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Long-term trends in black carbon from biomass and fossil fuel combustion detected at the JRC atmospheric observatory in Ispra

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Contents

Abstract 1

1 Introduction 2

2 Measurements and data processing 3

 2.1 Measurements 3

 2.2 Measurement data processing 3

 2.3 Emission data processing 4

3 Results 6

4 Conclusions 7

References 8

List of figures 9

Abstract

The concentrations of equivalent black carbon deriving from biomass burning $[eBC]_{bb}$ and fossil fuel combustion $[eBC]_{ff}$ have been estimated from measurements of aerosol light attenuation at several wavelengths (from infrared to ultraviolet) performed at the atmospheric observatory of the Joint Research Centre located in Ispra (Northern Italy). The data show repeated seasonal cycles from 2004 to 2016, which suggests that winter time wood burning for domestic heating is the main biomass burning activity in this area. The $[eBC]_{bb}/[eBC]_{ff}$ ratio has increased on average by +5%/yr over the 2007 – 2016 period. We compared these measurement-derived data with CO₂ emissions estimated from EDGAR relative to biomass burning for domestic heating and fossil fuel combustion for transport (Diesel) and residential heating (coal + oil) in the 0.4°x0.4° area centred on Ispra. The data shows an increase in CO₂ emissions from biomass burning compared to fossil fuel combustion from 2004 to 2008, and a rather constant ratio since then. There is no obvious correlation between the concentrations of $[eBC]$ and the statistics on CO₂ emissions from biofuel and fossil fuel combustion over the studied period. The impact of the economic crisis of 2009 on the use of biofuels for domestic heating cannot be rigorously demonstrated, neither from the measurement data nor from the emission inventory.

1 Introduction

Unlike fossil fuel combustion, biomass burning has been considered almost CO₂ neutral, since it mainly releases the CO₂ previously absorbed from the atmosphere by plants through photosynthesis. However, biomass burning also emits particulate matter (PM), including polycyclic aromatic hydrocarbons (PAHs) which are toxic. Biomass burning therefore degrades air quality. Biomass burning emitted PM also contains black carbon (BC), which is the most light-absorbing component of the atmospheric aerosol (Myhre, 2013). Therefore, wood burning also contributes to climate warming.

However, wood is currently the cheapest fuel for domestic heating in Europe. In rural and forested areas, it can even be gratis. Furthermore, incentives have also been provided in several European countries to promote the purchase of "clean stoves" burning wood pellets, etc (see for instance "Conto Termico" in Italy, from December 2012).

Has the economic crisis of 2008 – 2009 pushed European citizens to increase the share of solid biofuel in the fuel mix they use for domestic heating? Using the data we obtained at the Atmospheric Observatory of the Joint Research Centre in Ispra as a benchmark, we looked at long term variations in the contribution of biomass burning to black carbon concentrations. Ispra sits in a semi-rural zone in North-western Lombardy, which is the third most productive region in Europe from a total of 276 (Eurostat, 2017). Due to high emission rates, peculiar orography (between two high mountain ridges) and specific weather conditions (very low average wind speeds, very shallow mixed boundary layer in winter), Lombardy also experiences pollution levels that are amongst the highest in Europe (European Environment Agency, 2017), although improvements have been observed over the past 30 years, particularly regarding PM concentrations. Lombardy's gross domestic product (GDP) dropped significantly (-5%) in 2009, similar to many regions in Europe (Eurostat, 2017). Wood burning for domestic heating is an attractive alternative, especially in the Alpine and pre-Alpine areas, where fuelwood is quite abundant. It was indeed demonstrated in a previous study using measurements of organic and isotopic tracers, that biomass burning accounted for close to 50% of the elemental carbon concentration in Ispra during the periods January - March and October - December 2007 (Gilardoni et al., 2011). Therefore, measurements performed at the JRC-Ispra site offer a good opportunity to study the impact of the economic crisis on air pollution in relation to domestic wood burning.

