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Aethalometer measurements in a road tunnel: a step forward in the characterization of black carbon emissions from traffic

C. Blanco-Alegre [1], A.I. Calvo [1], C. Alves[2], P. Fialho[3], T. Nunes[2], J. Gomes[2], A. Castro [1], F. Oduber [1], E. Coz [4] and R. Fraile[1]

[(1)] Department of Physics, IMARENAB University of León, 24071 León, Spain

+34 987291543, rfral@unileon.es

[(2)] Centre for Environment and Marine Studies, Department of Physics, University of Aveiro, 3810-193 Aveiro, Portugal

[(3)] Research Institute of Volcanology and Evaluation – IVAR, Rua da Mãe de Deus, 9500-321 Ponta Delgada, Portugal

[(4)] Centre for Energy, Environment and Technology Research (CIEMAT), Department of the Environment, Madrid, Spain

ABSTRACT

A sampling campaign was conducted in the Liberdade Avenue tunnel (Braga, Portugal) during a week (with 56,000 vehicles) to monitor black carbon (eBC-equivalent black carbon) by means of an Aethalometer AE-31, and gaseous pollutants (CO_2, CO, NO_x). Inside the tunnel, the mean eBC mass concentration was 21 ± 10 µg m^{-3}, reaching a maximum hourly value of 49.0 µg m^{-3}. An hourly and weekday-weekend study was carried out. Regarding the Absorption Ångström exponent (AAE), a mean value of 0.97 ± 0.10 was obtained, for a source of practically pure traffic. There was a positive significant correlation between eBC and the number of light vehicles ($r=0.47$; $p<0.001$) and between eBC and the gaseous emissions: CO ($r=0.67$; $p<0.001$), CO_2 ($r=0.71$; $p<0.001$), NO ($r=0.63$; $p<0.001$) and NO_2 ($r=0.70$; $p<0.001$). The mean black carbon emission factors (EF_{BC}) inside the tunnel were 0.31 ± 0.08 g (kg fuel)$^{-1}$ and 0.11 ± 0.08 mg veh^{-1} km^{-1}, similar to those found in other studies for gasoline and diesel vehicles in road tunnels.

KEYWORDS: Absorption Ångström exponent, Aethalometer, black carbon, emission factors, pollutant relationships, vehicle exhaust emissions.

1. INTRODUCTION

Nowadays, airborne particulate matter with diameters below 10 µm (PM_{10}) has major effects on climatic change while airborne particulate matter with diameters below 2.5 µm ($PM_{2.5}$) poses a major risk to human health (WHO, 2013). In Europe, more than 90% of urban dwellers are exposed to $PM_{2.5}$ levels that exceed the reference value set by the WHO (EEA, 2013).

Traffic exhaust emissions of motorized vehicles are one of the main sources of $PM_{2.5}$ in many urban areas (Bycenkiene et al., 2014; Sun et al., 2013). Besides, traffic non-exhaust emissions, such as particles from tyre wear, brakes, road surface abrasion and dust resuspension, are one of the principal contributors to airborne particulate matter, especially in semi-enclosed places like tunnels (Querol et al., 2004; Thorpe and Harrison, 2008). While strict policies have led to significant reductions in exhaust emissions, currently non-exhaust emissions from road vehicles, are unabated (Padoan and Amato, 2018; Thorpe and Harrison, 2008). Data from European cities showed that exhaust and non-exhaust sources contribute, at least, equal amounts to total traffic-related emissions (Amato et al., 2016, 2014; Denier van der Gon et al., 2013). One of main constituents of $PM_{2.5}$ is black carbon (BC). It is emitted from incomplete combustion of fossil fuel or biomass and it is a carbonaceous material that is formed primarily in flames and directly emitted to the atmosphere. BC presents particular physical properties: it strongly absorbs visible light and is refractory with a vaporization temperature of around 3700 °C (Bond et al., 2013). BC pollution has been linked to respiratory infections (such as adverse effects on lung function and increased cancer risks) and cardiovascular diseases, as well as to increased morbidity and mortality among different age groups (Silverman et al., 2012; Suglia et al., 2008). Globally, BC is considered a major short-lived climate forcer through direct radiative forcing and cloud, sea-ice and snow effects. The global mean radiative forcing caused by BC was estimated to be from 0.4 to 1.2 W m^{-2}, becoming the second man-mad strongest contributor after CO_2 (Bond et al., 2013; Ramanathan and Carmichael, 2008). Hence, the study of BC concentration is crucial due to its effects on multiple essential policy objectives (e.g., climate, energy, air quality, public health, etc.) (EEA, 2016; Kinney, 2008; Tong et al., 2016).

The aethalometer (Hansen et al., 1984) has become extensively used over the last years, and especially the seven wavelength (from near-ultraviolet to near-infrared) model, to measure the aerosol light absorption. The use of the mass absorption cross-section, proposed by the manufacture, allows the calculation of the equivalent black carbon (eBC) concentration, defined as the light absorbing constituent considered BC (Sandradewi et al., 2008b).

Aiming at determining vehicle emissions, studies should be carried out in areas where traffic is the main pollution source. In the last years, different methods have been used to analyse vehicle emissions. The chassis dynamometer methods enable to test vehicles under controlled laboratory conditions. This procedure ensures a high repeatability of results, but is very costly (Alves et al., 2015a; Traver et al., 2002). In addition, it does not allow reproducing the real-world conditions (Franco et al., 2013). However, studies carried out in tunnels describe the real-world emission behaviour of on-road vehicles, capturing both exhaust and non-exhaust emissions (Alves et al., 2016b, 2016a; Handler et al., 2008; Kristensson et al., 2004; McGaughey et al., 2004; Pio et al., 2013). Hitherto, most of the studies in tunnels focused on PM_{10}, $PM_{2.5}$ and emissions factors of gaseous pollutants, but current information on black carbon concentration in tunnels is still scarce.

In this paper, black carbon emissions are analysed from a sampling campaign carried out in an urban roadway tunnel in Braga (Portugal). Furthermore, correlations between eBC and gaseous emissions and number of vehicles are discussed. This study, together with the ones already published about this campaign (Alves et al., 2015b, 2016b), provide a complete characterization of the particulate material emitted by vehicles. Aethalometer measurements in road tunnels can supply a valuable information regarding Absorption Ångström Exponent (AAE) and black carbon emission factors for application in models and for updating emission inventories.

2. EXPERIMENTAL

2.1. Sampling site and measurements

The study site is a road tunnel in Braga (Portugal), a city located in the west of the Iberian Peninsula (41°33′N, 08°25′W and 215 m above sea level). Braga is the third most populated city in Portugal, with about 200,000 residents in 2011 and a population density of 1000 inhabitants km^{-2}. The tunnel connects two main avenues to Liberdade Avenue in the centre of the city, and habitually has large traffic intensity (~ 15,000 vehicles per day) (Alves et al., 2015a,b). Sampling of BC and gaseous pollutants (CO, CO_2 and NO_x) has been carried out continuously for 7 days from 1 to 8 February 2013 (Friday to Thursday), at two sampling points, one outside (urban background site) and other inside the tunnel (Fig. 1). Except for the aethalometer, which was installed only in the tunnel, the other sampling devices were mounted at both locations.

