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Black Carbon in Lorraine: sources, geographical origins and model evaluation

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Abstract

Black carbon is mainly emitted by any combustion process: road traffic, wood and coal burning, shipping and various industrial combustions. Given, among others, the health effects of air pollution, and that BC is a good tracer and its sanitary impacts, monitoring of BC at various scales (remote, rural, urban and traffic) has nowadays become widespread. The present study uses long-term BC measurements in Metz (East of France) to determine the contribution of the two main sources, traffic and wood-burning, and their seasonalities. Wood-burning, in 2015, could represent up to 25% of BC during cold months. In paralell, a comparison with measurements at a rural background site located 80 km South-West of Metz during March 2015 helped to assess the role of background concentrations to urban air quality during an intense haze pollution event. Finally, a model evaluation was performed at both sites and showed an air-mass dependence on the performance of the model.

Keywords: Black Carbon, sources, origin, model

1 Introduction

Outdoor air pollution has been, since 2013, considered as carcinogenic for humans (IARC, 2013). The chemical composition of particulate matter (PM) and its granulometry plays a central role in this assessment (Harrison and Yin, 2000). In particular, the physical and chemical structure of Black Carbon (BC) can lead to human health issues. Its small size increases the penetration of soot deep inside our breathing system, and can be associated to (coated with) organic compounds,

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like Polycyclic Aromatic Hydrocarbons (HAP), potentially intensifying the inflammatory response (Ramgolam et al., 2009). It is for that matter that BC is considered as a good indicator of the impacts of air pollution (Janssen et al., 2011), being closely linked to its sources. BC is emitted from incomplete combustion of fossil fuels and biomass. Thus, in urban areas, where population density enhances the exposure to pollution, BC main sources are traffic and wood-burning. Indeed, Putaud et al. (2004) have highlighted a significant gradient of BC concentrations from rural background to kerbsides. But despite this very local pattern, case studies report examples of long-range advection of BC, observed in the Himalayas (Ming et al., 2007, 2010), or even in urban environments like Paris (Healy et al., 2012), where up to 21% of BC was determined to have a continental origin. At an European scale, Putaud et al. (2004) also observed a geographical heterogeneousness.

More specifically, the North-East of France regularly faces pollution episodes during winter and spring usually associated with continental air masses. Paris being located West from this area of France, and oceanic air masses being mainly occurring (Petit et al., 2015), advection from other French regions may be observed. But poor is known about the contribution of the sources of BC and their different geographical scales. This study focuses on Black carbon measurements in Metz, the second largest urban area in Lorraine. Long-term 7-wavelength aethalometer data are presented at urban background during 2015, to investigate the seasonality and the sources of black carbon. A more specific work has been performed during March 2015, associated with an intense pollution episode: a comparison between urban concentrations was performed with a rural background site (OPE), located around 80 km South-West of Metz, to investigate the potential role of rural background on urban BC levels, and inversely.

Moreover, the precise determination of the impacts of mitigation policies in order to improve air quality is closely linked to the capability to model, through mitigation scenarios, BC concentrations. Since few information about this are available in France, a comparison between predicted and observed BC levels at rural and urban environment is presented here during the pollution episode of March 2015.

2 Sites, instrumentation and methods

For the purpose of this study, two measuring sites have been used (Metz and OPE). The first one is located in Metz (49.11°N, 6.22°E), around 5 km South-East of the city center, and is representative of urban background pollution. This station is part of the air quality monitoring network in Lorraine, operated by Air Lorraine. The second site (OPE) is located around 80 km South-West of Metz (49.56°N, 5.51°E), representative of rural background concentrations. Due to this relative

proximity, these two sites experience similar meteorology, notably in terms of ambient temperatures and relative humidity. Figure 1 shows the comparison of these 2 parameters at both sites during 2015 (15-min averages).

