

Intercomparison of aerosol optical depth from Brewer ozone spectrophotometers and CIMEL sunphotometers measurements

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Abstract

The Langley plot method applied on the Brewer Ozone measurements can provide accurate Aerosol Optical Depth (AOD) in the UV-B. We present seven intercomparisons between AOD retrieved from Brewer Ozone measurements and AOD measured by CIMEL sunphotometer, which are stored in the international AERONET database. Only the intercomparisons between co-located instruments can be used to validate the Langley Plot method applied to the Brewer measurements: in this case, all the correlation coefficients are above 0.83. If the instruments are not at the same site, the correlation between the AOD retrieved by both instruments is much lower.

1 Introduction

The density of the aerosol particles is a key parameter in the radiative budget of the atmosphere. The direct aerosol effect on the radiative budget in absorbing and scattering the solar radiation is important (Roberts and Jones, 2004; Wild et al., 2005, 2007; See et al., 2006; Chou et al., 2006; Pfeifer et al., 2006; Mallet et al., 2005). Aerosol particles have also an important indirect effect as they play the role of the cloud condensation nuclei in the atmosphere (IPCC, 2001). The retrieval of the aerosol parameters as the Aerosol Optical Depth (AOD) appears to be an important challenge of the research community.

The Brewer network consists of about 150 spectrophotometers (<http://www.io3.ca/>), which are measuring in the UV-B radiation band. The retrieval of AOD from these instruments using the Langley Plot Method (LPM) represents a good complementary information to the AOD in the visible spectrum obtained with other instruments (de La Casinière et al., 2005). The method was developed and described in previous studies (Marenco et al., 1997; Carvalho and Henriques, 2000; Groebner et al., 2001; Cheymol and De Backer, 2003; Bais et al., 2004; Kazadzis et al., 2005). Validation studies have also already been done (Groebner and Meleti, 2004; Jaroslowski and Krzyscin, 2005;

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Savastiouk and Mc Elroy, 2005; Cheymol et al., 2006; Sellitto et al., 2006) to provide information on the accuracy of the AOD in the UV-B from Brewer instruments.

In this paper, the intercomparisons between AOD from Brewer spectrophotometers and CIMEL sunphotometers measurements confirm the accuracy of the AOD obtained from the Brewer Ozone measurements in using the LPM and under the condition that the Brewer instruments are well calibrated.

The method used to retrieve the AOD from Brewer Ozone measurements is explained shortly in the Sect. 2. The validation of the AOD obtained in using LPM will be made in Sect. 3 in comparing AOD from Brewer spectrophotometers and the CIMEL sunphotometers belonging to the AERONET database (<http://aeronet.gsfc.nasa.gov/>) at seven different stations.

2 Method

The LPM is used to retrieve the AOD from the Brewer spectrophotometer. The instruments are calibrated by using this linear regression method and then the Calibration Factor (CF) can be determined: the CFs for Uccle, El Arenosillo, Seoul, Norrköping and Rome are constant over the period considered here. For Toronto, as there was some technical changes in the Brewer's calibration, the CFs are levelled: they are constant from 1984 to February 1992, then they are shifted for the period March 1992 to December 2002 and changed again from January 2003 to December 2004 and finally shifted again from January 2005 up to now. For Hong Kong, the CFs are interpolated over the whole period as they change during the period. The error due to the error on the CFs are shown in Table 1 for each Brewer and for the wavelength 320.1 nm. The error on AOD due to error on CF is, for most of the Brewer, below 0.03 except for the Brewer in Hong Kong and Seoul, where it is equal to 0.05 and 0.04, respectively.

The AOD are calculated only for cloudless days. To select these days, several tests are applied on the Brewer ozone measurements (Cheymol et al., 2006). To remove the stray light effect, which contaminated the AOD values (Cede et al., 2006; Arola and

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Koskela, 2004), a test on the airmass factor is added compared to previous studies (Cheymol and De Backer, 2003; Cheymol et al., 2006). Once the calibration of the Brewer spectrophotometer is determined with LPM, the AOD can be calculated for each individual Direct Sun (DS) measurements. Below, the list of the tests to select cloudless days to perform LPM applied on the Direct Sun radiation are summarised:

1. The individual DS data for which the airmass is above 3 are removed.
2. The ozone column and standard deviation is computed on each group of five individual DS measurements for each wavelength. Data are accepted if the standard deviation is lower than 2.5 DU.
3. The range of zenith angles covered by valid DS observations on one day must be at least 20°.
4. The number of the individual DS data must be at least 50 per day (i.e. 10 sequences of 5 observations).
5. The distance between each point and the Langley Plot regression line must be lower than 4.
6. The daily mean absolute deviation of the distance between each point and the Langley Plot regression line must be lower than 0.055.

