

# Aerosol optical thickness from Brewer spectrophotometers and an investigation into the stray-light effect

Abel A. Silva and Volker W. J. H. Kirchhoff

The Langley method has been applied to the measurements of direct solar radiation made by Brewer spectrophotometers to obtain the aerosol optical thickness (AOT) of the atmosphere in the ultraviolet-B range. In several cases the AOT increased with wavelength, which raises suspicion about the stray-light effect. To investigate the quality of the AOT measurements and the possibility of stray light, we conducted a campaign by using single- and double-monochromator Brewers. The campaign's results have shown that both Brewers' AOT values are in good agreement and that stray light is not an important effect for AOT at wavelengths above 306 nm. © 2004 Optical Society of America

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## 1. Introduction

Brewer spectrophotometers are capable of making direct sun (ds) measurements looking straight into the solar beam. Total column values of ozone ( $O_3$ ) and sulfur dioxide ( $SO_2$ ) in the atmosphere are determined from those measurements. In addition, they have also been used to obtain the aerosol optical thickness (AOT)<sup>1,2</sup> in the ultraviolet B (UVB) by application of the Langley method to the wavelengths of 306.3, 310.1, 313.5, 316.8, and 320.1 nm.

Ozone Laboratory at the Instituto Nacional de Pesquisas Espaciais (INPE) in Brazil has used Brewer spectrophotometers in special campaigns to make ds measurements for obtaining AOT values in Brazil and other sites in South America. Site location has varied from the Brazilian seashore to the high altitudes of the Andes Mountains in Bolivia, passing through biomass burning, quiet rural, and polluted metropolitan regions in Brazil. The procedure for obtaining the AOT from Brewer ds measurements has been checked to guarantee the highest-quality values. Nevertheless, in some cases<sup>1,2</sup> the AOT values have shown a puzzling behavior by increasing

with wavelength. According to Angstrom's law, the opposite is expected; could the stray-light effect in the single monochromator produce erroneous results for AOT? The stray-light effect is an augmentation of the detector's photon count due to the arrival of undesirable photons of different wavelengths in the photomultiplier tube. For shorter wavelengths in the UVB range, such contamination can be the same order of magnitude as the real photon count, increasing UVB measurements as much as 10%. It suggests the misleading idea of a less opaque atmosphere. The main source of stray light in a spectrophotometer is its diffracting grating, followed by collimating and focusing mirrors. Marengo *et al.*,<sup>3</sup> with ds measurements from a double-monochromator Brewer, got results similar to Kirchhoff *et al.*<sup>1,2</sup> without using the Langley method and found, besides a wavelength increasing behavior, a strong oscillation of the AOT values below approximately 310 nm, which Marengo *et al.* believed to be caused by an imperfect removal of the ozone contribution or even a sulfur dioxide influence. Bais *et al.*<sup>4</sup> also compared UVB irradiances from single- and double-monochromator Brewers and concluded that for wavelengths greater than approximately 300 nm there was no significant effect of stray light.<sup>5,6</sup>

According to Bernhard and Seckmeyer,<sup>7</sup> in addition to stray light there are other effects also intrinsic to the spectrophotometer that can cause systematic errors affecting the measurements of UVB irradiances and their uncertainties. Those effects arise from radiometric calibration, cosine error, spectral resolution, wavelength misalignment, radiometric stability, nonlinearity between the instrument's sig-

A. A. Silva (abel@ieav.cta.br) is with the Centro Técnico Aeroespacial Rod. dos Tamoios, km 5,5, São José dos Campos, SP 12228-840, Brazil. V. W. J. H. Kirchhoff is with the Instituto Nacional de Pesquisas Espaciais, Av. Dos Astronautas 1758, São José dos Campos, SP 12227-010, Brazil.

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nal and the flux of photons, signal-to-noise ratio, and solar position timing errors. Stray light is the eighth in decreasing order of importance as a source of error among the above cited. Associated with spectral resolution and timing error effects, the cone form of the spectrophotometer field of view in ds measurements allows for undesirable forward-scattering photons that get inside the detector together with the other photons from the solar beam not scattered by the atmosphere. In other words, the Beer's law assumption [see Eq. (1)] of a direct solar beam of radiation at a wavelength  $\lambda$  only<sup>8-10</sup> is not strictly followed.