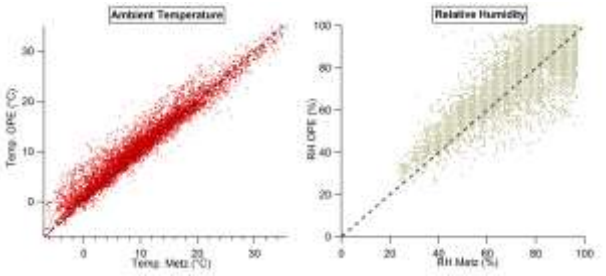
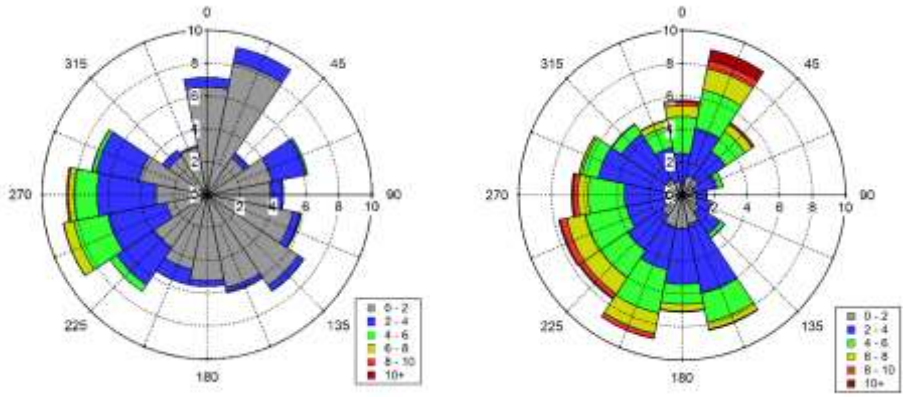


Figure 1. Comparison of ambient temperature and relative humidity between Metz and OPE in 2015 (15-min average)

It is however interesting to note that wind conditions are significantly different (Fig. 2), where OPE seems much more ventilated than Metz, which is mainly associated with calm winds, below 2 m/s. This can be partly associated to the topography of the sites: Metz is located within the valley of the Moselle river and is surrounded by hills, while OPE is on a plateau, cleared from any natural impediment.



a) b)

Figure 2. Wind rose in a) Metz and b) OPE, during 2015 (15-min wind data). Radial axis is the frequency (in %), wind speed classes in m/s.

Both sites were equipped with 7-wavelengths aethalometer instruments, AE31 model in OPE, and AE33 model in Metz. Extensive description of these

instruments can be found elsewhere (Drinovec et al., 2015 and references therein); they also have been found to be very robust over long term periods (Herich et al., 2011; Petit et al., 2015). In this study, BC concentration at 880 nm were used. Fossil-fuel and wood-burning fractions (BC_{ff} and BC_{wb}, respectively) were calculated in Metz following the algorithm proposed by Sandradewi et al. (2008).

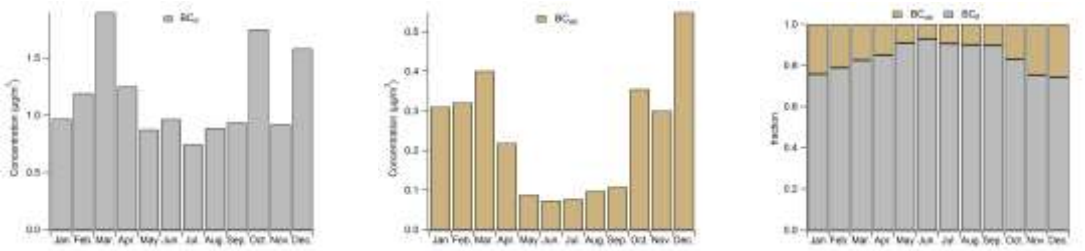
Wind analysis on BC concentrations has been performed at both sites by Non-parametric Wind Regression (NWR, Henry et al., 2009). Calculations were carried out with ZeFir, an Igor© tool for wind and trajectory analyses (Petit et al., submitted). In parallel, backtrajectories arriving hourly at OPE (at 100 m above ground level) were calculated with the PC-based version of Hysplit (Draxler, 1999). Following total spatial variance variations, cluster analysis led to 5 clusters, illustrated in Figure 7.

Black carbon modeled concentrations were determined from the Chemistry-Transport Model (CTM) CHIMERE (Menut et al., 2013), through the regional platform Prev'est. Chemical mechanisms are controlled by MELCHIOR 2 (Lattuati, 1997), thermodynamics by ISORROPIA (Nenes et al., 1998), WRF meteorological fields combined with a regional emission inventory were used. The evaluation of performance of the model was investigated by calculating different statistics: Fraction of predictions within a factor of two (FAC2), Mean bias (MB), Normalised mean bias (NMB), Root mean squared error (RMSE), Pearson correlation coefficient, Coefficient of Efficiency (COE, Legates and McCabe, 2013) and Index of Agreement (IOA, Willmott et al., 2012).