For more details on the method used to calibrate the Brewer instrument, see Cheymol and De Backer (2003) and Marengo (2002).

3 Intercomparisons between AOD from Brewer spectrophotometer and CIMEL sunphotometer

The automatic CIMEL observes the direct sun and scans the sky at several wavelengths (340, 440, 670, 870 and 1020 nm) according to AERONET protocols, as part of this network (Holben et al., 1998).

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During routine ozone observations, the Brewer spectrophotometer makes 5 individual Direct Sun (DS) ozone observations within 3 minutes at five wavelengths in the UV-B (306.3 nm, 310.1 nm, 313.5 nm, 316.7 nm and 320.1 nm) with a Full Width Half Maximum (FWHM) of about 0.6 nm. This intercomparison uses only the largest wavelength of the Brewer spectrophotometer at 320 nm as it is the closest wavelength to the wavelength of the CIMEL, which is for the CIMEL sites selected 340 nm or 440 nm (see Table 2). The sun tracking of the Brewer is checked regularly by visual inspection. The stability of the tracking system is claimed by Kipp and Zonen to be of the order of 0.2° . This should be a random error and should not cause a bias on the AOD measurements. For Brewer Mark II like at Uccle, Toronto, a stray light effect exists for wavelength below 313.5 nm, which influences the AODs (Arola and Koskela, 2004). In this study, as only the 320.1 nm wavelength is considered, this effect is negligible on AOD. The range of the zenith angle for all the Brewer is between at the lowest 13.68° (at El Arenosillo) up to 71.10° as all the AOD measurements for which the airmass is above 3 are removed (see Table 3).

Figure 1 shows the location of the different stations used in this study. The common period of each intercomparison is listed in Table 2.

At Norrköping in Sweden, the AODs obtained from the Brewer ozone measurements have already been validated with AOD from a co-located sunphotometer (Cheymol et al., 2006).

Quasi simultaneous AOD from the Brewer and the CIMEL are compared. We consider only measurements of both instruments with a maximum time difference of three minutes.

3.1 Co-located instruments

3.1.1 Uccle, Belgium

Uccle is located near Brussels in Belgium ($50^\circ 48' N$, $4^\circ 21' E$, 100 m a.s.l.) in a residential area. It is strongly affected by pollution (BIM, 2004). Two Brewer instruments

are situated on the roof of a building of the Royal Meteorological Institute of Belgium (RMIB) and are re-calibrated every 2–3 years since 2001. The AOD from the single monochromator MKII #016 will be used for this intercomparison with the co-located CIMEL belonging to the Belgian Institute for Space Aeronomy.

The single monochromator Brewer MARK II #016 (SCI TEC, 1988) and the CIMEL sunphotometer (www.cimel.fr/index_us.html) are 100 m far from each other. They are considered as co-located instruments. The two instruments measure the direct solar intensity at 320 nm and at 340 nm, respectively. The CIMEL sunphotometer operated since July 2006. Since 1984, the Brewer spectrophotometer #016, which is a single monochromator is operated routinely at Uccle to measure the total ozone column in the atmosphere from the Direct Sun (DS) ultraviolet radiation.

Figure 2 shows the scatter plot of the 368 AODs from Brewer #016 and the CIMEL sunphotometer at Uccle in Belgium since April 2006. There is a very good linear relation between AODs from the two instruments: the correlation coefficient, the slope and the intercept of the regression line are 0.96, 0.93 ± 0.01 and -0.06 ± -0.01 , respectively. This bias of 0.06 is statistically significant. Nevertheless, this value is one order of magnitude below the one of the AOD value at Uccle.

3.1.2 Toronto, Canada

The Brewer spectrophotometer #008 is located at the top of the building in a big Canadian city ($43^{\circ}78' N$, $79^{\circ}468' W$). Its calibration is maintained by Environment Canada, who also keeps the World standard triad for Brewer observations (Fioletov et al., 2005). It is a Mark II spectrophotometer. This instrument is a part of the Brewer triad and is calibrated on a regular basis at Mauna Loa, Hawaii. The site is moderately affected by pollution. It is located in urban area near the intersection of two busy streets in the vicinity of a large park. The co-located CIMEL sunphotometer measures at 340 nm.

