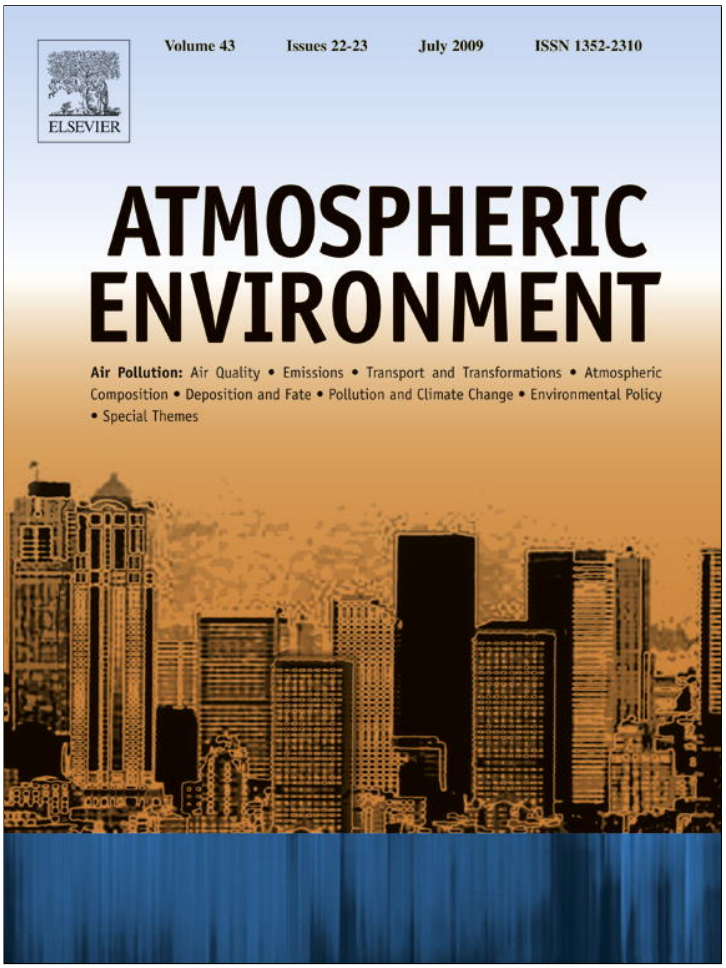

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Short communication

Evidence for a significant contribution of wood burning aerosols to $PM_{2.5}$ during the winter season in Paris, France

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ABSTRACT

Although particulate emissions from residential wood burning have become a subject of great scientific concern for a few years, data related to their impact on the air quality of large European urban centres are still missing. In the present study, we investigated the chemical and optical properties of fine ($PM_{2.5}$) carbonaceous aerosols in Paris during the 2005 winter season in order to track the presence of wood burning emissions in such a large city. The use of a seven wavelength Aethalometer allowed us to document shortwave light absorption by brown-carbon-containing organic aerosols of biomass burning origin. In particular, a well-marked diurnal pattern of the spectral dependence of light absorption, with maxima during the night, could be observed every day of the campaign and attributed to wood burning emissions. Relatively high absorption Ångstrom exponents and WSOC/OC ratios (respectively 1.25 and 0.35 on average for the period of study) also indicated the importance of biomass burning aerosols in the Paris atmosphere in winter. Finally, a rough estimate of the contribution of wood burning carbonaceous aerosols to $PM_{2.5}$ could be achieved. This contribution was found to be as high as 20 ± 10% on average at the Paris background site investigated here.

1. Introduction

Carbonaceous aerosols originating from biomass burning are known to contain high amounts of carcinogenic polycyclic aromatic hydrocarbons (e.g. Lewtas, 2007) as well as light-absorbing species (black and brown carbon) which significantly influence the aerosol radiative forcing and the atmospheric photochemistry (e.g. Andreae and Gelencsér, 2006). It is thus of prime importance to evaluate the contribution of wood burning aerosols to particulate matter in the ambient air. A growing number of studies have been demonstrating the importance of this contribution in industrialized countries during the winter season (see for instance Puxbaum et al., 2007, and references therein). However, data are still missing for large European urban centres. In this study, we investigated the impact of residential wood burning on Paris air pollution in winter, using multi-wavelength Aethalometer absorption measurements.