3 Results & Discussion

Figure 3 exposes the monthly averages of BC_{ff}, BC_{wb} and their relative contributions during 2015 at Metz (urban background). BC_{ff} does not exhibit pronounced seasonal variations, despite highest averages being observed in March, October and December, reaching respectively 1.9, 1.8 and 1.6 $\mu g/m^3$, while the base level is around 1 $\mu g/m^3$, observed during summer. On the contrary, BC_{wb} has a strong seasonal pattern, with highest concentrations during winter, late autumn and early spring. Whatever the season (except for BC_{wb} in summer), both fractions have typical, very pronounced diurnal variations (not shown here): BC_{ff} has a bimodal pattern, with intense peaks in the morning and the evening, corresponding to daily commuting. The station is indeed located near main highways and is thus very reactive to traffic primary emissions. BC_{wb} has only one significant increase during the evening, slightly shifted from the evening traffic peak, and corresponds wood-burning emissions for residential heating. During winter, wood burning represents around 25% of total BC concentrations, underlining the role of this source in urban areas, as this has also been shown in

other French cities (e.g. Paris: Bressi et al., 2014; Favez et al., 2009; or Grenoble: Favez et al., 2010) or in Europe (e.g. London: Fuller et al., 2014).



a) b) c)

Figure 3. Monthly averages of BC_{ff} (a), BC_{wb} (b), and their relative contributions (c) in 2015

During March 2015, Metz has experienced one of the most intense pollution episode since many years. Briefly, two distinct sub-episodes occurred: the first one, at the beginning of the month (from March 7th to 11th), was characterized by local/regional signals, dominated by primary emissions. The second sub-episode, between March 19th and 22nd exhibited a more advected pattern, with notably a much higher contribution of secondary pollution (especially ammonium nitrate). An in-depth characterization of this episode, spread over most of the French territory, will be available in Petit et al., in prep. However, a specific focus on BC is made here.

A raw comparison of BC levels in Metz and OPE during March reveals a significant contribution of local emissions, rather than rural background (Fig. 4) . It represents, in median, around 20% of the concentrations measured at urban background. However, this contribution greatly depends on the concentration, as it decreases with increasing BC masses. It is important to note that this result is representative of this episode only, with all the specificities it may be associated with. A similar work on a longer dataset (e.g. a year), with upwind/downwind discrimination, as performed by Petetin et al. (2014) in Paris, may lead to global/refined conclusions.

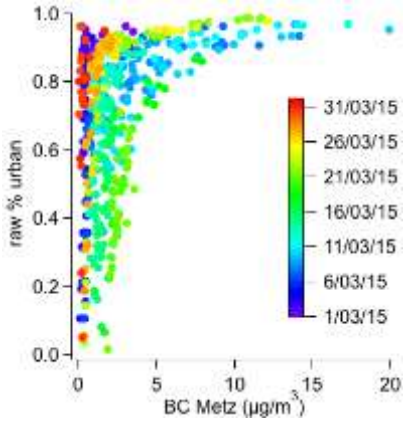


Figure 4. Fraction of urban BC compared to rural background during March 2015.

The wind analyses performed at both sites on BC concentrations during March 2015 show significant discrepancies in terms of geographical origins (Fig.5). In Metz, highest concentrations are observed at wind speeds close to 0 km/h, confirming the strong influence of local emissions. Also, a clear pathway from the North is observed with intermediate wind speeds (between 5 and 10 km/h). The precise geographical origin of this contribution cannot be precisely determined, mainly because wind speed is not directly proportional to a distance (in km). On the contrary, a completely different pattern is observed at OPE, in relation to dispersion conditions more favourable to advection (Fig. 2). A clear hotspot is observed in the NNE and NE sector, with intermediate wind speeds. Metz and Nancy (the other major city in Lorraine) fall within this sector and could thus significantly influence rural background at OPE. A major motorway connects both urban areas and has dense heavy-duty vehicles, due to the proximity with Belgium and Germany, and could also participate in observed background levels. Finally, long range advection, particularly with NE winds, could occur, since it has already been shown in Paris (Healy et al., 2012) or Lens (Waked et al., 2014). But the presence major of urban areas like Metz or Nancy upwind of OPE makes this assessment more laborious.