At Toronto, the single monochromator MARK II Brewer #008 and the CIMEL sunphotometer are operated since 1984 and 1996, respectively. 577 quasi simultaneous AODs at 320 nm and at 340 nm are compared from the Brewer and the CIMEL, re-

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spectively on Figure 3. The correlation coefficient is high, 0.99. The slope and the intercept of the regression line are 0.91 ± 0.006 and 0.019 ± 0.002 , respectively. A very high correlation coefficient is also found at Toronto.

3.1.3 El Arenosillo, Spain

5 The Brewer spectrophotometer #150 and the CIMEL sunphotometer are operated since 2000 on the roof of INTA (Instituto Nacional de Tecnica Aeroespacial), which is in El Arenosillo located near Huelva (37° N, $6^\circ 12'$ W, 17 m a.s.l.) on the Southwestern Spanish coast. El Arenosillo is surrounded by a pine tree forest and is located within Donana National Park, a protected coastal area.

10 The Brewer spectrophotometer #150, which is a MARK III, operates routinely at El Arenosillo to provide ozone values and global UV irradiance since 1998.

The CIMEL sunphotometer and a MARK III Brewer spectrophotometer are co-located and measure the direct solar radiation at 440 nm and 320 nm, respectively. Figure 4 shows the scatter plot of the 7351 AODs from Brewer #150 and the CIMEL sunphotometer at El Arenosillo in Spain. It shows that there is a good linear relation between AODs from the two instruments: the correlation coefficient, the slope and the intercept of the regression line are 0.83, 0.60 ± 0.005 and 0.016 ± 0.0016 , respectively.

15 Nevertheless, a bias still exists. This can be attributed to the large difference of wavelengths used for both instruments. The important difference here could also be explained by the wavelength dependence of AOD. This depends on the type and size of the aerosol. Larger is the difference in particle composition, larger is the discrepancy between AOD from the two instruments.

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3.2 Brewer and CIMEL at different sites

3.2.1 Seoul, Korea

Seoul is a big city in Asia (37°34' N, 126°58' E), which is mostly influenced by pollution from industry in the surroundings. The Brewer Mark IV spectrophotometer #148 and the CIMEL sunphotometer are located at Yonsei University and Seoul National University, respectively, which is 16 km apart within Seoul in Korea. Yonsei university is located in downtown Seoul, while Seoul National University is at the southern part, which is less polluted. They are operated since 2000 and 1999, respectively. The AOD at 440 nm from the CIMEL and 320 nm from the Brewer are compared.

In Seoul, 45 quasi simultaneous AODs at 320 nm from the Brewer #148 are compared with AOD from the CIMEL at 440 nm on Fig. 5. The correlation coefficient, the slope and the intercept are 0.79, 0.60 ± 0.07 and 0.11 ± 0.04 , respectively. There is a good linear relation between the two instruments but there is a noticeable bias. Nevertheless, a large difference in wavelength also exist (320 and 440 nm for the Brewer and the sunphotometer, respectively).

As we only have 45 data, no final conclusion can be done on the accuracy of the AOD retrieved by the Brewer spectrophotometer in Seoul.

3.2.2 Hong Kong in China

The spectrophotometer MARK IV spectrophotometer #115 is operated in the south eastern part of Hong Kong, which is a big city in China (22°13' N, 114°15' E, 60 m a.s.l.). It is located at a coastal site on top of a cliff facing South China Sea. This remote station encounters continental outflow in wintertime and clean maritime air in summertime. The CIMEL sunphotometer is operated in the centre of the city on the roof of the library of the Hong Kong Polytechnic University (22°18' N, 114°10' E, 30 m a.s.l.), which means that it represents the urban aerosol conditions. The CIMEL is 12 km far from the Brewer. The AOD at 340 nm is taken account for this study as it is the closest wave-

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length to the one of the Brewer at 320 nm. The Brewer and the CIMEL are operated since 1995 and 2005, respectively.

161 AODs are compared from the Brewer spectrophotometer #115 at 320 nm and the CIMEL sunphotometer at 340 nm on Fig. 6. The linear relation between the instruments is poor (only 0.59), the slope and the intercept of the regression line are 0.50 ± 0.05 and 0.36 ± 0.04 , respectively. Figure 7 shows the time series of the AOD from the CIMEL (in blue) and the Brewer (in red) during the common period. In 2006, the CIMEL measures larger AOD than the Brewer, which is quite not usual: in all the stations, the Brewer measure larger AOD than the CIMEL.