Light absorption by aerosols is usually parameterized as proportional to $\lambda^{-\alpha}$, where λ is the light wavelength and α stands for the absorption Ångstrom exponent. While the spectral dependence of black carbon light absorption is demonstrated to be relatively low ($\alpha \sim 1$), other aerosol components, i.e. hematite and brown carbon, exhibit a much higher absorption exponent (see for instance Kirchstetter et al., 2004; Andreae and Gelencsér, 2006; Hadley et al., 2008). Based on these optical properties, a few studies have been using multi-wavelength Aethalometer measurements to detect the presence of biomass burning aerosols in ambient air (e.g. Sandradewi et al., 2008a, and references therein). In a more quantitative way, Sandradewi et al. (2008b) recently proposed a source apportionment model allowing the quantification of biomass burning carbonaceous aerosols in environments where carbonaceous material is primarily made of wood burning and non-brown-carbon-containing combustion aerosols. We argue here that it is the case for the fine aerosol fraction in Paris at wintertime, so that this model could be applied to our dataset.

2. Aerosol sampling

Results presented here were obtained from 10 January to 21 February 2005 on the terraced roof of the Laboratoire d'Hygiène de la Ville de Paris (Paris, 13th district). This site, detailed by Favez et al. (2007), corresponds to a station of the AIRPARIF air quality

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monitoring network and is representative of Paris background air pollution (www.airparif.fr). Mass concentrations of the fine aerosol fraction ($PM_{2.5}$) were obtained every minute using an optical particle counter (OPC, GRIMM Aerosol Technik, model 1.108) and applying a "field mass calibration" fully described in Sciare et al. (2007). The accuracy of this methodology was checked via an intercomparison exercise with $PM_{2.5}$ measurements from a collocated Tapered Element Oscillating Microbalance (TEOM, Rupprecht & Patashnik, model 1400a, equipped with a Filter Dynamic Measurement System, series 8500), showing a good correlation between both measurements (slope = 0.98 and $r = 0.96$, for hourly-averaged data). In order to investigate carbonaceous aerosols, filter samples were collected using a Stack Filter Units (SFU), consisting of a 47 mm-diameter Whatman QMA filter mounted downstream of an 8 μm Nuclepore filter. At the flow-rate used in this study (1 m^3 h^{-1}), the 50% cut-point diameter of the 8 μm filter is approximately 2.5 μm. Filter samples were collected on a 24 h base during weekdays (20 sampling periods) and on a 72 h base during week-ends (5 sampling periods). Finally, aerosol absorption coefficients were obtained on a 5 min base at seven different wavelengths (370, 470, 520, 590, 660, 880 and 950 nm) using a Magee Scientific Aethalometer (model AE-31) equipped with a $PM_{2.5}$ cut-off inlet. This instrument was operating at a flow rate of 5 l min^{-1} in an automated mode, under which the filter tape advanced when the attenuation at 370 nm reached 75. For comparison purpose, results obtained from Aethalometer measurements during a summer field campaign (28 July to 8 September 2005) are also presented in this paper. These measurements were performed at the same site, using the same instrument, and results were obtained following the same data treatment procedure.

3. Corrections of aethalometer measurements

Due to the methodology used within the Aethalometer (filter-based measurement), absorption coefficients directly obtained from this instrument are affected by various sampling and analytical artefacts, mostly referred as multiple scattering and shadowing effects (e.g. Weingartner et al., 2003). In order to correct these artefacts, the procedure introduced by Weingartner et al. (2003) was applied to our dataset following:

$$b_{\mathrm{abs},\lambda,t} = \frac{b_{\mathrm{aeth},\lambda,t}}{2.14 \times R(\mathrm{ATN})_{\lambda,t}} \quad (1)$$

where, at a given time (t) and a given wavelength (λ), $b_{\mathrm{abs},\lambda,t}$ and $b_{\mathrm{aeth},\lambda,t}$ correspond to the corrected absorption coefficient and the raw absorption coefficient respectively. The constant 2.14 stands for multiple scattering of the light beam at the filter fibres in the unloaded filter. Finally, $R(\mathrm{ATN})_{\lambda,t}$ describes the decrease of the latter artefact with the gradual accumulation of particles on/in the filter (i.e. correction of the shadowing effect). $R(\mathrm{ATN})_{\lambda,t}$ was determined following the equation:

