	0.82	0.835	0.83
The Pantanal	August 30	0.92	0.85	0.87	0.875	0.92
Near Porto Velho	September 7	0.88	0.86	0.89	0.89	0.94



fact that only three low-volume filters were taken, compared to nine for the IP technique and continuous measurements from the PSAP). This agreement is likely a result of the large sample sizes. On average, four OR, five IP, and continuous PSAP samples were obtained in each vertical profile. Thus while the scatter in  $\omega_o$  values between individual flight levels is approximately  $\pm 0.05$ , the final column-average values of  $\omega_o$ , obtained from the three optical techniques, generally converge.

The agreement between the three in situ optical techniques and the SPCTRAL2 model is excellent, although the model predicts slightly greater  $\omega_o$  values for some cases (by +0.01 to +0.02) than was derived from the OR, IP, and PSAP techniques. Differences of this magnitude can be explained, in part, by a lack of exact time coincidence between the aircraft flights and the cloudless time periods used for the remote sensing retrievals on August 30 and September 7. Also, if thin cirrus or nonabsorbing aerosols were present above the haze layer they may have contributed to higher estimates of total column  $\omega_o$ . Regardless, these differences are within the expected accuracy of all the  $\omega_o$  retrieval techniques.

The 6S model overestimated  $\omega_o$  compared to the three in situ optical techniques in two of the three cases studied. This is partly due to the inputs to the 6S model. For example, while a real index of refraction of 1.53 (the value assumed for the retrieval) is typical for most inorganic aerosols, the value for biomass burning aerosols is not known. A sensitivity test was performed by varying the real component of the index of refraction from 1.53 to 1.4 (typical for many organic species plus accounting for water uptake by aerosols). This change decreased the derived value of  $\omega_o$  by 0.015. There were also significant differences in the measured aerosol size distributions on the various overflights. For example, the volume median diameter of the aerosol over Cuiabá was 0.23  $\mu\text{m}$  [Reid *et al.*, this issue], while over the Pantanal and near Porto Velho, it was 0.29  $\mu\text{m}$ . The smoke size distribution models of Remer *et al.* [1997], which we utilized, have volume median diameters of 0.264 and 0.34  $\mu\text{m}$  for Cuiabá and the Pantanal/Porto Velho, respectively. This difference can account for a further 0.015. Finally, there are differences in the aerosol optical depth ( $\tau$ ) between the sites. At Cuiabá,  $\tau$  at 500 nm was 0.9, while over the Pantanal and Porto Velho,  $\tau$  was 2.1. Both irradiance models (6S and SPCTRAL2) may have larger uncertainties at very high  $\tau$  values due to approximations in the radiative transfer code. Thus the combination of inputs to the 6S model and the high optical depth could account for the differences in the derived values of the single-scattering albedo.

## 5. Discussion and Conclusions

The results of this intercomparison study are encouraging. While the uncertainty in individual samples may be large, with an adequate sample size, all of the optical methods yielded a value of  $\sigma_a$  that converged to within  $\pm 20\%$  of the same value. Because there are no "absolute" standards for measuring aerosol absorption or BC concentrations, one cannot say which method is "correct," only that the methods discussed in this paper yielded similar results. For example, for each region (Cuiabá, Porto Velho, and Marabá) the IP, OR, and PSAP techniques gave mean values for  $\omega_o$  that agreed to within  $\pm 0.02$ . The slight differences in values are probably due to the systematic errors in the calibration, differences in the

wavelength at which the measurements were made (green or broadband), and random errors.

The OR method compared best to the OEC for in-plume samples, with a variance in the derived value of  $\sigma_a$  of less than 17%. Since the OEC measures  $\sigma_a$  to better than 10%, it is likely that the variance in  $\sigma_a$  from the OR method is better than 14%. This, in turn, translates into an error in the derived single-scattering albedo of less than  $\pm 0.03$ . Although we do not have a primary standard for the measurements obtained in the regional hazes, it is likely that uncertainties are similar for this environment. However, for low values of  $\sigma_a$  the OR technique systematically underestimated absorption compared to the OEC by an average of 15%. This could be due to minor errors in the calibration of the OR technique. The OR technique yielded the highest values of  $\omega_o$  from the optical techniques.

The IP technique was used most frequently in SCAR-B. Random uncertainties in  $\sigma_a$  values from individual samples vary with filter loading. Filters with aerial densities  $>20 \mu\text{g m}^{-2}$  yield values of  $\sigma_a$  with uncertainties  $< 30\%$ . However, overall, the uncertainty in individual samples increases to over 40%. This translates into uncertainties of  $\pm 0.03$  to  $\pm 0.1$  in  $\omega_o$ .