2 Measurements and data processing

Measurement data were all obtained by the Air and Climate Unit at the atmospheric observatory of the JRC located in Ispra.

Emission data were estimated from JRC's EDGAR system, based on official national fuel consumption statistics.

2.1 Measurements

Measurements were performed with a light absorption photometer over the period 2004 – 2016. This instrument (Aethalometer AE-31, Magee) measures the attenuation of light through aerosol particles deposited on a filter tape, which sequentially moves into the measurement chamber. Measurements are performed at 7 wavelengths from ultraviolet (370 nm) to infrared (950 nm). The Aethalometer converts attenuation measurements to mass concentrations of "equivalent black carbon", [eBC], which are the concentrations of a virtual black carbon substance (with an assumed absorption cross section of 8.0 m²/g at 520 nm) that would lead to the light attenuation measured by the instrument. The Aethalometer produces 7 [eBC] concentrations corresponding to the 7 wavelengths every 2 to 5 min. When soot is black, [eBC] concentrations at all 7 wavelengths are equal. When soot has a more brownish hue, [eBC] is greater at wavelengths corresponding to blue and ultraviolet lights.

2.2 Measurement data processing

Light attenuation measurements performed with Aethalometers can be converted to aerosol light absorption in the atmosphere using several algorithms accounting for light scattering (which also contributes to light attenuation) and multiple scattering (which occurs in the aerosol deposited on the filter but not significantly in the air). A simple approach has recently been developed within the Aerosol, Cloud and Trace gas Research InfraStructure (ACTRIS) project, to directly estimate aerosol light absorption coefficients from multi-wavelengths absorption photometers with an uncertainty value of ±20% (Mueller, 2015).

For the Aethalometer AE-31, the formula is:

$$\alpha_{\lambda} = \frac{k}{\lambda} [eBC]_{\lambda} \quad \text{Eq. 1}$$

where α_{λ} is the aerosol light absorption coefficient (Mm⁻¹) at wavelength λ , λ is the light wavelength (nm), $[eBC]_{\lambda}$ is the concentration measured at wavelength λ (ng/m³), and k is a constant (=14.625/3.5).

Aerosol light absorption coefficients at 370 nm (UV) and 880 nm (IR) were estimated from [eBC] measurements according to Eq. 1.

Assuming that light absorbing particles come from only 2 sources, namely fossil fuel combustion (from transport, domestic heating, electricity generation, etc...) and biomass burning, we have:

$$\alpha_{370} = \alpha_{370_{ff}} + \alpha_{370_{bb}} \quad \text{Eq. 2}$$

and

$$\alpha_{880} = \alpha_{880_{ff}} + \alpha_{880_{bb}} \quad \text{Eq. 3}$$

The aerosol light absorption coefficient depends on the light wavelengths according to a power law:

$$\frac{\alpha_{\lambda_1}}{\alpha_{\lambda_2}} = \left(\frac{\lambda_1}{\lambda_2}\right)^{-\mathring{A}} \quad \text{Eq. 4}$$

where \mathring{A} is the absorption Ångström exponent, which depends on the "colour" of the absorbing particles. Several studies show that \mathring{A} is close to 1.0 for particles emitted by

fossil fuel combustion, while $\dot{A} = 1.9 \pm 0.1$ for particles emitted from biomass burning (Sandradewi et al., 2008).

Substituting $\alpha_{370_{ff}}$ and $\alpha_{370_{bb}}$ in Eq. 2 using Eq. 4 for both fossil fuel combustion ($\dot{A} = 1.0$) and biomass burning ($\dot{A} = 1.9$), we could calculate $\alpha_{880_{ff}}$ and $\alpha_{880_{bb}}$, and therefore the concentrations of eBC coming from fossil fuel combustion ($[eBC]_{ff}$) and biomass burning ($[eBC]_{bb}$) using Eq. 1.