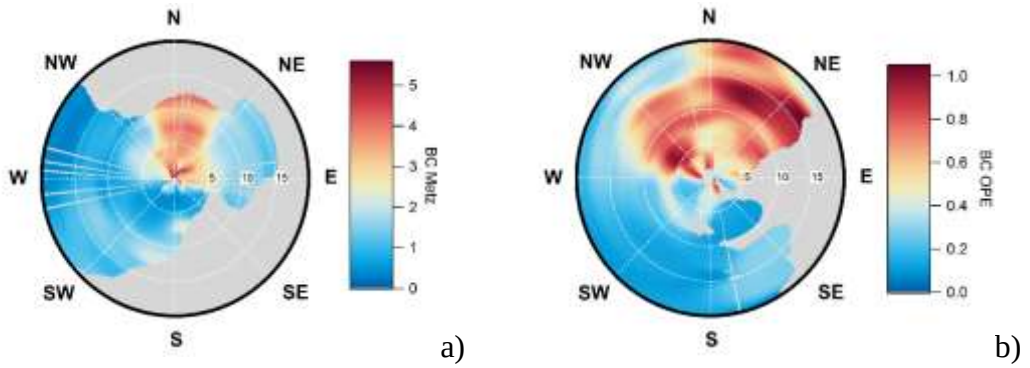


Figure 5. Wind analysis results on BC in a) Metz and b) OPE during March 2015. Radial axis represents wind speed (in km/h), colorscale is relative to the estimated concentrations (in µg/m^3)

Because the evaluation of the effectiveness of mitigation policies on BC comes to a satisfying ability of air quality models to represent its concentrations, it is critical to compare observed and modeled concentrations at different contribution levels (e.g. urban and rural). Such evaluation work is scarce in the literature. Gilardoni et al. (2011) proposed statistical tools to evaluate black carbon modeling using remote and rural sites across the world. Since BC is chemically inert in the atmosphere, the observed discrepancies between measured and modeled BC cannot be attributable to unknown chemical transformation pathways, but more to emission inventory issues and/or meteorological conditions. For example, Gadhavi et al. (2015) have shown seasonal variations in the agreement of the model they used, on a case study at a rural site in India, with highest discrepancies between January and April, associated with Southern air masses which are linked to an underestimation of open biomass burning emissions.

Here, it is very interesting to observe that modeled BC concentrations during the regional pollution episode (07/03 to 11/03) agree very well with the measurements at rural background (OPE), but not at urban background (Metz) (Fig. 6). Overall, Prev'est shows a much better performance at OPE than Metz, with for example a Normalized Mean Bias of -66% in Metz against -34% at OPE. The FAC2, fraction of modeled values within a factor of 2 of the observed values, is also much smaller in Metz (0.39), underlining the fact that the model has been unable to represent the amplitude of concentrations at urban background during this particular episode. On the other hand, modeled BC at rural background (OPE) stayed very low during the second sub-episode (between 19/03 and 22/03), characterized by an advected pattern.

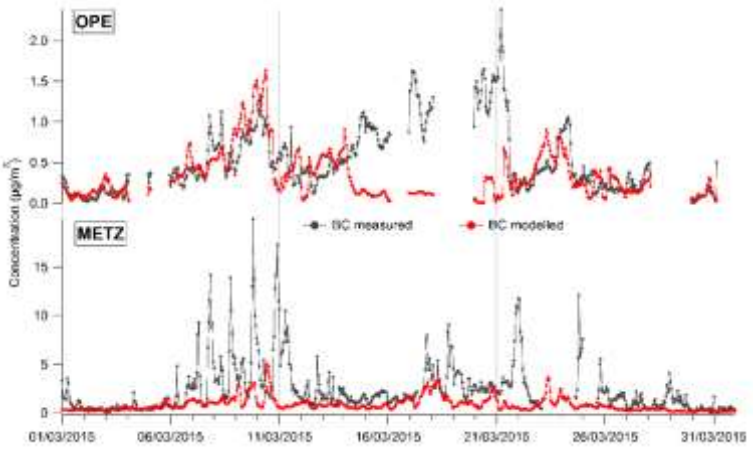


Figure 6. Comparison between BC measured and modeled at OPE and Metz

Table 1. Model evaluation statistics for BC at Metz and OPE during March 2015.