The Brewer and the CIMEL are representing the urban and the rural atmospheric conditions respectively, which can explain part of this difference.

3.2.3 Rome in Italy

Rome ($41^{\circ}54' N$, $12^{\circ}31' E$) is a very populated city strongly influenced by anthropogenic activities (Meloni et al., 2000). The CIMEL sunphotometer operated since 2001 on the roof of the ISAC-CNR building in the southern suburbs of Rome of about 15 km far from the city centre in a semi-rural site. The Brewer spectrophotometer #067 is operated since 1992 in the centre of Rome westerly from the CIMEL. The AOD at wavelength 440 nm from the CIMEL are compared to the AOD at 320 nm from the Brewer.

3418 quasi simultaneous data are compared on Figs. 8 and 9. There is a low linear relation (0.52) between the AODs from the two instruments and it is probably due to the different location of the instruments: the Brewer spectrophotometer is strongly influenced by anthropogenic activities and the CIMEL is in a semi-rural site, which is less affected by pollution. Moreover, some of the AOD retrieved from the Brewer spectrophotometer are too high in 2003 (above two) to be realistic. This could be attributed to technical problem. The two instruments measure also at different wavelength, which increase the difference between the AOD.

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Table 4 summarises all the intercomparisons. Three main conclusions can be made:

- If the instruments are co-located and measure at wavelength very close to each other (320 nm and 340 nm), the correlation coefficient is very high, the slope of the regression line is about 0.9 and its intercept is virtually equal to 0.
- With two co-located instruments measuring at two wavelength far from each other, the intercomparisons is worse. A bias appears as in El Arenosillo.
- For two instruments operated at different sites like in Rome or in Hong Kong, the correlation is very poor. It confirms the previous conclusion in Cheymol et al. (2006): AOD validation needs to have co-located instruments. The difference between AOD from the two instruments have different cause: the difference of the wavelength (El Arenosillo), the poor co-location (Hong Kong and Rome) and probably technical problems (Rome). Moreover, some error can be induced by an error on the neutral density filter values (Cheymol et al., 2006). This study is needed to go further to determine the main source of the discrepancy between AOD from Brewer and CIMEL at El Arenosillo, Hong Kong and Rome.

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Table 1. Contribution of the Error on Calibration Factor (CF) to the Error on the AOD's error for the wavelength 320,1 nm for each sites. N is the number of CFs used to calculate the average of the CF.

Site	Mean CF	Error on AOD due to CF error	N
Uccle	17.83±0.21	0.03	64
Toronto	17.30±0.15	0.02 (1984–1992)	91
	18.36±0.12	0.01 (1992–2003)	77
	18.46±0.13	0.03 (2003–2004)	27
	18.34±0.11	0.02 (2005–2007)	29
El Arenosillo	19.61±0.11	0.01	353
Norrköping	18.92±0.09	0.02	21
Seoul	18.47±0.09	0.04	5
Hong Kong	18.11±0.34	0.05	45
Rome	19.03±0.17	0.02	50

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Table 2. List of the sites (Uccle in Belgium, Toronto in Canada, El Arenosillo in Spain, Norrköping in Sweden, Seoul in Korea, Hong Kong in China and Rome in Italy), the instruments (B**** for the Brewer) and the distance between the instruments, the wavelength λ (in boldface for Brewer) and the period of the data used. The instruments in Sweden are also listed.

Site	Instruments	λ (nm)	Period used
Uccle	co-located: B#016 MkII , CIMEL	320.1 , 340	2006-2007
Toronto	co-located: B#008 MkII , CIMEL	320.1 , 340	1996,2003-2007
El Arenosillo	co-located: B#150 MIII , CIMEL	320.1 , 440	2000-2006
Norrköping	co-located: B#128 MkIII , CSEM	320.1 , 368	2004
Seoul	16km far: B#148 MkIV , CIMEL	320.1 , 440	2000-2001
Hong Kong	12 km far: B#115 MarkIV , CIMEL	320.1 , 340	2005-2006
Rome	15km far: B#067 MkIV , CIMEL	320.1 , 440	2002-2006

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Site	Range of zenith angle in °
Uccle	28.44–71.09
Toronto	20.34–71.08
El Arenosillo	13.68–71.09
Norrköping	35.16–71.09
Seoul	52.70–70.59
Hong Kong	41.21–70.93
Rome	18.46–71.09

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Table 4. Comparisons between AODs from different Brewer and CIMEL sunphotometers : at Uccle in Belgium (#016), at Toronto in Canada (#008), at El Arenosillo in Spain (#150), at Norrköping in Sweden (Cheymol et al., 2006), at Seoul in Korea (#148), at Hong Kong in China (#115) and at Rome in Italy (#067). The slope (a) and the intercept (b) of the regression line are in parenthesis and the correlation coefficient (c) is outside the parenthesis. λ_{CIMEL} is the wavelength used for the CIMEL and N is the number of measurements compared.