The tunnel consists of a single parallelepiped shaped reinforced concrete bore that is 1040 m long, carrying two lanes in most of its extension of one-way traffic. Traffic volume by vehicle type through the tunnel was manually counted at 15-minute intervals throughout each of the sampling days (8:00–20:00 h, local time). Traffic count data was grouped as follows: light vehicles (a), trucks (b), heavy diesel vehicles (c) and total number of vehicles (d). The ventilation system (smoke extraction fans) was cut off during the sampling campaign.

An automatic CO and CO_2 infrared monitor from Gray Wolf (WolfSense IQ-610) was installed inside the tunnel after calibration and intercomparison with an air quality meter from TSI, model 7525, which was used outdoors. The continuous monitoring of NO and NO_2 was done by using chemiluminescence analysers from Environment S.A. (model 31M). The campaign included parallel high-volume PM_{10} sampling on quartz filters, from 8:30–12:00 h, 12:00–16:00 h and 16:00–18:00 h, in the tunnel and at the urban background station. The filters were then analysed for organic and elemental carbon (OC and EC) by a thermal-optical system (Pio et al., 2011). A more detailed description of the sampling campaign can be found in Alves et al. (2015b).

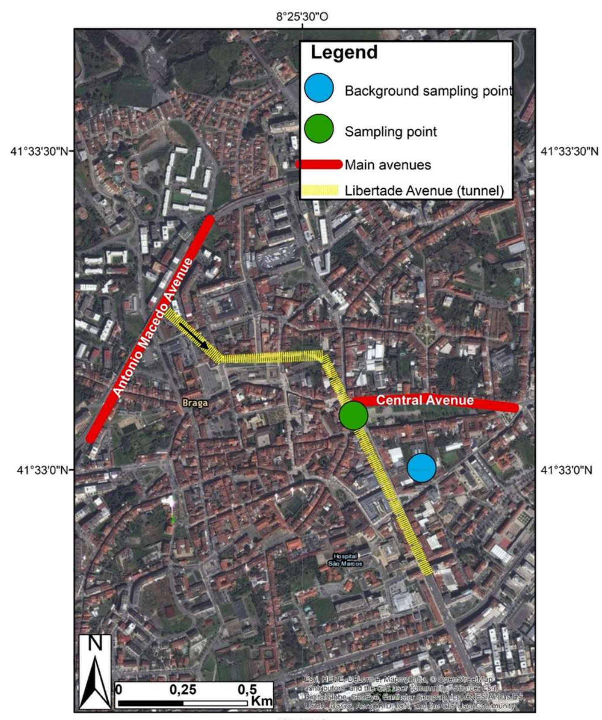

Fig. 1. Geographic location of the sampling site in Braga (Portugal). Yellow fractional line indicates the tunnel, the arrow the traffic direction, the blue dot represents the background sampling point and the green dot the sampling point inside the tunnel. The main avenues of Braga were represented with continuous red lines.

2.2. Black carbon data

Aerosol light-absorption at seven wavelengths (370, 470, 520, 590, 660, 880 and 950 nm) was continuously measured during the sampling campaign with an aethalometer Model AE-31 (Magee Scientific, USA). The instrument operated at a flow rate between 2.3 and 3.2 STP L min^{-1} with a time resolution of 5 minutes. The Aethalometer uses a differentialradiometric optical transmission technique to determine the eBC aerosol particles suspended in the sampled air (Hansen et al., 1984). It is equipped with a quartz filter tape (Pallflex, type Q250F) to collect the aerosol particles. The concentration of eBC was determined by measuring the change in the transmittance through the filter. A detailed description of the instrument can be found in Hansen et al. (1984), Weingartner et al. (2003) and Virkkula et al. (2007). Although the measurements were made every 5-minutes, the data were averaged at a resolution of 1 h to reduce the uncertainties derived from instrumental noise, flow rate, filter spot area and detector response (Corrigan et al., 2006).

The contribution from fossil fuel (eBC_{ff}) and biomass burning (eBC_{bb}) was estimated through the aethalometer model (Sandradewi et al., 2008a). For this purpose, the absorption Ångström exponent between 470 and 950 nm ($AAE_{470-950}$) was estimated (Becerril-Valle et al., 2017; Harrison et al., 2013; Sandradewi et al., 2008a). The wavelength at 470 nm has been used rather than the 370 nm one, because results using the latter could be distorted by the presence of secondary organic aerosol (Zotter et al., 2017). The limits used for the aethalometer model in this case are $AAE_{ff} = 0.97$ (corresponding to AAE values during rush-hour traffic) and $AAE_{bb} = 1.68$ (Zotter et al., 2017).

The eBC data recorded during the sampling period were corrected following WMO/GAW Aerosol Measurement Procedures, Guidelines and Recommendations (WMO, 2016). Aethalometer data were also corrected for loading effect by using the Weingartner et al. (2003) model with the winter campaign parameters proposed by Sandradewi et al. (2008b).

2.3. Statistical analyses

A univariate analysis (i.e. mean, median, minimum, maximum, quartiles and standard deviation) and a bivariate correlation (Pearson correlations with 95% confidence intervals around the point estimates) were performed to characterize eBC in the tunnel. Pearson correlations were computed to determine the relationships between eBC parameters, gaseous emission factors (CO, CO_2, NOx) and number of vehicles.

2.4. Emission factors

Emission factors (EFs) were estimated from measurements of eBC, CO_2 and CO concentrations using the following equation (McGaughey et al., 2004):

$$EF_{eBC} = \frac{\Delta[eBC]}{\Delta[CO_2] + \Delta[CO]} \times \omega_c$$

where EF is the emission factor defined as mass of pollutant emitted per kilogram of fuel burned; $\Delta[\mathrm{eBC}]$ is the black carbon concentration inside the tunnel subtracted from the background levels (μg m^{-3}); $\Delta[\mathrm{CO_2}]$ and $\Delta[\mathrm{CO}]$ are the background-subtracted concentrations of CO_2 and CO given in μgC m^{-3} (i.e., when converting concentrations of CO_2 and CO from mol fractions to mass units, a molecular weight of 12 g mol^{-1}, rather than 44 g mol^{-1} and 28 g mol^{-1} for CO_2 and CO, respectively, was used), and ω_c is the carbon weight fraction of the fuel, 0.87 for diesel and gasoline (EPA, 2015). Organic compounds can be ignored in the denominator because their contribution to total carbon concentrations in the tunnels is negligible compared to those made by CO_2 and CO (Kirchstetter et al., 1999; McGaughey et al., 2004). It should be noted that, since no aethalometer was available outside the tunnel, the eBC concentrations in the urban background atmosphere were estimated from a ratio EC_{in}/EC_{out} of 16.3 (Alves et al., 2015b) obtained in the same sampling campaign.