In this paper we compare the AOT in the UVB range from simultaneous ds measurements made by two Brewers to investigate whether the stray-light effect acts on measurements. Measurements of ds are likely to be analyzed by the Langley method after having been converted from raw counts into count rates following exactly the same Brewer algorithm's routine.<sup>11</sup> In 2002 a special campaign was conducted in São José dos Campos (23.18° S, 45.89° W, 600 m above sea level) by use of ds measurements from single- and double-monochromator Brewers in the first part of the campaign, and two single-monochromator Brewers in the second part. For both parts, Brewers had worked side by side under the same schedule. Because double monochromators can reject stray light much more efficiently than single ones, it is believed that any measurable difference between AOT values from ds measurements from single- and double-monochromator Brewers could be detected, if stray-light effects are important in these proceedings.

## 2. Instruments and Methods

During four weeks in 2002, two Brewer spectrophotometers were set simultaneously side by side in the Ozone Lab facilities at INPE and operated together under the same schedule to make ds measurements at the wavelengths 306.3, 310.1, 313.5, 316.8, and 320.1 nm with spectral resolution of 0.6 nm FWHM. In the first part of the campaign, two Brewers, a double-monochromator MKIII (#167) and a single-monochromator MKIV (#068), made several ds measurements during the mornings and afternoons. Brewer checkout routines were conducted periodically to ensure the best quality of measurements. In the second part of the campaign the same Brewer MKIV of the first part and a single-monochromator Brewer MKII (#056) were used. Unblocked solar beam conditions for ds measurements were checked by visual surveillance of the sky. Despite this procedure, it is impossible to guarantee no influence on sky-thickness values from invisible high-altitude clouds or variable plumes of aerosols during measurements.

According to Beer's law, the solar beam intensity ( $I_\lambda$ ) at the wavelength  $\lambda$  depends exponentially on the atmospheric optical thickness ( $\tau_{at}$ ):

$$I_\lambda = I_{\lambda_0} \exp(-m\tau_{at}). \quad (1)$$

$I_{\lambda_0}$  is the intensity of radiation at the Earth's atmospheric top, and  $m$  is the air mass that equals the inverse of the cosine of the solar zenith angle for angles less than 60°. Rewriting Eq. (1), we obtain

$$\ln(I_\lambda) = \ln(I_{\lambda_0}) - \tau_{at}m, \quad (2)$$

where  $I_\lambda$  is a Brewer ds measurement at air mass  $m$  from a set of data ( $m, I_\lambda$ ), which can be analyzed by the Langley method to obtain  $\tau_{at}$  if both  $I_{\lambda_0}$  and  $\tau_{at}$  stay constant for some period of time  $\Delta t$ . That means  $\ln(I_\lambda)$  is proportional to  $m$ .<sup>12</sup>

In the UVB band the AOT is equal to  $\tau_{at}$  minus the sum of the optical thicknesses of all other atmospheric components that attenuate the solar beam intensity along its path in the atmosphere except for aerosols:

$$\tau_{ae} = \tau_{at} - (\tau_{O_3} + \tau_{SO_2} + \tau_R). \quad (3)$$

$\tau_{O_3}$ ,  $\tau_{SO_2}$ , and  $\tau_R$  are the optical thicknesses due to  $O_3$  and  $SO_2$  absorption and the Rayleigh scattering effect, respectively.  $\tau_R$  can be easily calculated with the Hansen and Travis formula<sup>13,14</sup> corrected for local atmospheric pressure that was  $(960 \pm 10)$  mbar for both parts of the campaign.  $\tau_{O_3}$  and  $\tau_{SO_2}$  are a function of the gas cross section<sup>15,16</sup> ( $\sigma$ ) and the total column gas value ( $N$ ) supplied by Brewer ds measurements and come from

$$\tau_i = \sigma N 2.69 \times 10^{16}, \quad i = O_3 \text{ or } SO_2. \quad (4)$$

Contributions of trace gases such as formaldehyde (HCHO), nitrous acid (HONO), and others were not taken into account because  $\tau_{ae}$  is not significantly affected by the poor concentration of those trace gases in the atmosphere as Kirchhoff *et al.*<sup>2</sup> demonstrated.