2.3 Emission data processing

CO₂ emission estimates were calculated by the EDGAR system, using the official fuel consumption statistics provided by the International Energy Agency (IEA, 2017), and technologies and emission factors of EDGARv4.3.2 (Janssens-Maenhout et al., submitted).

The IEA 2017 biomass consumption data are very different from previous estimates, as they are no longer based on the biomass sold, but instead are modelled considering information on biomass availability and population for a certain area.

National CO₂ emissions were then gridded at 0.1°x0.1° resolution using the proxy data of EDGARv4.3.2 (Janssens-Maenhout et al., submitted). In particular, emissions from the residential sector were gridded using the gridded population provided by the Center for International Earth Science Information Network (CIESIN, 2005). For what concerns road transport, emissions were distributed on three road types (highways, primary and secondary, residential and commercial roads) obtained from the OpenStreetMap of Geofabrik (2015), and weighted considering the different type of vehicles circulating on the different types of roads.

Emissions from the area centred on Ispra comprised between 45.6° and 46.0°N, and between 8.4° and 8.8°E were calculated from the gridded 0.1°x0.1° emission fields.

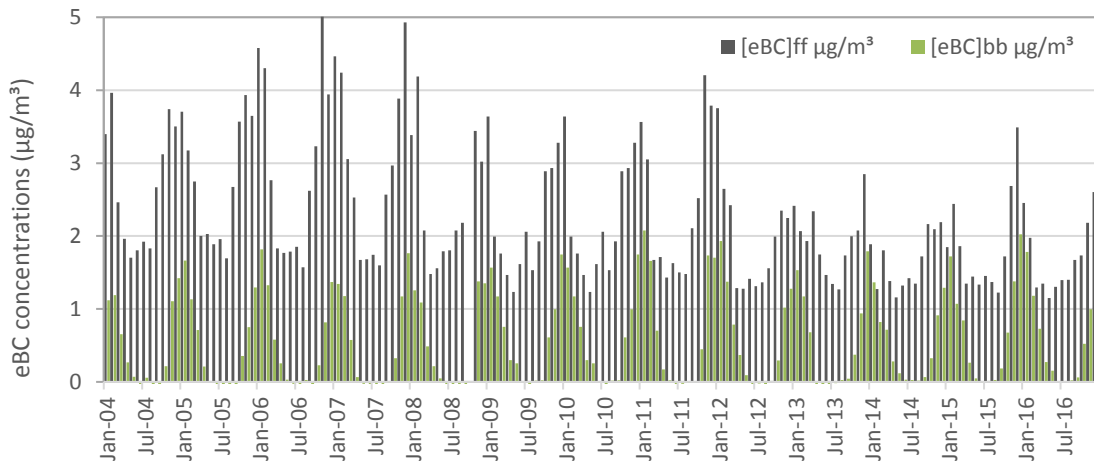


Figure 1: Monthly mean concentrations of equivalent Black Carbon coming from fossil fuel combustion ($[eBC]_{ff}$) and from biomass burning ($[eBC]_{bb}$).