Brewer	c, a and b	λ_{CIMEL} (nm)	N
#016	0.96 (0.93±0.01;-0.06±0.01)	340	368
#008	0.99 (0.91±0.006;0.019±0.002)	340	577
#150	0.83 (0.60±0.005;0.016±0.002)	440	7351
#128	0.98 (0.85±0.004;0.02±0.0014)	368	1763
#148	0.79 (0.60±0.005;0.016±0.002)	440	45
#115	0.59 (0.50±0.05;0.36±0.04)	340	161
#067	0.52 (0.19±0.005;0.18±0.003)	440	3418

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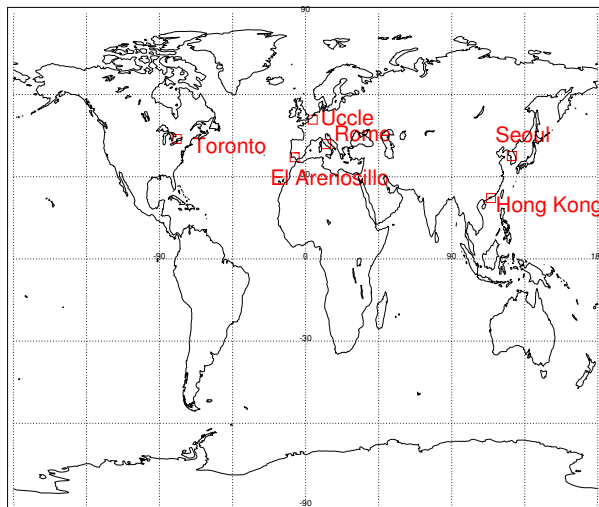


Fig. 1. The six stations used in this paper: Uccle in Belgium, Rome in Italy, El Arenosillo in Spain, Toronto in Canada, Seoul in Korea and Hong Kong in China.

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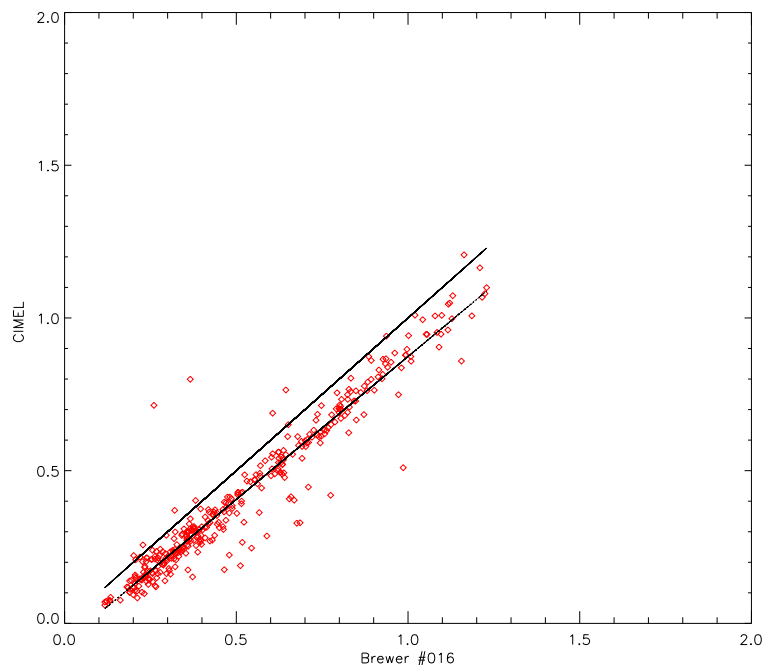


Fig. 2. 368 AODs from CIMEL at 340 nm versus the AODs from Brewer #016 at Uccle in Belgium since April 2006 up to now. The thick solid and the dotted lines represent the equation $f(x)=x$ and the linear regression line for the data, respectively. The correlation coefficient, the slope and the intercept are 0.96, 0.93 ± 0.01 , -0.06 ± -0.01 , respectively.