These emission factors are commonly normalized to vehicle distance travelled. For this, a typical fuel consumption per unit of distance travelled by vehicle class (mass emitted per kilometre) is assumed. A composite fuel consumption value (g km^{-1}) was estimated after weighting typical consumption values by the percentage of vehicles in each category obtained through the traffic counts in the tunnel. Fuel consumptions of 48.48 L fuel / (100 km) for the diesel fleet and 8.78 L fuel / (100 km) for gasoline vehicles were taken from Brimblecombe et al. (2015).

3. RESULTS AND DISCUSSION

The eBC results complement those compiled in previous publications (Alves et al., 2015b, 2016b) from the same sampling campaign, which provide information on gaseous pollutants, and carbonaceous and elemental composition of size-segregated particles.

3.1. Equivalent Black Carbon values

Based on traffic counts, around 56,000 vehicles circulated in the tunnel during daytime hours (8:00–20:00 h, local time). The total number of vehicles for all week was estimated to be 105,000. The percentage of heavy-duty traffic in the tunnel was very low. In fact, 96% of the circulating fleet was composed of passenger cars and light-duty vehicles. Motorbikes represented 1% of the fleet, while the remaining 3% were composed of heavy-duty vehicles, from which 30% was compressed natural gas buses (Alves et al., 2016b). The daily mean of total vehicles during weekdays was around 10,000 vehicles, while during weekend a decreased in the traffic density of around 40% was registered (Table 1). The traffic of light-duty vehicles in the tunnel can be considered representative of the fleet in Portugal. At the time of the campaign, and according to data provided by the Portuguese Institute of Statistics, the percentages of passenger cars for different European emission norms were as follows: 15.0 (Euro 5), 22.8 (Euro 4), 18.4 (Euro 3), 22.9 (Euro 2), 13.2 (Euro 1) and 7.0 (pre-Euro).

In the tunnel, the daily mean eBC mass concentration was 21 ± 10 μg m^{-3} (Fig. 2), close to the limit proposed by the World Health Organization (WHO) for daily $PM_{2.5}$ concentrations (25 μg m^{-3}). The eBC concentrations reached a maximum value of 49.0 μg m^{-3} (Tuesday, 05 February 2013 at 1800 UTC) lower than that reported by Miguel et al. (1998) in Oakland, and a minimum of 0.14 μg m^{-3} (Table 1). Regarding eBC sources, eBC_{ff} was 20.7 ± 10.3 μg m^{-3}, representing 98% of total eBC, while eBC_{bb} was 0.4 ± 0.8 μg m^{-3}, showing a residual penetration of eBC into the tunnel from residential biomass combustion emissions in the city. Thus, exhaust emissions from traffic are clearly the main BC source in the tunnel. Hourly mean eBC/OC and EC/eBC ratios of 0.60 and 1.38 were obtained, respectively (Table 2). The PM_{10}/EC ratio (4.40) presented a similar value than those reported in other studies (Handler et al., 2008). The maximum values occurred between 1400 and 1900 UTC, during the rush hours, when traffic density is greater because of commuting from work. However, during the weekend, a peak was observed between 0900 and 1200 UTC, probably due to leisure and shopping activities (Fig. 3). Based on Mann-Whitney U-test, no statistically significant differences between eBC concentration during workdays and weekend, were observed (p>0.05).

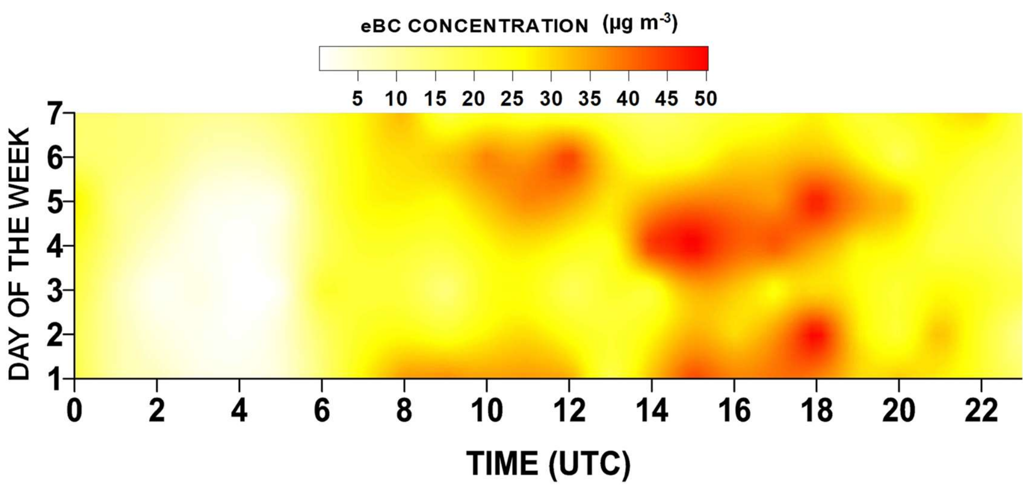

Fig. 2. Evolution of the eBC concentration (μg m^{-3}) throughout the week. The first day of the week is Monday.

Table 1. Daily eBC (μg m^{-3}), $AAE_{470\text{-}950}$ (mean, minimum and maximum), number of vehicles registered and percentage of data available during the sampling campaign.

Date	eBC			$AAE_{470\text{-}950}$			Sum of veh.	Veh. h^{-1}
	Mean	Max.	Min.	Mean	Max.	Min.		
01 February 2013	20 ± 12	45.0	1.4	0.95 ± 0.09	1.22	0.84	7319	477
02 February 2013	23 ± 8	41.7	12.6	0.92 ± 0.07	1.09	0.83	6705	339
03 February 2013	19.8 ± 4.0	27.8	13.4	1.00 ± 0.06	1.24	0.92	3738	196

04 February 2013	23 ± 12	40.0	2.0	1.00 ± 0.13	1.27	0.84	9376	426
05 February 2013	24 ± 9	49.0	9.7	0.95 ± 0.04	1.03	0.87	8865	454
06 February 2013	17 ± 8	30.1	0.14	0.98 ± 0.13	1.47	0.81	10119	426
07 February 2013	19 ± 9	30.7	1.9	1.00 ± 0.10	1.26	0.89	9844	447
Mean	21 ± 10	49.0	0.14	0.97 ± 0.10	1.47	0.81	55966	395

During weekdays, after rapid traffic intensification at 0700 UTC, eBC values were consistently higher than 25 µg m^{-3}. During nighttime (0000 to 0700 UTC), values were between 0 and 15 µg m^{-3} along all the week. However, the daily maximum reached during weekdays (49.0 µg m^{-3}) was greater than that attained during the weekend (41.7 µg m^{-3}). On weekdays, between 0600 and 2300 UTC a mean eBC concentration of 25.0 µg m^{-3} was registered, while during the weekend a mean value of 22.6 µg m^{-3} was obtained (10 % decreased compared with weekdays). A similar pattern was observed for other pollutants (CO or NO_x) in tunnels (Kristensson et al., 2004; Martins et al., 2006) or in ambient measurements like León (Spain), although with much lower eBC values (Blanco-Alegre et al., 2018), due to the dependence on traffic intensity and dispersion.