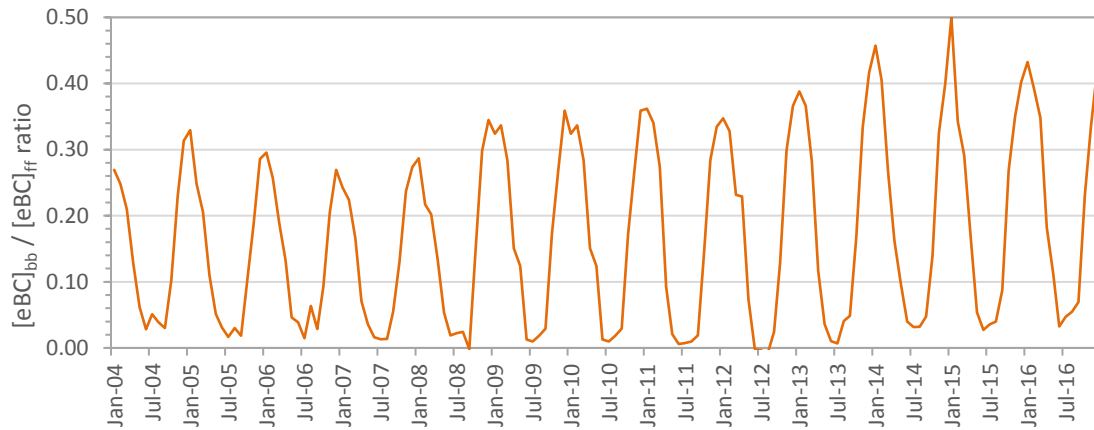


Figure 2: Monthly mean ratios between equivalent Black Carbon concentrations coming from biomass burning ($[eBC]_{bb}$) and from fossil fuel combustion ($[eBC]_{ff}$).

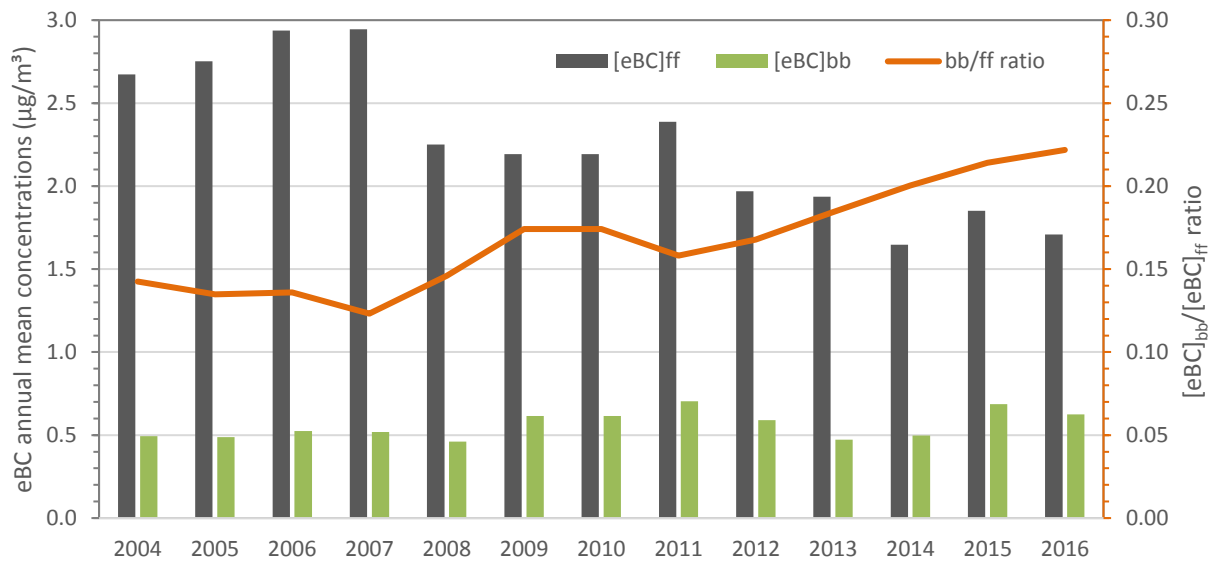


Figure 3: Annual average concentrations of equivalent Black Carbon (eBC) coming from fossil fuel combustion ($[eBC]_{ff}$) and from biomass burning ($[eBC]_{bb}$) on the left hand axis; annual mean ratios between $[eBC]_{bb}$ and $[eBC]_{ff}$, right hand axis.