The uncertainty of  $\tau_{ae}$  ( $\delta_{ae}$ ) must reflect the parameters and processes used in its calculation. That means the error propagation analysis must be applied to Eqs. (3) and (4) and the Hansen and Travis formula.  $\tau_R$  uncertainty is supposed to depend on the local atmospheric pressure only and is equal to approximately 1%.  $\tau_{O_3}$  and  $\tau_{SO_2}$  uncertainties come from the following formula:

$$\delta_i = 2.69 \times 10^{16} [(N\delta_\sigma)^2 + (\sigma\delta_N)^2]^{0.5}, \quad i = O_3 \text{ or } SO_2. \quad (5)$$

The uncertainties  $\delta_\sigma$  and  $\delta_N$  (related to  $\sigma$  and  $N$ ) for  $O_3$  are less than 1% and 2%, respectively, whereas for  $SO_2$  they can be much larger. Nevertheless, owing to its low concentration in the atmosphere at the scenarios of this paper, the  $SO_2$  optical thickness and its uncertainty have small, or even negligible, influence on  $\tau_{ae}$ . The uncertainty of  $\tau_{at}$  is drawn from the linear regression analysis used to calculate the Langley method's parameters, and it is usually the largest source of error for  $\tau_{ae}$ . Then the uncertainty of  $\tau_{ae}$  is equal to

$$\delta_{ae} = [(\delta_{at})^2 + (\delta_{O_3})^2 + (\delta_{SO_2})^2 + (\delta_R)^2]^{0.5}. \quad (6)$$

**Table 1. Report of ds Measurements Made by Brewers MKIII and MKIV in the First Part of the Campaign**

Day of Year	$\Delta t$ (hr)	$\Delta m$	Number of ds Measures	Brewer MKIII $R^2$	Brewer MKIV $R^2$	$D$ (%)
60	0.33	0.093	5	0.90	0.88	16.1
63	0.97	0.155	14	0.92	0.92	1.1
64	1.75	0.549	23	0.96	0.95	7.7
66	0.76	0.219	11	0.91	0.89	9.2
69	2.00	0.481	25	0.93	0.92	11.0
72	1.41	0.215	17	0.86	0.88	5.6
75	2.41	0.295	20	0.93	0.92	3.6
76	1.80	0.304	21	0.92	0.94	9.4

**3. Results and Discussion**

The campaign was organized to obtain ds measurements for the application of the Langley method during periods of time  $\Delta t$  (or intervals  $\Delta m$  of  $m$ ) in the mornings or afternoons. Table 1 shows  $\Delta t$  and the corresponding number of  $n$  ds measurements released in the first part of the campaign by Brewers MKIII and MKIV. Table 2 shows the same for the second part of the campaign by use of Brewers MKII and MKIV. Time periods as short as 0.33 h and as large as 4.11 h, with a proportional number  $n$  of ds measurements, were obtained.

The optical thicknesses for  $O_3$  and  $SO_2$  absorption come from the representative values of  $O_3$  and  $SO_2$  for the period of time of  $n$  ds measurements. Tables 1 and 2 show  $n$  as the number of ds measures. The representative values of  $O_3$  and  $SO_2$  are averages among  $n$  ds measurements with one standard deviation (sd) taken as uncertainty. Figures 1 and 2 show those values in the two parts of the campaign. Even though there is some disagreement on days of year 60, 282, 302, and 307, such disagreement neither significantly affects the corresponding AOT values nor changes their wavelength behavior. Typical values for the optical thickness of aerosols,  $O_3$ ,  $SO_2$ , Rayleigh scattering, and the atmosphere obtained in the campaign can be seen in Table 3.

At the end of the campaign, 16 groups of  $n$  ds measurements were obtained (15 in the mornings and only one in the afternoons). After applying the

**Table 2. Report of ds Measurements Made by Brewers MKII and MKIV in the Second Part of the Campaign**

Day of Year	$\Delta t$ (hr)	$\Delta m$	Number of ds Measures	Brewer MKII $R^2$	Brewer MKIV $R^2$	$D$ (%)
282	1.08	0.081	7	0.92	0.95	15.5
283	4.11	0.912	34	0.92	0.95	13.1
284	3.08	0.629	14	0.85	0.89	15.7
285	4.10	0.893	30	0.99	0.99	5.0
286	4.04	0.885	31	0.96	0.97	4.5
289	2.16	0.396	20	0.95	0.95	11.1
302	2.33	0.424	21	0.97	0.97	5.6
307	3.41	0.504	29	0.94	0.97	6.1