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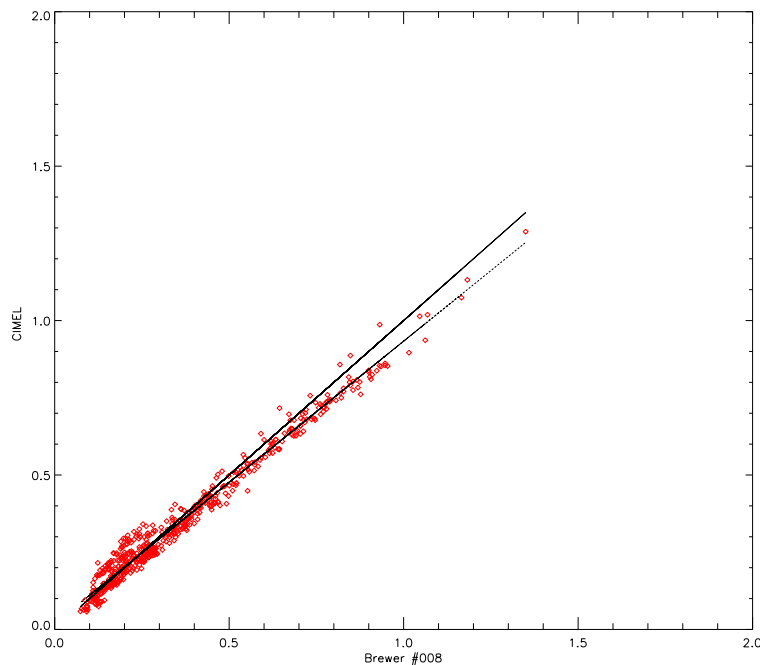


Fig. 3. 577 AODs from CIMEL at 340 nm versus the AODs from Brewer #008 at Toronto in Canada since 1996 up to now. The thick solid and the dotted lines represent the equation $f(x)=x$ and the linear regression line for the data, respectively. The correlation coefficient, the slope and the intercept are 0.99 , 0.91 ± 0.006 , 0.019 ± 0.002 , respectively.

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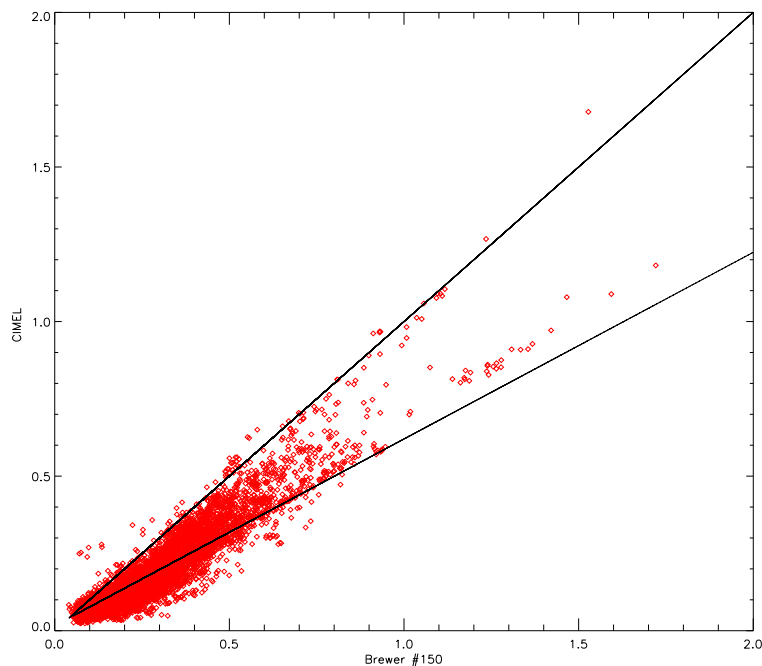


Fig. 4. 7351 AODs from CIMEL at 440 nm versus the AODs from Brewer #150 at El Arenosillo in Spain. The thick solid and the dotted lines represent the equation $f(x)=x$ and the linear regression line for the data, respectively. The correlation coefficient, the slope and the intercept are 0.83, 0.60 ± 0.005 , 0.016 ± 0.0016 , respectively.