$$R(\mathrm{ATN})_{\lambda,t} = \left(\frac{1}{f_\lambda} - 1\right) \times \frac{\ln\left(\mathrm{ATN}_{\lambda,t}\right) - \ln(10)}{\ln(50) - \ln(10)} + 1 \quad (2)$$

where $\mathrm{ATN}_{\lambda,t}$ corresponds to the light attenuation measured by the aethalometer at a given time and a given wavelength, and f_λ allows the correction of the instrumental error that occurs when the shadowing effect is disregarded. As no measurement of light scattering coefficients were available for this study, it was not possible to calculate f_λ as a function of the single scattering albedo. Alternatively, based on Sandradewi et al. (2008a) and Yang et al. (2008), this free parameter was determined by minimising the difference between the ratio of absorption coefficients and the ratio of aerosol mass concentrations ($PM_{2.5}$ calculated from OPC measurements) before and after filter tape advancements. The overall uncertainty of absorption coefficients calculated this way is of the order of 20%. However, this uncertainty is expected to affect measurements at each wavelength in a relatively similar way, so that a highest confidence level is assumed for the spectral shape of light absorption.

4. Carbon analysis

Particulate elemental carbon (EC) and organic carbon (OC) were determined using a thermo-optical carbon analyser (EC-OC Sunset Lab. Instrument) working in transmittance and implemented with the IMPROVE thermal program, as described in Sciare et al. (2003). Prior to analysis, carbonates were removed under HCl fumes, and positive sampling artefacts were minimised by pre-heating filter samples at 60 °C during 20 min. The detection limit and the analysis uncertainty of the EC-OC Sunset Lab. Instrument are of 0.2 μgC and 5%, respectively. OC particles were also discriminated between their water soluble and water insoluble fractions (WSOC and WIOC). WSOC analyses were conducted using a Total Organic Carbon analyser (Sievers TOC 900) and following the procedure fully detailed by Favez et al. (2008). Briefly, extraction of the soluble fraction was conducted in ultra pure water and under soft shaking (~16 h). Prior to analysis, the extract solution was filtered through Teflon filters in order to remove any suspended particle. WIOC was calculated as the difference between OC and WSOC. The overall uncertainty in determination of WSOC and WIOC is estimated to be of the order of 10%. Finally, water-insoluble/water-soluble organic matter (WIOM and WSOM respectively) were obtained applying specific WIOC-to-WIOM and WSOC-to-WSOM conversion factors accounting for non-carbon atoms present in the organic matter. Based on the works of Turpin and Lim (2001) and Zhang et al. (2005), these conversion factors were assumed to be of 1.4 and 2.1 respectively.

Interestingly, a mean WSOC/OC ratio of 0.35 ± 0.05 was obtained for this study, corresponding to the upper range of values previously reported for wintertime WSOC/OC ratios in urban environment (e.g. Miyazaki et al., 2006; Viana et al., 2007). Biomass burning aerosols are known to be particularly rich in water-soluble organics, and relatively high WSOC/OC ratios observed here are first indicators of the significant influence of residential wood burning emissions in Paris during the winter season.

5. Spectral dependence of light absorption

The mean spectral shape of light absorption obtained in this study is compared in Fig. 1 to that obtained during a summer field campaign performed at the same site. As shown in this figure, a mean $\alpha_{370-950\mathrm{nm}}$ (i.e. the absorption exponent calculated using the seven Aethalometer wavelengths) of 1.25 ± 0.11 was obtained in winter, which is significantly higher than the mean $\alpha_{370-950\mathrm{nm}}$ calculated for the summer campaign (1.07 ± 0.04). No significant change in the aerosol size distribution was observed between the winter and summer experiments (Cachier et al., 2009), so that this seasonal trend is rather related to a change in the aerosol chemical composition and the presence of other light absorbers than black carbon, such as brown carbon and/or iron oxides, has to be envisaged for the winter season. The influence of iron oxides is expected to be very limited here, notably due to the location of the study (i.e. far away from iron oxide sources). The impact of organic aerosols on the spectral dependence of light absorption is further confirmed by the good correlation obtained between OC/EC ratios and absorption exponents ($r = 0.96$), as shown in Fig. 2.