	BC Metz	BC OPE
FAC_2	0.39	0.625
MB	-1.56 µg/m^3	-0.18 µg/m^3
NMB	-0.66	-0.34
RMSE	2.91	0.51
r	0.34	0.15
COE	0.04	0.03
IOA	0.52	0.52

Figure 7 illustrates the difference between predicted and observed BC concentrations at OPE, colorcoded with the origin of the air masses, defined as clusters, from 1 to 5. Difference values range from +0.6 to -2.3 µg/m^3, highest discrepancies being observe exclusively with cluster 5, corresponding NE air masses. This air mass dependence is particularly highlighted in Fig. 8, illustrating the variations of the model evaluation statistics (described in Table 1) for the 5 clusters. Indeed, each statistic degrades in cluster 5 compared to the other clusters. For example, while RMSE stays below 0.21 for oceanic air masses (cluster 1 to 3), it reaches 0.32 and 0.83 for continental influences (clusters 4 and 5), respectively.

This can suggest that the transport of BC from other regions than Lorraine, but also from Metz for example since BC concentrations are badly modeled at urban background, is not well represented. Overall, these results emphasize the critical need of refined emission inventories in urban areas like Metz, but also a better knowledge (and/or consideration) of border emissions.

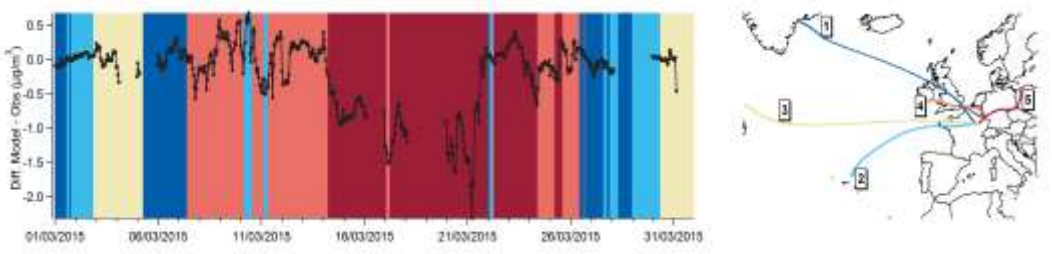


Figure 7. Temporal variation of the difference between BC modeled and observed at OPE. Colors in the background refer to clusters which are described in the right panel.

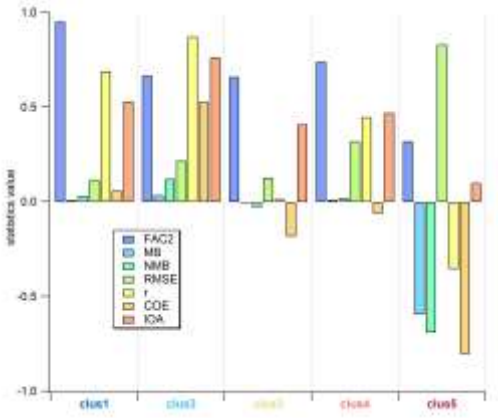


Figure 8. Model evaluation statistics for BC at OPE divided by clusters

Finally, it is important to note that this analysis could suffer from statistical artifact, as the 5 clusters are not temporally equally distributed (e.g. cluster 5 represents 32% of the air masses during this period, against 12% for cluster 2).

4 Conclusion

This study presents long-term black carbon measurements in a major urban area in the East of France. The fossil fuel fraction, linked to traffic emissions, makes up the majority of BC year long, but wood-burning exhibits a strong

seasonality, with highest concentrations during winter and early spring. During March 2015, the comparison between urban and rural levels revealed that most of urban concentrations (80% on median, but up to 98%) are associated with local emissions. Wind analysis performed on BC at the rural site showed that Metz and Nancy, the two major urban areas in Lorraine, influence rural background concentrations during the pollution episode. The role of Paris emissions, yet one the strongest emitter of BC in France, could not be highlighted, mainly because air masses from the West have a low representativeness during the period of study.

Finally, a model evaluation at both sites showed very contrasted results. Overall, the model is more reliable at rural than urban background (relative mean bias of around -34% and -66% respectively). This emphasizes the need of a better knowledge of i) emission factors and ii) emission inventories within urban areas. Also, cluster analysis showed the influence of air mass origin on the performance of the model, which dramatically decreased with continental air masses. Trans-regional modeling work is thus needed for a better prediction of black carbon concentrations.

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