Although there was no primary standard available to evaluate measurements from the PSAP, comparisons with the OR method in the regional haze validate the PSAP data. There was a variance of 45% between the PSAP and the OR technique. Thus the 17% uncertainty in the OR method implies an uncertainty in the PSAP of approximately 40%. This translates into an uncertainty in  $\omega_o$  of  $\pm 0.05$  for a 1 min sample.

Application of the SPCTRAL2 radiative transfer model to Sun photometer and irradiance data yielded single-scattering albedos slightly higher (by 0.01) than the columnar-averaged values derived from in situ optical measurements made aboard the UW aircraft. The difference could be due to the lack of exact time coincidence between the flights and the cloudless periods utilized for the remote sensing retrievals. However, this small difference is within the expected accuracy of  $\omega_o$  retrievals for all techniques.

The 6S radiative transfer model showed varied results. While it agreed with the other methods for one vertical profile, in two other profiles it gave values for the single-scattering albedo which were well above the mean. This is likely due to the extremely high optical depth of the haze ( $\tau_{500}=2.1$ ), the assumed particle size distribution, and the assumed real index of refraction of the particles. Reid *et al.* [this issue] report significant variability in the size distribution of particles in the regional hazes. This variability, coupled with the uncertainty in the value of the real index of refraction, appears to have a significant effect on the values of  $\omega_o$  derived from the 6S model.

This intercomparison study demonstrates the difficulty in deriving reliable values of  $\sigma_a$  from measurements of the BC content using the TE method (and vice versa). It is clear that for the biomass burning aerosols studied here, variability in data from the TE method and the use of equation (1) to relate  $\sigma_a$  to  $c_{\text{mbc}}$  can lead to large systematic errors. If these errors can be overcome, the uncertainty in  $\sigma_a$  from the TE would be about 30%. In this study, the errors in the derived single-scattering albedo were in excess of  $\pm 0.1$  for all samples.

It is possible that most of the systematic error in the TE method is from our underestimation of  $\alpha_{\text{abc}}$ . However, this

leads to an unphysically high mass absorption efficiency for the biomass smoke studied here of approximately  $25 \text{ m}^2 \text{ g}^{-1}$ . The TE method used in SCAR-B probably underestimated the amount of black carbon. Huffman [1996] suggested that BC can evolve at temperatures below the threshold that most investigators use to separate BC from OC. Novakov and Corrigan [1995] reported that catalytic reactions with K and Na (both of which are present in large quantities in biomass burning aerosols: Ward et al. [1992], Gaudichet et al. [1995], and Ferek et al. [this issue]) can reduce the evolution temperature of BC by  $100^\circ\text{C}$ . Values of  $c_{\text{mbc}}$  obtained from the TE method should probably be increased by 25% for the SCAR-B data set. This increase in  $c_{\text{mbc}}$ , coupled with a mass scattering efficiency of  $\sim 18 \text{ m}^2 \text{ g}^{-1}$ , would yield systematic agreement between the TE method and the OEC.

The results reported here have some important implications for previous research on biomass burning in Brazil. Artaxo et al. [1994] used the same OR method that we used to derive 4 years of BC data in smoky regions in Brazil. Using their values of  $c_{\text{mbc}}$ , highly accurate values of  $\sigma_a$  can be derived using an effective mass absorption efficiency ( $\alpha_{\text{abc}}$ ) of  $6.8 \text{ m}^2 \text{ g}^{-1}$ . However, Artaxo et al.'s values for  $c_{\text{mbc}}$  do not correlate very well with the BC concentration of the atmosphere as measured by TE. To retrieve the true black carbon content, one would have to correct the data using the actual value of  $\alpha_{\text{abc}}$  for smoke particles from biomass burning as well as account for the effects of other absorbing aerosols (e.g., industrial pollution, soils, and absorbing organics). Furthermore, as discussed in this paper, the value of  $\alpha_{\text{abc}}$  for biomass burning aerosol is highly variable. Thus it is unlikely that the true BC content of individual aerosol samples can be retrieved. Because the true value for  $\alpha_{\text{abc}}$  for biomass burning aerosol is of the order of  $15$  to  $20 \text{ m}^2 \text{ g}^{-1}$ , and Artaxo et al. assumed a value for  $\alpha_{\text{abc}}$  of  $6.8 \text{ m}^2 \text{ g}^{-1}$ , it is likely that Artaxo et al.'s BC values are high by almost a factor of 2. However, considering the ensemble of data, one may make a zeroth-order correction and retrieve the average BC concentration as a percentage of total mass by multiplying Artaxo et al.'s values by  $0.55 \pm 0.2$ .

Hobbs et al. [1997] presented mean values for  $\omega_0$  derived from the IP method for the regional hazes studied in SCAR-B. They used a value for  $c_p$  of 0.5, and no shadowing correction was applied ( $R=1$ ). Thus their derived values for  $\omega_0$  for each region were slightly higher (although not statistically different) from those derived here from the OR and PSAP techniques.

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