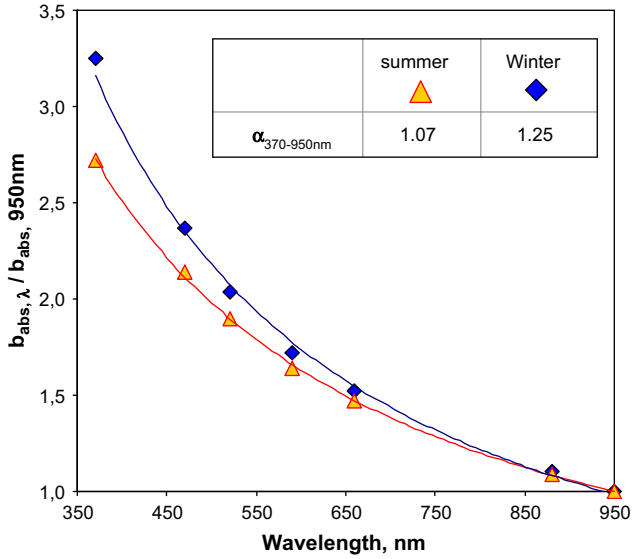

Fig. 1. Mean spectral dependence of light absorption during the winter and summer field campaigns. For comparison purpose, the absorption coefficient at each Aethalometer wavelength was normalized to the absorption coefficient at 950 nm. Triangles: data obtained for the summer campaign; diamonds: data obtained for the winter campaign. Solid lines correspond to power law fits and $\alpha_{370-950nm}$ corresponds to the absorption Ångstrom exponents obtained from these power law fits. Assuming the summer spectral shape as background conditions, the difference between the two solid lines is assessed to correspond to wintertime light absorption by wood burning organic aerosols.

A striking feature arising from this study is the well-marked wintertime diurnal pattern displayed by the absorption exponent. This diurnal pattern is compared in Fig. 3 to that obtained for the summer experiment. For both seasons, minimum values were obtained during traffic rush-hours (i.e. 7:00–900 and 17:00–19:00), which can be explained by the low spectral dependence of traffic emissions (e.g. Andreae and Gelencsér, 2006). During summer, this diurnal pattern displayed limited amplitude (from 1.02 ± 0.04 during the morning traffic peak to 1.08 ± 0.04 during night/afternoon periods). Conversely, important variations were observed in winter, with a mean absorption exponent of 1.33 ± 0.10 during the night. Similar winter/summer diurnal patterns were observed by Sandradewi et al. (2008a) in an Alpine village. It should be noted that the well-marked wintertime diurnal pattern was observed every day during the campaign (Fig. 4), which seems to indicate

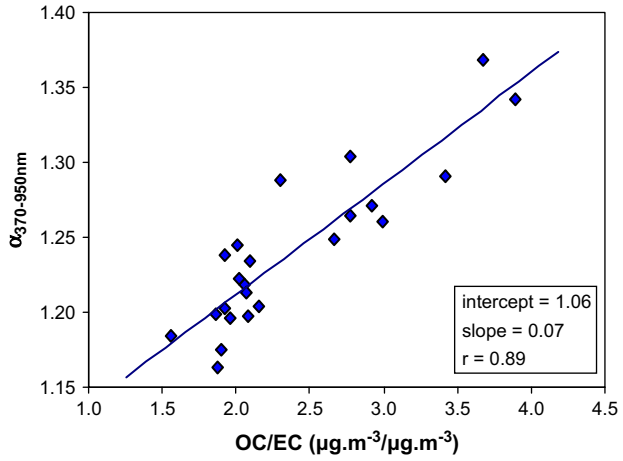

Fig. 2. Scatter plot of absorption Ångstrom exponents (averaged over the duration of filter samplings) vs. OC/EC ratios during the winter field campaign.