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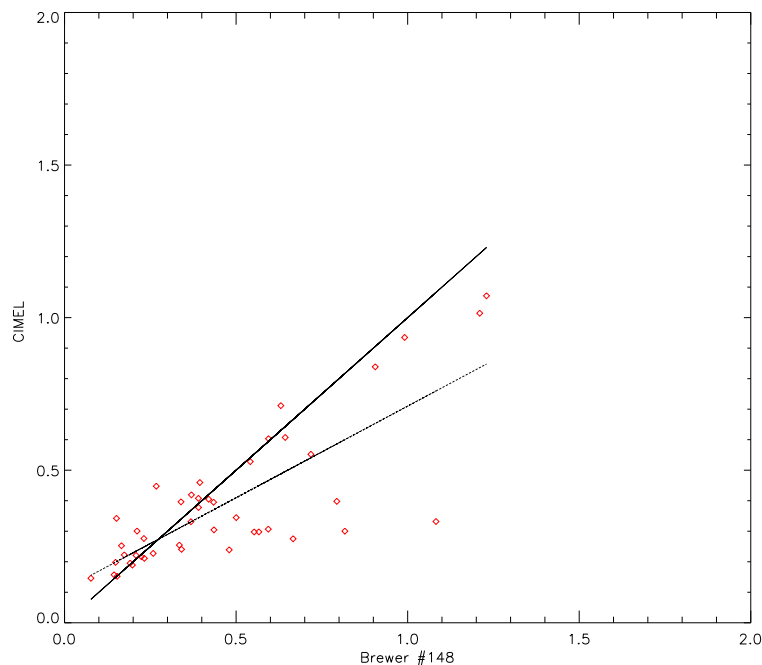


Fig. 5. 45 AODs from CIMEL at 440 nm versus the AODs from Brewer #148 at Seoul in Korea. The thick solid and the dotted lines represent the equation $f(x)=x$ and the linear regression line for the data, respectively. The correlation coefficient, the slope and the intercept are 0.79 , 0.60 ± 0.07 , 0.11 ± 0.04 , respectively.

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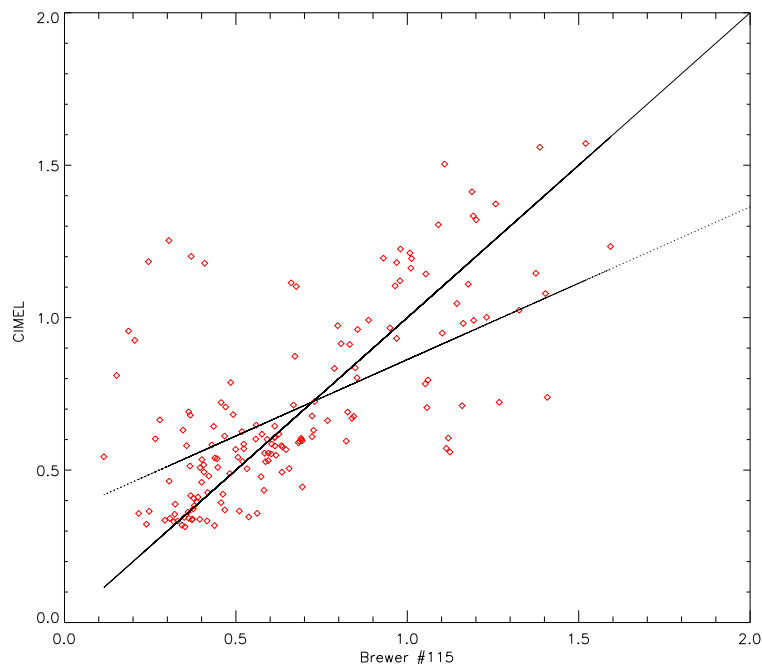


Fig. 6a. 161 AODs from CIMEL at 340 nm versus the AODs from Brewer #115 at Hong Kong in China. The thick solid and the dotted lines represent the equation $f(x)=x$ and the linear regression line for the data, respectively. The correlation coefficient, the slope and the intercept are 0.59 , 0.50 ± 0.05 , 0.36 ± 0.04 , respectively.