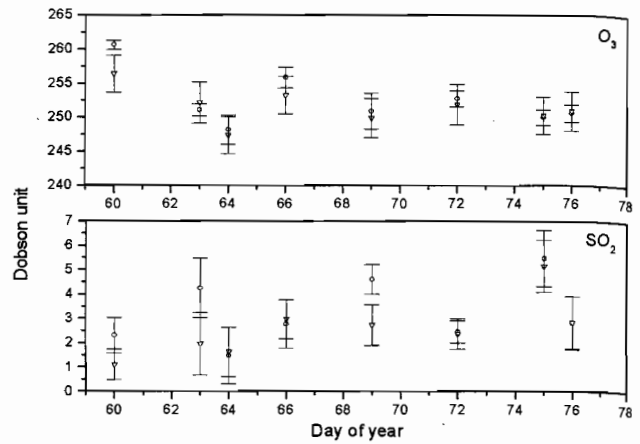


Fig. 1. Representative  $O_3$  and  $SO_2$  values from Brewers MKIV (circles) and MKIII (triangles) in the first part of the campaign.

Langley method to those groups and calculating  $\tau_{O_3}$ ,  $\tau_{SO_2}$ , and  $\tau_R$  values, we used Eqs. (3) and (6) to obtain the corresponding AOT values and their uncertainties, respectively. Figure 3 shows the AOT values for the first part of the campaign, in which they ranged from  $0.35 \pm 0.05$  to  $1.91 \pm 0.18$  at 306.3 nm, with Brewer MKIV and Brewer MKIII values as circles and triangles, respectively. The day of year is shown in the corner of the figures, and uncertainties are 1 sd. Figure 4 shows the AOT values for the second part of the campaign with Brewer MKIV (circles) and Brewer MKII (triangles). In this case the AOT values range from  $0.20 \pm 0.18$  to  $2.63 \pm 0.39$  at 306.3 nm and are larger than the ones in the first part of the campaign. That is because October, in which almost all the second part of the campaign was carried out, marks the end of the dry season, which is typically more polluted than the rainy season of March in the first part of the campaign.

AOT uncertainties have been chosen as 1 sd to emphasize the agreement between the Brewers. As can be seen from a typical value of AOT in Table 3, although  $\delta_{ae}$  is wavelength decreasing by approxi-

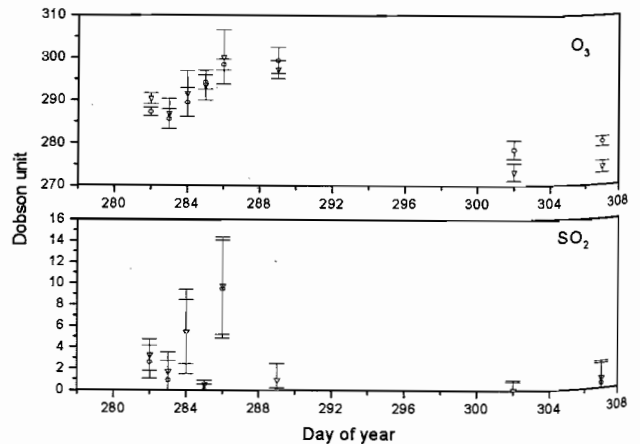


Fig. 2. Representative  $O_3$  and  $SO_2$  values from Brewers MKIV (circles) and MKII (triangles) in the second part of the campaign.

Table 3. Typical Values of Optical Thickness for Aerosol, O<sub>3</sub>, SO<sub>2</sub>, Rayleigh Scattering, and the Atmosphere Obtained from Brewer MKIII on Day 72 as a Function of Wavelength

<i>i</i>	306.3 nm	310.1 nm	313.5 nm	316.8 nm	320.1 nm
Aerosol	1.09 ± 0.18	1.14 ± 0.17	1.15 ± 0.17	1.18 ± 0.17	1.21 ± 0.16
O <sub>3</sub>	1.06 ± 0.02	0.58 ± 0.01	0.42 ± 0.01	0.23 ± 0.00	0.18 ± 0.00
SO <sub>2</sub>	0.04 ± 0.01	0.01 ± 0.00	0.01 ± 0.00	0.01 ± 0.00	0.00 ± 0.00
Rayleigh scattering	1.05 ± 0.01	0.99 ± 0.01	0.95 ± 0.01	0.91 ± 0.01	0.87 ± 0.01
Atmosphere	3.24 ± 0.18	2.72 ± 0.17	2.53 ± 0.17	2.33 ± 0.17	2.26 ± 0.16

mately 0.02, AOT is wavelength increasing by approximately 0.12. Because the latter is as much as six times the former, it can hardly be believed that the wavelength behavior of the AOT values is affected by the uncertainties, even though some minor influence cannot be denied.