Table 2. EC, OC, PM_{10} mean values in the tunnel and ratios between PM_{10}, carbonaceous constituents and percentage of data available during the sampling campaign.

Day	Weekday	EC (µg m^{-3})	OC (µg m^{-3})	PM_{10} (µg m^{-3})	PM_{10}/EC	EC/eBC	eBC/OC	eBC/PM_{10}	N/N_T(%)
01 February 2013	Friday	21.9	32.4	143.9	6.56	1.08	0.62	0.14	44%
02 February 2013	Saturday	28.0	28.2	127.3	4.54	1.20	0.83	0.18	42%
03 February 2013	Sunday	26.2	31.2	94.6	3.61	1.33	0.63	0.21	44%
04 February 2013	Monday	26.2	51.6	134.3	5.12	1.16	0.44	0.17	44%
05 February 2013	Tuesday	34.5	37.6	150.9	4.37	1.44	0.64	0.16	43%
06 February 2013	Wednesday	30.5	25.0	106.3	3.49	1.85	0.66	0.16	45%
07 February 2013	Thursday	32.5	36.5	133.3	4.10	1.71	0.52	0.14	44%
Mean		29.1	35.3	128.1	4.40	1.38	0.60	0.16	44%

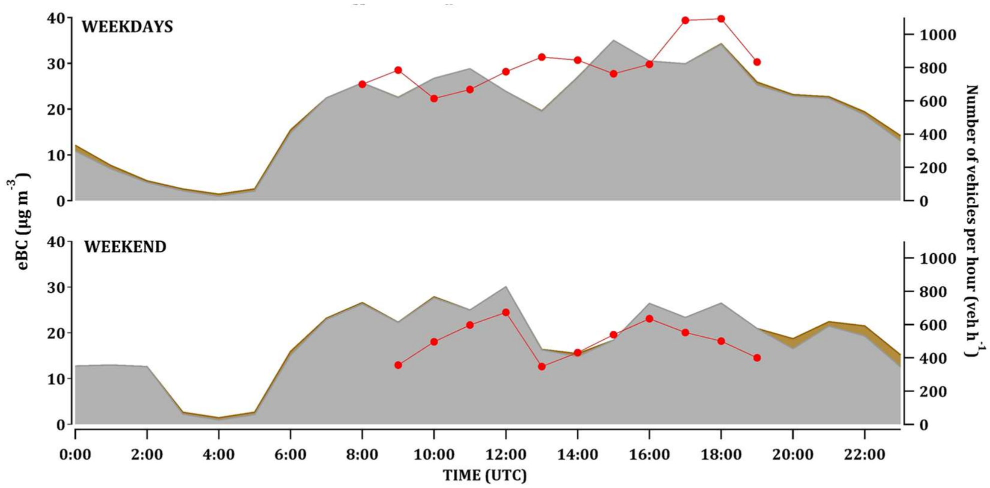

Fig. 3. Evolution of eBC_{ff}, eBC_{bb} concentration (µg m^{-3}) and number of vehicles per hour throughout the day along weekdays and weekend.

3.2. Absorption Ångström Exponent

During daytime hours (8:00–20:00 h, local time), a mean Absorption Ångström Exponent ($AAE_{470\text{-}950}$) value of 0.93 ± 0.04 was obtained, ranging from a minimum value of 0.81 and a maximum of 1.07 (Fig. 4). At night, when the traffic density was low, a maximum AAE of 1.47 was recorded. Taking into account nocturnal data (without traffic counts), the mean value of $AAE_{470\text{-}950}$ was 0.97 ± 0.10 (Table 1). The highest concentrations of eBC corresponded to $AAE_{470\text{-}950}$ values between 0.85 and 0.95. In general, values greater than 1.0 occurred at night-time, with eBC values less than 20 µg m^{-3}. These low values may be due to the low dispersion of pollutants inside the tunnel, a semi-enclosed place. AAE values estimated in the tunnel, where practically pure traffic emissions are overwhelming, are similar to those documented in other studies, such as Zotter et al. (2016), who obtained an AAE_{ff} of 0.9 from ^{14}C measurements of EC fractions on filter samples in Switzerland. Likewise, values in the tunnel are similar to those measured for outdoor traffic in León (Spain) (Blanco-Alegre et al., 2018) and in the range of values (0.6-0.9) reported for traffic events in New Delhi (India) (Garg et al., 2016). It has to be emphasized that because of the low AAE values obtained, the mineral dust interference in results is minimal (Petzold et al., 2013). Furthermore, $AAE_{370\text{-}950}$ and $AAE_{470\text{-}950}$ values did not present statistically significant differences each other ($r=0.97$; $p<0.001$). In Figure S1, BC has been plotted as a function of attenuation BC(ATN) for the wavelengths of 470 and 950 nm) with slope values close to 0, indicating no dependence on ATN (Drinovec et al., 2015). Figure S2 provides complementary information to Figure 4, showing the great variability of AAE values at night, when the traffic volume is much lower.

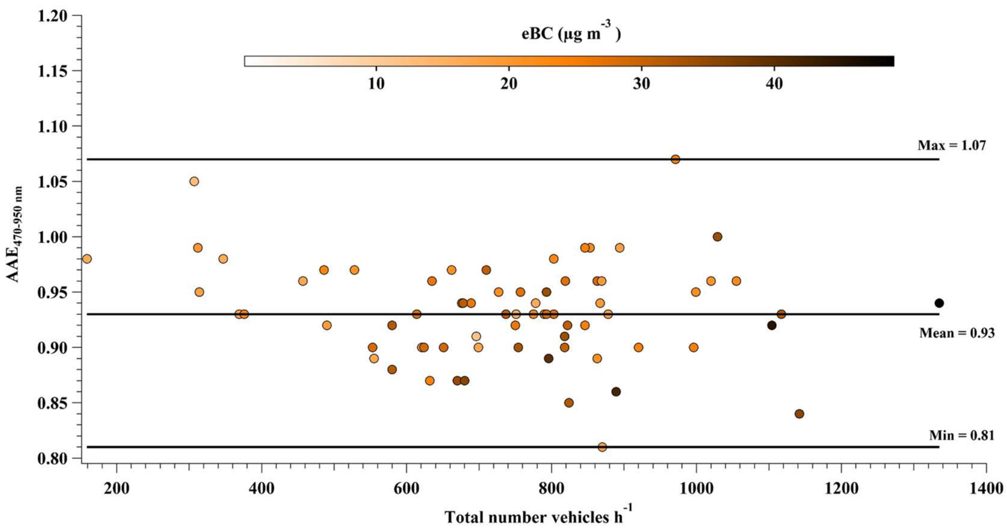

Fig. 4. Absorption Ångström exponent ($AAE_{470-950}$) vs total vehicles per hour in the tunnel during daytime hours (8:00–20:00 h, local time).