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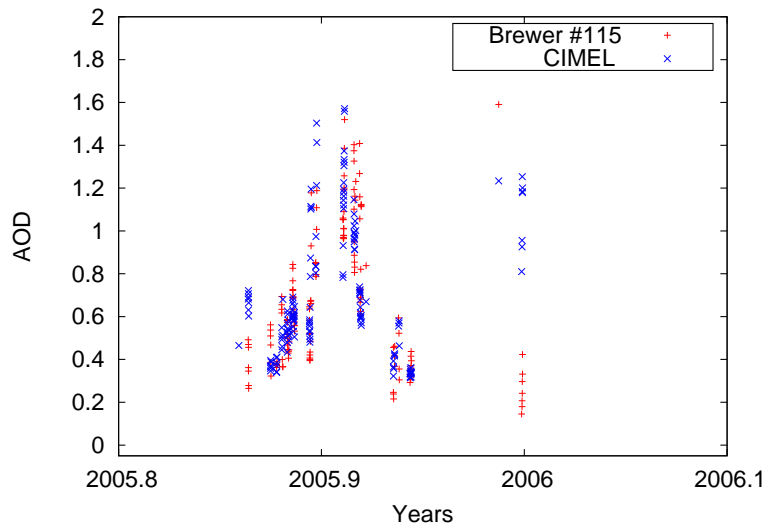


Fig. 6b. Time series of the 161 AODs from CIMEL at 340 nm and the AODs from Brewer #115 at Hong Kong in China.

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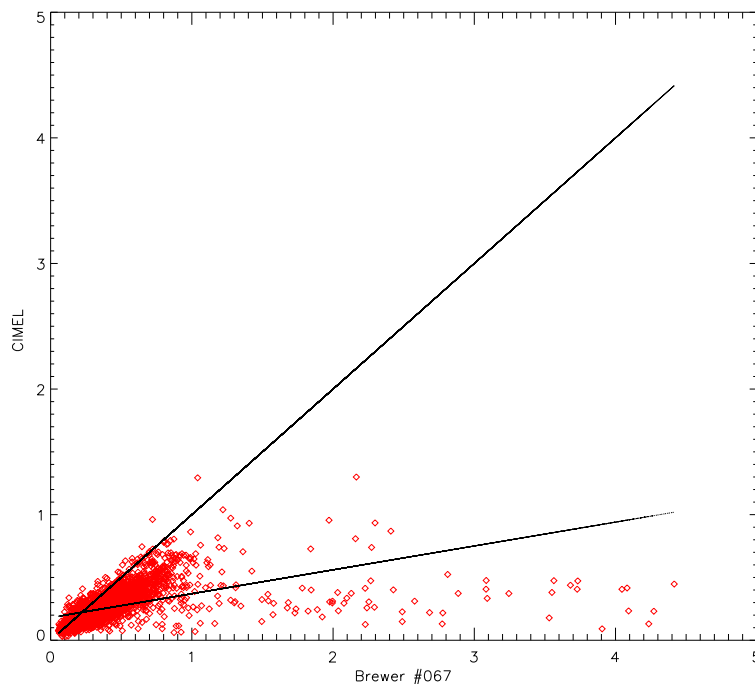


Fig. 7. 3418 AODs from CIMEL at 440 nm versus the AODs from Brewer #067 at Rome in Italy. The thick solid and the dotted lines represent the equation $f(x)=x$ and the linear regression line for the data, respectively. The correlation coefficient, the slope and the intercept are 0.52, 0.19 ± 0.005 , 0.18 ± 0.003 , respectively.

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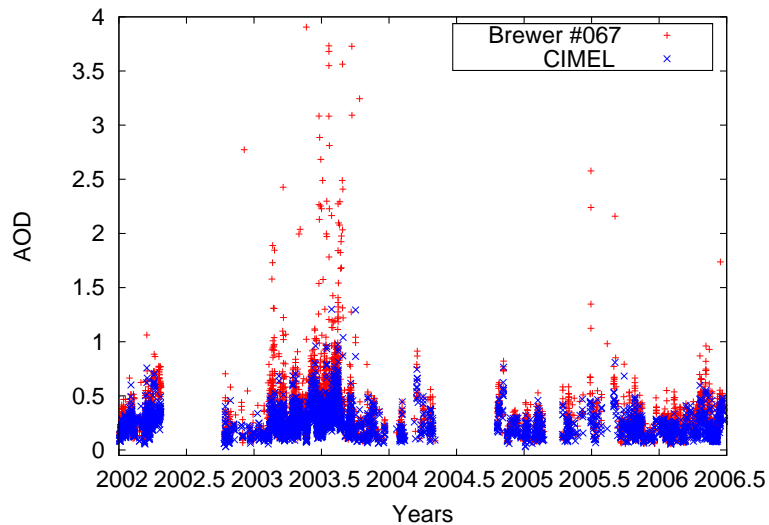


Fig. 8. Time series of the 3418 AODs from CIMEL at 440 nm and the AODs from Brewer #067 at Rome in Italy.

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