The Brewers had worked side by side under the same schedule. AOT values have been compared by use of both the square correlation coefficient ( $R^2$ ), from the linear regression for obtaining  $\tau_{at}$ , and the average relative difference ( $D$ ). For a wavelength  $\lambda$  the ratio of the difference between the AOT values of the two Brewers to their average is the relative difference. Then  $D$  is the average among the relative difference values for the five wavelengths. Those parameters are shown in Tables 1 and 2. The limits for acceptance of  $\tau_{ae}$  have been chosen as  $R^2 \geq 0.85$  and  $D \leq 20\%$  because they are associated with reliable sets of ds measurements. Sometimes, even for an apparently cloudless day condition, ds measurements plotted along a period can show some incongruities due to AOT variations that are undetectable by a naked-eye observation. Every time any heterogeneous plume of aerosols had been detected in the

sky during a set of ds measurements, AOT values of  $R^2 < 0.85$  and  $D > 20\%$  were obtained. This fact has been used to disqualify the AOT values associated with it.

A question rising from  $D \leq 20\%$  as a limit is if two Brewers were working under identical conditions, looking at the same point in the sky at the same time, why is  $D$  as large as 20%? Brewer ds measurements are not calibrated ones, and they depend on the efficiency of the photomultiplier tube inside the Brewer. Thus ds measurements from two Brewers are photon counts that can be different from each other even though the Brewers can be looking at the same point. As Eq. (1) suggests, the lack of calibration of ds measurements is not important because  $\tau_{at}$  is not dependent on the units of  $I_\lambda$  and  $I_{\lambda_0}$ . Nevertheless, Bais<sup>17</sup> has proposed a method to calibrate Brewer ds measurements.

The AOT variation during ds measurements can pose a huge problem to determine AOT values by the application of the Langley method. The coefficient  $\ln(I_{\lambda_0})$  in Eq. (1) could be used to get some information about the variation of the AOT. If a set of ds measurements were made daily under aerosol stable

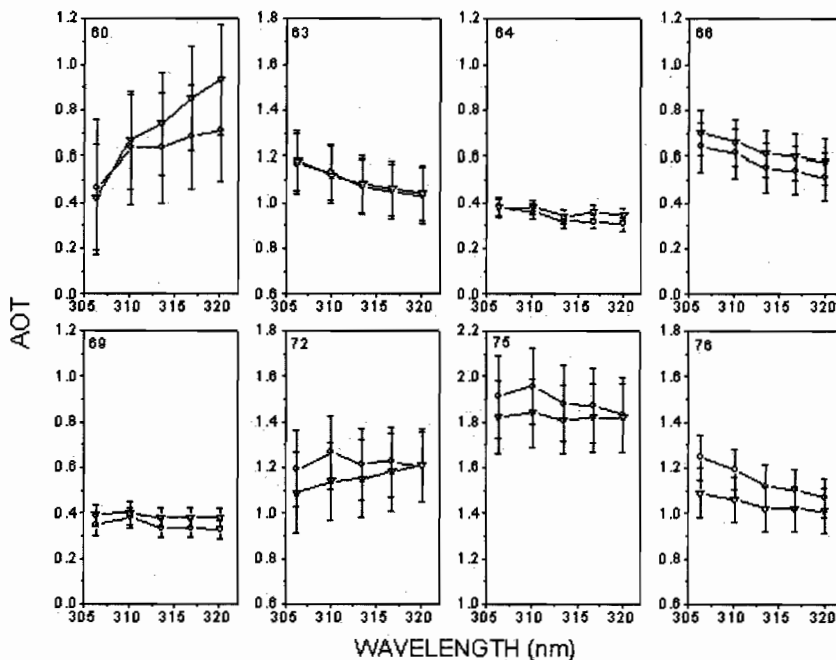


Fig. 3. AOT values as a function of wavelength from Brewers MKIV (circles) and MKIII (triangles). Except for day 69, all values are related to morning periods.