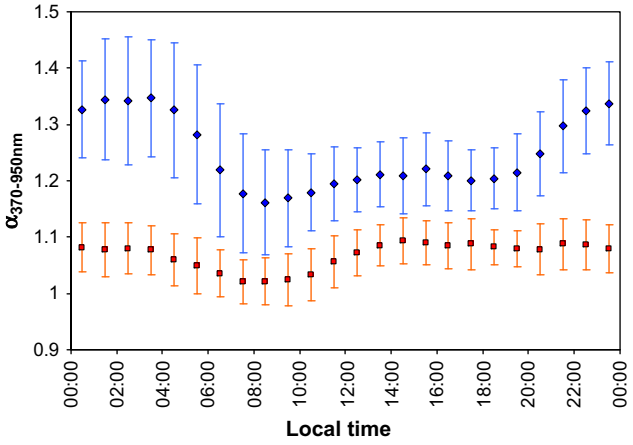

Fig. 3. Mean diurnal pattern of the absorption Ångstrom exponent ($\alpha_{370-950nm}$) during the winter and summer field campaigns (mean value ± standard deviation).

a predominant influence of local sources rather than of long-range transport. In order to further investigate the impact of long-range transport, we computed absorption exponents as a function of wind directions. Results indicated only a slight increase of the absorption exponent for air masses originating from the continental sector (1.27 ± 0.13 against 1.24 ± 0.11 for the oceanic sector).

All these results are indicative of significant emissions of brown-carbon-containing aerosols in Greater Paris (Paris city + suburbs) during winter, and especially during the night. It appears reasonable to consider that these emissions are related to residential heating, which is supported by the fact that highest absorption exponents were observed during week-ends (Fig. 4). Possible candidates for these brown-carbon-containing emissions are coal burning and wood burning. Coal is almost not used any longer in France, while wood burning for heating purpose is still a very common practice. Results presented here are thus assessed to be primarily due to residential wood burning.

Interestingly, the Angstrom absorption exponent seemed to globally increase as ambient air temperature decreased ($r = -0.62$ for the correlation between 24-h averaged $\alpha_{370-950}$ and temperature). This phenomenon could be explained by an increase of residential heating during coldest periods. It could also be partially related to a more efficient condensation of semi-volatile polycyclic aromatic hydrocarbons (from wood burning origin) as temperature decreased. The impact of such a mechanism on particulate air pollution in Paris has still to be investigated.

6. Contribution of wood burning aerosols to particulate matter

We present here an attempt to evaluate the contribution of wood burning carbonaceous material (CM_{wb}) to total $PM_{2.5}$ concentrations during the winter experiment. This source apportionment exercise is based on the work of Sandradewi et al. (2008b). As proposed by the latter study, total carbonaceous material (CM_{total}) comprised in the fine aerosol fraction in winter could be primarily considered as the sum of brown-carbon-containing carbonaceous material (i.e. CM_{wb} here) and of non-brown-carbon-containing carbonaceous material originating from fossil fuel combustion (CM_{ff}) following:

$$CM_{total} = CM_{ff} + CM_{wb} = C_1 \times b_{abs,ff,950nm} + C_2 \times b_{abs,wb,470nm} \quad (4)$$

where C_1 and C_2 relate the light absorption to the particulate mass of both sources, $b_{abs,ff,950nm}$ represents the absorption coefficient of