3.3. Vehicles-eBC relationship

Figure 5 shows the linear regression between eBC concentration and the number of different types of vehicles inside the tunnel: light vehicles (a), trucks (b), heavy diesel vehicles (c) and total number of vehicles (d). Almost all the fleet in circulation (94 %) consisted of light-duty vehicles. The contribution of light automobiles was higher than that of heavy diesel vehicles and trucks, thus, results of (a) and (d) were similar. There was a positive correlation, statistically significant, between eBC and light vehicles ($r=0.48$; $p<0.001$) and between eBC and the total number of vehicles ($r=0.47$; $p<0.001$). Thus, these results highlight the clear relationship between a high traffic density and eBC concentration. However, there were not statistically significant correlations between trucks and heavy diesel vehicles and eBC, probably due to their low number. Besides, another factor that can affect the dispersion of values can be the turbulence of the air promoted by the number of vehicles in circulation and high speeds (Kristensson et al., 2004).

In Figure 5, the slope was higher for trucks and heavy diesel vehicles (0.169 and 0.153 µg m^{-3} $vehicle^{-1}$, respectively) than for light vehicles (0.016 µg m^{-3} $vehicle^{-1}$). Thus, a small number of trucks and heavy diesel vehicles cause a high concentration of BC. From the intercept values, it can be seen that trucks and heavy vehicles contribute to a higher concentration of BC than light vehicles (23.42 vs. 13.68 µg m^{-3}).

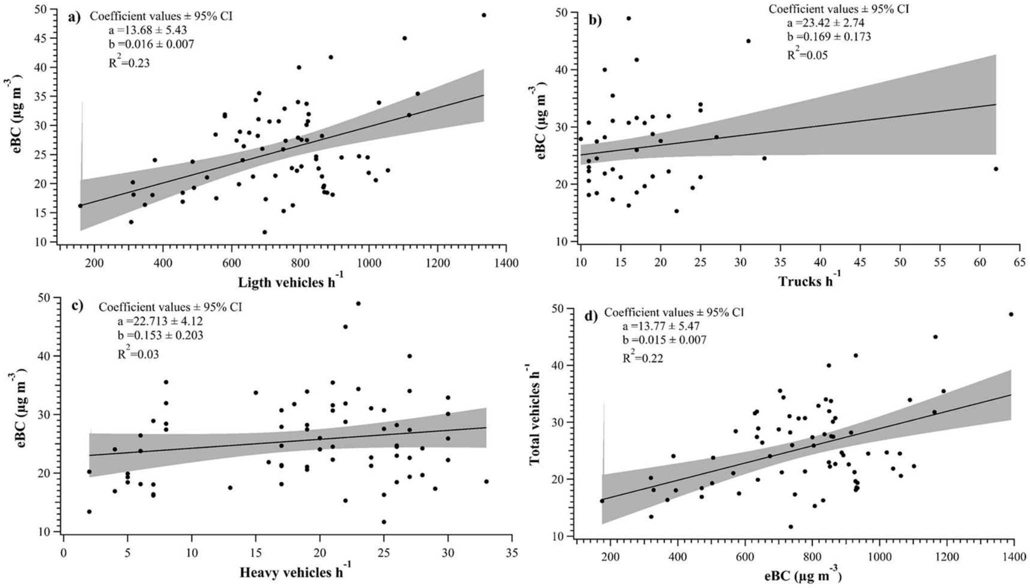

Fig. 5. Linear regression and confidence bands (shaded) with 95% significance level between eBC (µg m^{-3}) and a) number of light vehicles per hour; b) number of trucks per hour; c) number of heavy diesel vehicles per hour; d) total number of vehicles per hour. The parameter *a* is the intercept and *b* is the slope.

3.4. Gaseous emissions-eBC relationship

Correlations between eBC concentrations and gaseous compounds are depicted in Fig. 6. According to Alves et al. (2015b), the mean emission factors (g veh^{-1} km^{-1}) of these gaseous pollutants were: EF_{CO2} (212 ± 18.2), EF_{CO} (4.09 ± 2.52), EF_{NO} (0.61 ± 0.14) and EF_{NO2} (0.29 ± 0.07), similar to other studies in tunnels in Brazil (Martins et al., 2006) and Sweden (Kristensson et al., 2004). On average, the concentrations of CO, CO_2, NO and NO_2 were 20, 1.6, 53 and 43 higher in the tunnel than at the urban background, respectively (Alves et al., 2015b). Statistically significant positive correlations between eBC and all gaseous pollutants were found: CO (r=0.67; p<0.001), CO_2 (r=0.71; p<0.001), NO (r=0.63; p<0.001) and NO_2 (r=0.70; p<0.001). The sum of NOx, which reflects the primary emission more than the individual oxides, presented a significant positive correlation with eBC concentration (r=0.66; p<0.001). Similar CO-eBC results were obtained by Latha and Badarinath (2004) at an urban site. However, the eBC-NO_2 relationship does not follow the one described by Wang et al. (2012), who determined on-road diesel vehicle emission factors for nitrogen oxides and black carbon in two Chinese cities, observing that high eBC emission trucks are usually not high NOx emission sources and vice-versa. Their results suggested that, while a small number of high BC emission trucks contribute disproportionally to the total BC emissions, high NOx emission trucks do not dominate the total NOx emissions.

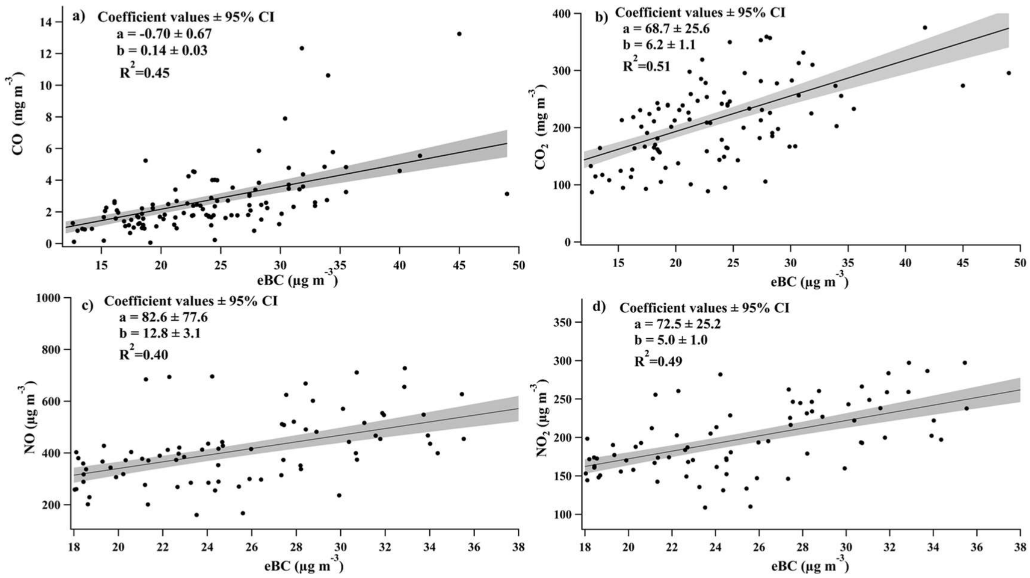

Fig. 6. Linear regression and confidence bands (shaded) with 95% significance level between eBC concentrations and levels of: a) CO b) CO_2 c) NO and d) NO_2.