3 Results

Figure 1 shows the variations in monthly mean concentrations of equivalent Black Carbon from fossil fuel combustion ($[eBC]_{ff}$) and biomass burning ($[eBC]_{bb}$) in Ispra from 2004 to 2016, as derived from Aethalometer measurements using Eq. 1 to 4. Seasonal variations are primarily due to variations in pollution dilution: the thickness of the mixed boundary layer (the fraction of the atmosphere where pollutants emitted from the Earth's surface are dispersed) is less during winter time, as previously demonstrated by Barnaba et al., 2010. However, conversely to $[eBC]_{ff}$, $[eBC]_{bb}$ drops down to $<0.1 \mu\text{g}/\text{m}^3$ every year from May to September, which is consistent with the fact that eBC_{bb} mainly derives from wood burning for domestic heating. The seasonal cycle in the $[eBC]_{bb}/[eBC]_{ff}$ ratio is obvious, with summer minimum values ≤ 0.03 , and winter maximum values ranging from 0.25 to 0.50 (Figure 2). Winter time maxima increased from 2008 to 2015, while summer time minima were greater in 2014 – 2016 than during the previous years.

Monthly data shown in Figure 1 and Figure 2 have been aggregated to annual averages. Figure 3 shows that while the concentration $[eBC]_{ff}$ dropped stepwise from 2007 to 2016, the concentration $[eBC]_{bb}$ was higher than average in 2009 – 2011 and 2015 – 2016. As a consequence, the ratio $[eBC]_{bb}/[eBC]_{ff}$ increased by almost 50% since 2007, with a statistically significant (confidence level = 99%) slope of +5%/yr, and a temporary maximum in 2009 – 2010 (Figure 3).

For comparison, CO_2 emissions from fossil fuel combustion (Diesel engines + coal & oil for domestic heating) and biomass burning in a $0.4^\circ \times 0.4^\circ$ ($12 \times 9 \text{ km}^2$) grid cell centred on Ispra are shown in Figure 4. CO_2 emission data suggest a steady decrease in the combustion of fossil fuels from 2006, and an increase in wood burning for domestic heating from 2004 to 2008, followed by a slow decrease since then, with a local minimum in 2011. The share of wood burning to the CO_2 emissions in the area around Ispra was large in 2008 – 2010 (Figure 4), at the peak of the economic crisis, but not greater than in 2012 – 2013.

From 2004 to 2015, the ratio in CO_2 emissions from fossil fuel combustion and wood burning around Ispra (0.39) was much larger than the $[eBC]_{bb}/[eBC]_{ff}$ ratio (0.16), which suggests that eBC emission factors from these two sources are different. None of the variations observed in the $[eBC]_{bb}/[eBC]_{ff}$ ratio can be explained from variations in the ratio between CO_2 emissions from wood and fossil fuels in the 108 km^2 area centred on Ispra, except perhaps the low value of 2011.