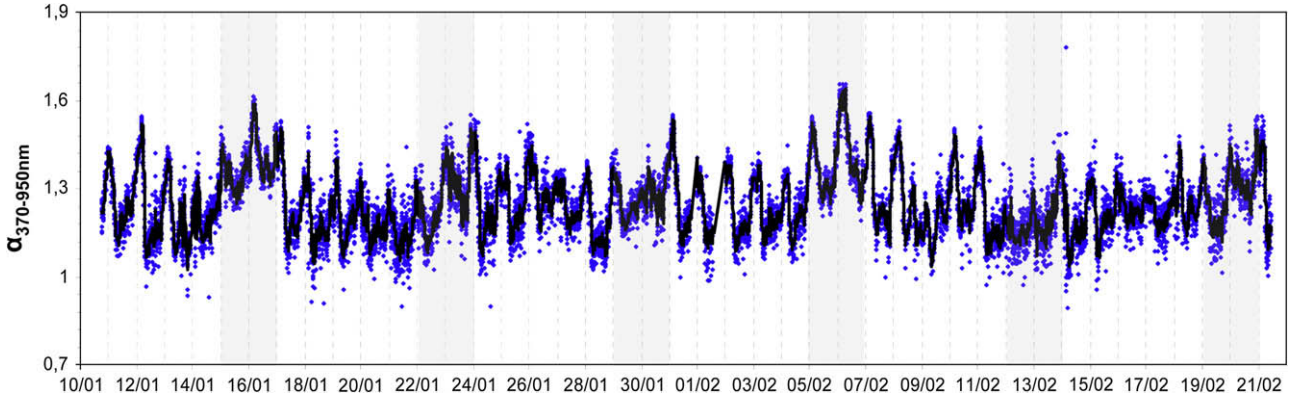

Fig. 4. Time series of the absorption Ångstrom exponent ($\alpha_{370-950nm}$) during the winter field campaigns. Coloured bands correspond to week-ends (15–16 January, 22–23 January).

CM_{ff} at 950 nm and $b_{abs,wb,470nm}$ represents the absorption coefficient of CM_{wb} at 470 nm. In this calculation, the contribution of primary and secondary biogenic organic aerosols to CM_{total} is assumed to be negligible, which appears reasonable during the winter season and within the fine aerosol fraction investigated in the present study. Anthropogenic secondary organic aerosols, and especially those formed from biomass burning gaseous emissions, are assessed to be partly taken into account in this Aethalometer model, as suggested by Sandradewi et al. (2008c). It should also be noted that CM_{ff} is assumed here to comprise traffic emissions as well as carbonaceous aerosols originating from fuel oil and natural gas combustion. Equation (4) can be solved when combined with the following ones:

$$CM_{total} = EC + WIOM + WSOM \quad (5)$$

$$b_{abs,\lambda} = b_{abs,ff,\lambda} + b_{abs,wb,\lambda} \quad (6)$$

$$\frac{b_{abs,ff,470nm}}{b_{abs,ff,950nm}} = \left(\frac{470}{950}\right)^{-\alpha_{ff}} \quad (7)$$

$$\frac{b_{abs,wb,470\ nm}}{b_{abs,wb,950\ nm}} = \left(\frac{470}{950}\right)^{-\alpha_{wb}} \quad (8)$$

where α_{ff} and α_{wb} represent the Angstrom absorption exponents of both aerosol sources. In these calculations, α_{ff} was set at 1.1, based on the mean absorption exponent ($\alpha_{470-950nm}$) obtained for the summer campaign. The absorption exponent of CM_{wb} (α_{wb}) was roughly estimated to be 2, based on values previously reported for wood burning aerosols (e.g. Sandradewi et al., 2008b; Lewis et al., 2008; and references therein). Via the propagation of errors, mostly related to the correction of absorption coefficients and to the assumption of negligible secondary organic aerosol contents, the overall uncertainty of this calculation is estimated here to about 50%. However, values obtained for C_1 and C_2 (264 895 and 724 256 respectively) are very close to those proposed by Sandradewi et al. (258 831 and ~630 000 respectively), which seems to validate our calculation.

Results of this calculation indicated a mean contribution of CM_{wb} to total carbonaceous aerosols of 46%. This value is in the same order of magnitude than those previously reported in the few papers focusing on European cities (e.g. Zdráhal et al., 2002; Szidat et al., 2006; Lanz et al., 2008). If considering only filters collected during week-ends, when traffic is limited and residential wood burning is expected to be more important, this contribution was found to be on average of 64%. Related now to the total fine aerosol fraction, carbonaceous aerosols originating from residential wood burning were evaluated to represent about 20% of $PM_{2.5}$ in Paris during the period of study. Due to the number of approximations made for this calculation, a relatively high uncertainty is linked to this estimate (±10%). However, results presented here clearly indicate that the contribution of wood burning aerosols to PM concentrations could definitely not be neglected in Paris at wintertime. For a more precise estimate of this contribution, the use of specific organic tracers (such as levoglucosan) and of real-time measurements of carbonaceous aerosols should be envisaged. Modelling studies are also still needed to investigate the climatic impacts of these wood burning emissions at a regional scale.

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