3.5. Emission factors

The mean EF_{BC} estimated inside the tunnel was 0.31 ± 0.08 g (kg fuel)$^{-1}$. The EFs obtained in the present work are in the range of values reported by other studies (Ban-Weiss et al., 2009; Dallmann et al., 2013, 2012; Gëller et al., 2005; Grieshop et al., 2006; Jezek et al., 2015; Miguel et al., 1998; Strawa et al., 2010) (Table 3). Emission factors depend on traffic intensity, emission category (Euro standards), driving modes (idle, low- and high-speed acceleration, low- and high-speed cruise), vehicle age, load, fuel type, installed emission control technologies, as well as on external factors, such as local mixing and meteorology (Grieshop et al., 2006; Park et al., 2011; Wang et al., 2018). In the morning (0800-1300 UTC), a mean value of 0.32 ± 0.07 g (kg fuel)$^{-1}$ has been estimated, while in the afternoon (1400-2100 UTC) the EF_{BC} was 0.30 ± 0.08 g (kg fuel)$^{-1}$. During the sampling campaign, a maximum EF_{BC} of 0.48 g (kg fuel)$^{-1}$ (on Tuesday at 1800 UTC) and a minimum of 0.20 g (kg fuel)$^{-1}$ (on Wednesday at 0900 UTC) were registered. When these EFs were converted into mass emitted per km and vehicle, EF_{BC} presented a mean value of 0.11 ± 0.08 mg veh^{-1} km^{-1}, ranging from 0.05 mg veh^{-1} km^{-1} to 0.52 mg veh^{-1} km^{-1}. These values are lower than those reported for road tunnels in São Paulo, Brazil (Sánchez-Coyllo et al., 2009). However, it should be borne in mind that, in this latter work, BC was estimated thought reflectance analysis of filters.

During the mornings, a mean value of 0.13 ± 0.10 mg veh^{-1} km^{-1} has been estimated in our study, while during the afternoons the value was 0.10 ± 0.05 mg veh^{-1} km^{-1}. For weekdays, a mean value of 0.08 ± 0.02 mg veh^{-1} km^{-1} has been estimated, whilst weekend days presented a higher value of 0.19 ± 0.10 mg veh^{-1} km^{-1}.

Table 3. Mean BC emission factors measured in this study compared with other field measurements.

Study	City	Study type	EF_{BC} (g (kg fuel)$^{-1}$)	Vehicle type
Grieshop et al. (2006)	Pittsburgh (USA)	Tunnel	0.03	Light duty gasoline and diesel vehicles
Strawa et al. (2010)	Oakland (USA)	Tunnel	0.022	Light duty gasoline and diesel vehicles
Miguel et al. (1998)	Oakland (USA)	Tunnel	0.03	Light duty gasoline and diesel vehicles
Miguel et al. (1998)	Oakland (USA)	Tunnel	1.44	Heavy duty diesel
Ban-Weiss et al. (2009)	Oakland (USA)	Tunnel	1.7	Light duty gasoline and diesel vehicles
Gëller et al. (2005)	California (USA)	Tunnel	0.02	Light duty gasoline and diesel vehicles
Park et al. (2011)	Wilmington (USA)	Mobile platform in a tunnel	0.09	Light duty gasoline vehicle
Dallmann et al. (2012)	Oakland (USA)	Tunnel	0.54	Heavy-duty diesel trucks
Dallmann et al. (2013)	Oakland (USA)	Tunnel	0.10	Roadside measurement
Jezek et al. (2015)	Slovenia	Chasing	0.28	Petrol cars
Jezek et al. (2015)	Slovenia	Chasing	0.64	Light duty gasoline and diesel vehicles
Brimblecombe et al. (2015)	Hong Kong	Mobile platform in a tunnel	1.28	Diesel fleet
This study	Braga (Portugal)	Tunnel	0.31 ± 0.08	Light and heavy-duty gasoline and diesel vehicles

4. CONCLUSIONS

The following main conclusions could be extracted from this sampling campaign, carried out continuously for 7 days in a road tunnel in Braga (Portugal) with an aethalometer:

- Inside the tunnel, the mean eBC mass concentration was 21 ± 10 µg m^{-3}, close to the limit proposed by the WHO for daily $PM_{2.5}$ concentrations (25 µg m^{-3}). eBC concentrations reached an hourly maximum of 49.0 µg m^{-3}.
- The maximum values reached during weekdays occurred between 1400 and 1900 UTC, during the rush hours, when traffic density is greater due to commuting.

However, during the weekend days, a peak between 0900 and 1200 UTC was observed, probably due to leisure and shopping activities.

- A mean Absorption Ångström Exponent ($AAE_{470\text{-}950}$) of 0.97 ± 0.10 was obtained, with a maximum of 1.07 during daytime hours, for a source of practically pure traffic.
- There was a positive correlation, statistically significant, between eBC and light vehicles ($r=0.48$; $p<0.001$) and between eBC and total number of vehicles ($r=0.47$; $p<0.001$).
- There was a statistically significant positive correlation between eBC and gaseous emissions (CO, CO_2, NO and NO_2).
- The mean eBC emission factor, EF_{BC} inside the tunnel was 0.31 ± 0.08 g (kg fuel)$^{-1}$. When this EF was converted into mass emitted per km, EF_{BC} presented a mean value of 0.11 ± 0.08 mg veh^{-1} km^{-1}.

The study of black carbon in a road tunnel contributes to better characterize emissions of this pollutant from traffic in real circulation conditions and without influence from other sources providing valuable information on BC emission factors, which are useful as input data to climate and air quality models, as well as to updated emission inventories. Furthermore, the quantification of BC is essential to assess air quality in road tunnels and, thus, improve ventilation systems.