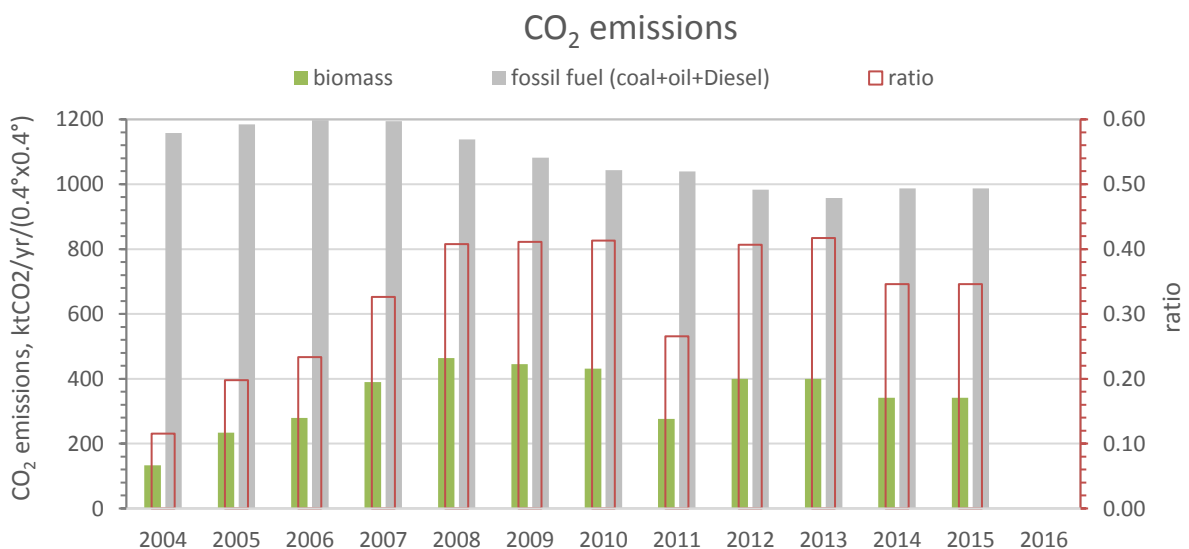


Figure 4: CO_2 (ktCO_2/yr) emissions from fossil fuel combustion and biomass burning for domestic heating in the $0.4^\circ \times 0.4^\circ$ cell centred on Ispra (left hand axis); ratio between CO_2 emissions from biomass burning and fossil fuel combustion (right hand axis).

4 Conclusions

Over the period 2004 – 2016, the ratio between the concentrations of equivalent Black Carbon attributed to biomass burning ($[eBC]_{bb}$) and fossil fuel combustion ($[eBC]_{ff}$) detected at the atmospheric observatory of the JRC in Ispra has followed a strong seasonal cycle with minimum values (<0.03) in summer and maximum values (0.25 to 0.50) in winter. This seems to indicate that most biomass burning was from winter time wood burning for domestic heating. Annual average $[eBC]_{bb}/[eBC]_{ff}$ ratios increased significantly from 2007 onwards ($+5\%/yr$). The overall average $[eBC]_{bb}/[eBC]_{ff}$ ratio over 2004 – 2016 was 0.17.

The ratio between CO_2 emissions from biomass burning for domestic heating and fossil fuel (oil + coal) combustion from transport and domestic heating in the $0.4^\circ \times 0.4^\circ$ ($12 \times 8 \text{ km}^2$) grid cell around Ispra increased during the period 2004 - 2008, remained quite constant from 2008 to 2013 (with a local minimum in 2011), and generally decreased since then.

Although both data sets show temporal maxima around 2009 and a local minimum in 2011, there is no statistically significant correlation between the eBC concentration ratio and the CO_2 emission ratio from biomass and fossil fuel combustions. The long term increase in the $[eBC]_{bb}/[eBC]_{ff}$ ratio can be explained by a dramatic increase in the eBC_{bb} emission factor, or by a decrease in eBC_{ff} emission factor, or by a combination of both. The increase in the $[eBC]_{bb}/[eBC]_{ff}$ observed in 2009 – 2010 is possibly related to the economic crisis. The steady increasing rate observed from 2011 to 2016 could also be related to the economic stagnation observed in Lombardy over the same period, but a more detailed knowledge of the biofuel consumption in the region would be needed before definitive conclusions can be drawn.

It would be worth expanding this study to a set of atmospheric data from stations where similar multi-wavelength aerosol light absorption data series are available.

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List of figures

Figure 1: monthly mean concentrations of equivalent Black Carbon coming from fossil fuel combustion ([eBC]_{ff}) and from biomass burning ([eBC]_{bb}). 5

Figure 2: monthly mean ratios between equivalent Black Carbon concentrations coming from biomass burning ([eBC]_{bb}) and from fossil fuel combustion ([eBC]_{ff}). 5

Figure 3: annual averages of equivalent Black Carbon (eBC) coming from fossil fuel combustion ([eBC]_{ff}) and from biomass burning ([eBC]_{bb}) on the left hand axis; annual mean ratios between [eBC]_{bb} and [eBC]_{ff}, right hand axis. 5

Figure 4: CO₂ (ktCO₂/yr) emissions from fossil fuel combustion and biomass burning for domestic heating in the 0.4°x0.4° cell centred on Ispra (left hand axis); ratio between CO₂ emissions from biomass burning and fossil fuel combustion (right hand axis)..... 6

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