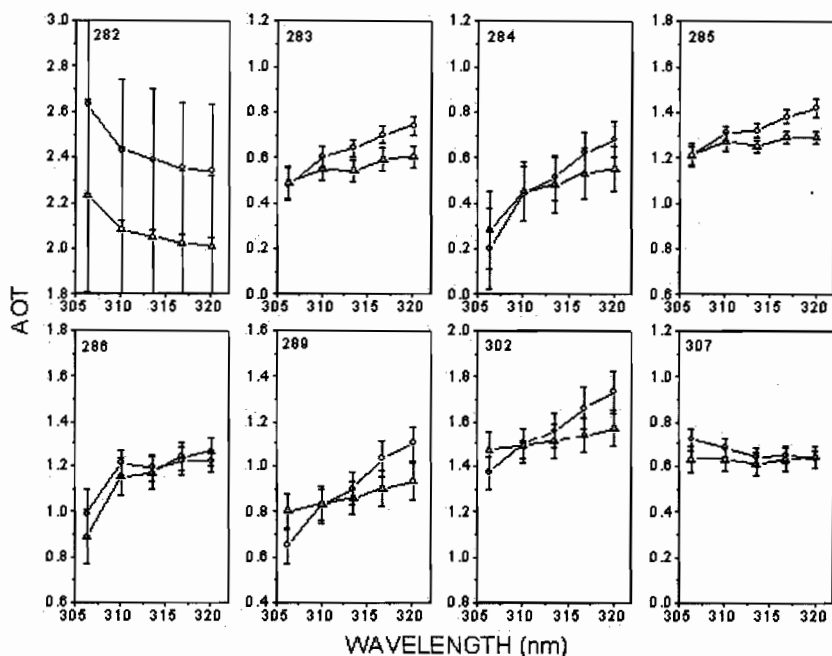


Fig. 4. AOT values as a function of wavelength from Brewers MKIV (circles) and MKII (triangles). All values are related to morning periods.

layer conditions, low variability could be expected from the  $\ln(I_{\lambda 0})$  value for all the measured UVB wavelengths in a couple of days. In the first part of our campaign,  $\ln(I_{\lambda 0})$  equals  $20.778 \pm 0.568$  (1 sd). Even if we had used Bais's method to convert  $I_{\lambda 0}$  into physical units [watts per (square meter times nanometer)], the interpretation of low variability for it as a sign of constancy of the atmospheric condition must be taken with care because AOT variations along a set of ds measurements can be offset (see Fig. 3 at Kirchoff *et al.*<sup>1</sup>), leading to a value of  $I_{\lambda 0}$  similar to that for a stable atmosphere.

Research has been done to develop a criterion to improve the confidence in the AOT values from the application of the Langley method under aerosol layer variable conditions. That is a hard issue in that changes can be not only in the quantity but also in the chemistry and size of the aerosols.<sup>18,19</sup> A quantity variation can be reflected in  $\tau_{ae}$  by augmentation (or diminution) of its value and growth of  $\delta_{ae}$ . Nevertheless, such a type of variation does not change the AOT wavelength behavior. On the other hand, according to Abel *et al.*,<sup>19</sup> changes in the chemical composition and size of aerosols in a course of a few hours can lead to variations in their optical properties. Could this result in AOT wavelength behavior changes? Some recent papers such as Jaroslowski *et al.*<sup>20</sup> have found similar results but regarded AOT wavelength behavior as a problem linked to the Angstrom coefficient. As can be seen in Figs. 3 and 4, there are cases of both AOT wavelength increasing and decreasing behavior.

The whole campaign had been driven to show the consistency of the methodology in obtaining AOT by use of Brewer ds measurements. The first part had

also been intended to determine whether AOT values were affected by stray light. For this, Fig. 3 shows four out of eight cases (days of year 60, 69, 72, and 75) in which, at least for the two shortest wavelengths, a wavelength increasing behavior was observed. Researchers, who had been suspecting stray-light contamination, were surprised to see that Brewer MKIII data showed not only a wavelength increasing behavior but also, as seen on days 60 and 72, a stronger one when compared with MKIV data. After these results, it can be said stray-light effect was not present, or at least not significantly, on the AOT values obtained in this proceedings.

#### 4. Conclusions

In this paper we investigated the procedure to obtain AOT values from Brewer ds measurements and determine whether stray light was acting on them. The Brewers' AOT values, which are in good agreement, have been associated with square correlation coefficients  $R^2 \geq 0.85$  and average relative difference  $D \leq 20\%$ . Even though such a criterion has delivered values matching quite well, we believe that more applications to compare AOT values among Brewers and other instruments should be made to improve the confidence in the procedure.

The doubt about stray light as causing the wavelength increasing behavior of the AOT values has been removed in that the comparison between values from double- and single-monochromator Brewers in the first part of the campaign has clearly showed the lack, or the low significance, of such an effect on the measurements. In some cases the double monochromator had even determined AOT values with stronger wavelength increasing behavior.

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