ACKNOWLEDGEMENTS

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Aethalometer measurements in a road tunnel: a step forward in the characterization of black carbon emissions from traffic

C. Blanco-Alegre [1], A.I. Calvo [1], C. Alves[2], P. Fialho[3], T. Nunes[2], J. Gomes[2], A. Castro [1], F. Oduber [1], E. Coz [4] and R. Fraile[1]

[(1)] Department of Physics, IMARENAB University of León, 24071 León, Spain

+34 987291543, rfral@unileon.es

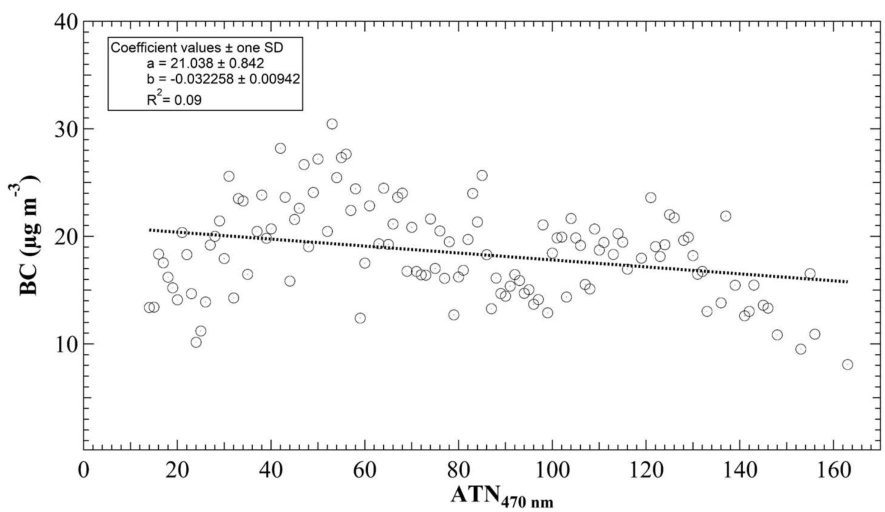
[(2)] Centre for Environment and Marine Studies, Department of Physics, University of Aveiro, 3810-193 Aveiro, Portugal

[(3)] Research Institute of Volcanology and Evaluation – IVAR, Rua da Mãe de Deus, 9500-321 Ponta Delgada, Portugal

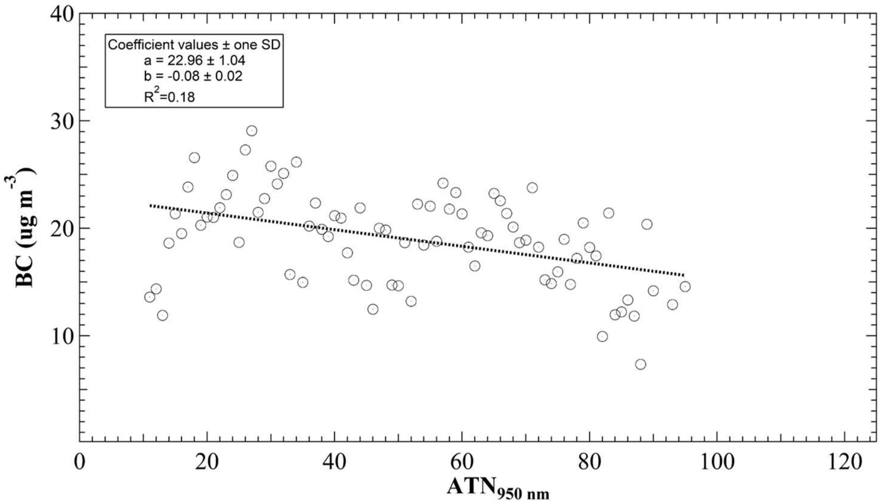
[(4)] Centre for Energy, Environment and Technology Research (CIEMAT), Department of the Environment, Madrid, Spain

SUPPLEMENTARY MATERIAL

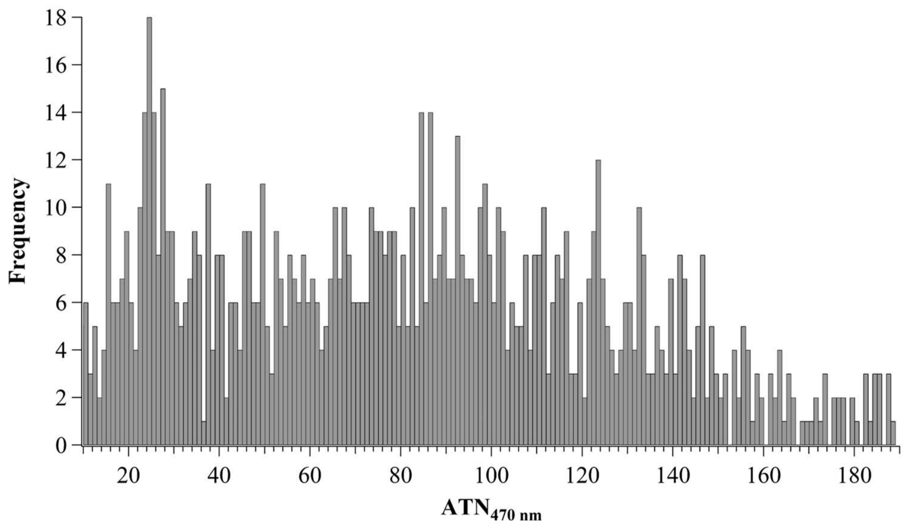
a)



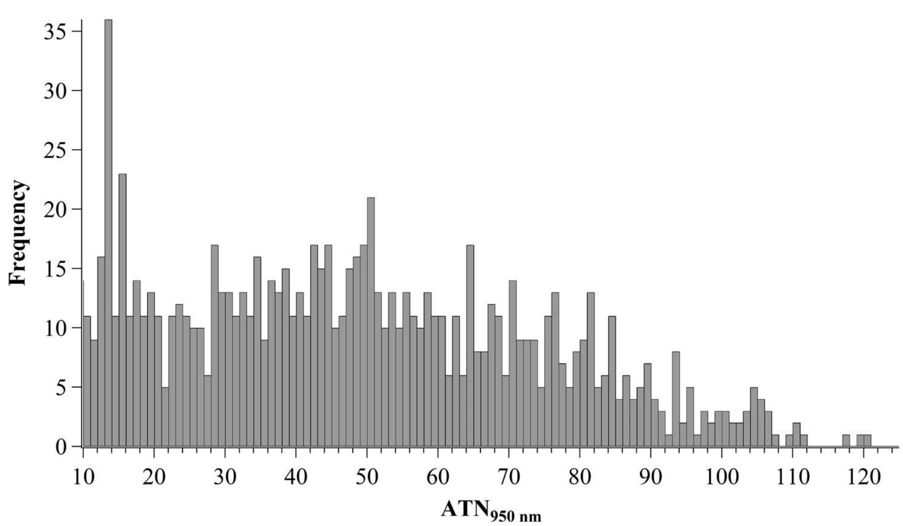
b)


Coefficient values ± one SD
a = 22.96 ± 1.04
b = -0.08 ± 0.02
R^2=0.18
BC (ug m^{-3})
$ATN_{950\ nm}$

c)


Frequency
$ATN_{470\ nm}$

d)


Frequency
$ATN_{950\ nm}$

e)

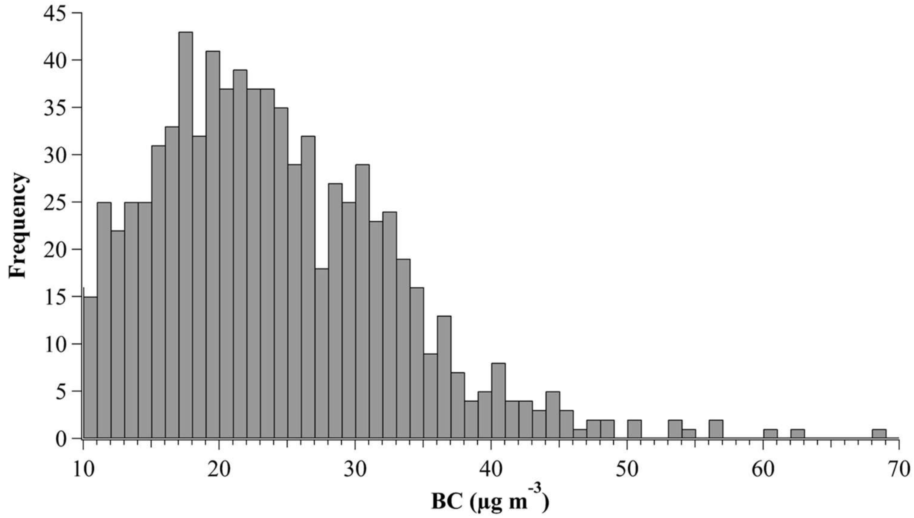
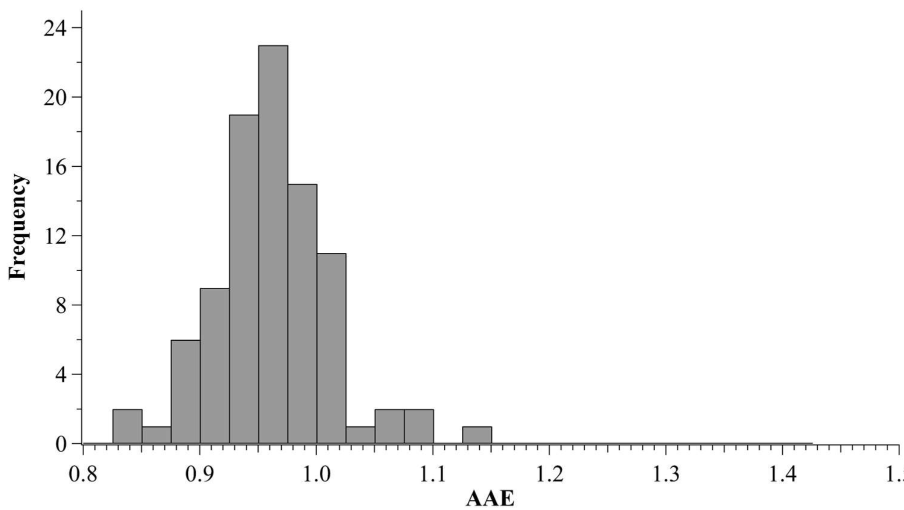
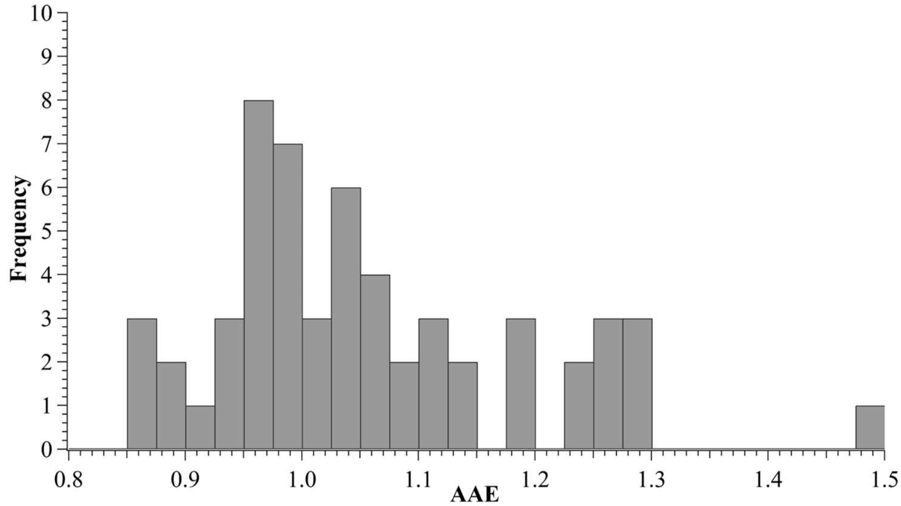


Fig. S1. BC as a function of a) $ATN_{470\ nm}$ and b) $ATN_{950\ nm}$ plot (only considers the average values for the 1 ATN bin with at least 4 values to average); c) Frequency distribution of the number of measurements per $ATN_{470\ nm}$; c) Frequency distribution of the number of measurements per $ATN_{950\ nm}$; e) The BC frequency in the ATN range of 10-125.

a)



b)



c)

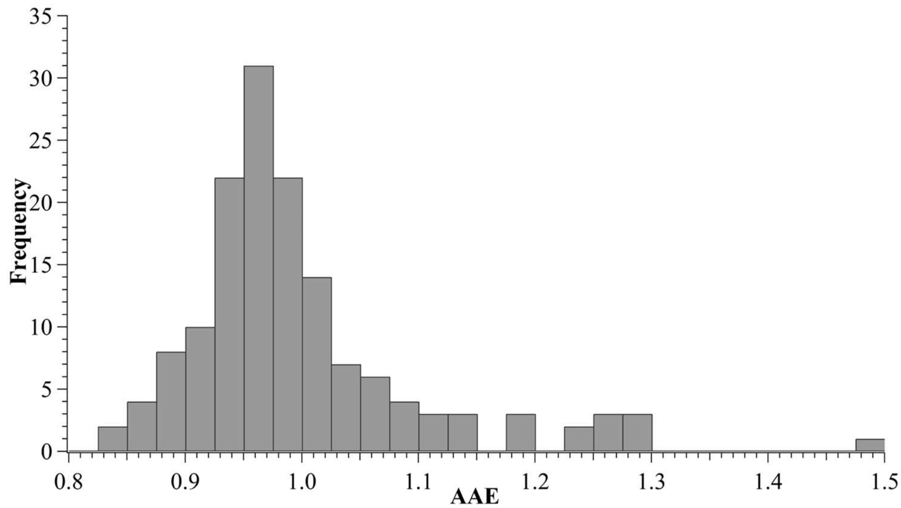


Fig. S2. Frequency distribution of hourly AAE for daytime (a), nighttime(b) and the whole campaign (c).