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IDENTIFYING BLACK CARBON SOURCES AND CONCENTRATIONS IN AN URBAN HOTSPOT OF THE CENTRAL PO VALLEY FOR TARGETED MITIGATION STRATEGIES USING HIGH-RESOLUTION DISPERSION MODELLING AND MICRO-AETHALOMETERS

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Abstract: Black carbon (BC), a component of particulate matter resulting from the incomplete combustion of fossil fuels and biomass, has received increasing attention due to its adverse effects on human health and its significant impact on the earth's radiation balance. Understanding sources of BC in cities is crucial for air quality management. While past research has examined BC levels in rural and remote areas, data for urban environments, particularly regarding spatial distribution and source identification, has been limited. This study addresses this gap by investigating BC levels and sources in Modena, Italy, a representative medium-sized European city.

Using a combination of micro-aethalometer measurements and modelling simulations, we analysed BC concentrations and performed a source apportionment analysis focusing on two consecutive winter seasons (February-March 2020 and December 2020-January 2021). Micro-aethalometer measurements were apportioned to sources (fossil fuel combustion vs. biomass burning) and components (BC vs. brown carbon) using the multi-wavelength absorption analyser model, while dispersion modelling simulations were conducted with a hybrid modelling system consisting of the chemical transport model CHIMERE and the urban modelling suite GRAMM-GRAL. Analysis of the measurements revealed clear patterns. Brown carbon had minimal daily variation, while fossil fuel BC combustion sources peaked during rush hours, particularly at the traffic site. Biomass combustion sources, on the other hand, showed less variation throughout the day. The hybrid modelling system highlighted the most critical areas of the city in terms of concentration peaks. Additionally, it showed that more than half (52% ± 10%) of the BC concentrations originated from sources located in Modena, with biomass burning being the main source (35% ± 15%), followed by fossil fuel combustion (9% ± 4%). Long-distance transport also played a role, contributing 48% ± 10% (28% ± 1% from biomass combustion and 15% ± 2% from fossil fuel combustion) of BC. These results provide valuable insights for policy makers and urban planners. By considering both local and long-distance sources, they can develop effective strategies to manage BC levels in Modena and similar cities.

***Key words:** Black carbon, source apportionment, micro-aethalometers, hybrid Eulerian-Lagrangian modelling tool.*

INTRODUCTION

Atmospheric aerosol particles play an important role in the earth's radiative balance by directly scattering and absorbing solar and terrestrial radiation. Indirectly, they act as cloud condensation nuclei, influencing cloud formation and properties. Among the major aerosol constituents, black carbon (BC) is emerging as an important player in the atmospheric system due to its ability to absorb light over a wide range of wavelengths, from the UV to the IR, leading to localised atmospheric warming and influencing the distribution of solar radiation within the atmosphere. In addition to its impact on climate, BC also has critical implications for public health, as exposure to its concentrations has been linked to a range of respiratory and cardiovascular diseases, from aggravation of asthma to chronic bronchitis, cardiovascular disease and even lung cancer. For these reasons, understanding the role of BC in atmospheric processes remains of paramount importance for both environmental and public health concerns.

The growing concern about health and climate impacts has sparked an increase in research focused on urban areas, particularly examining BC concentrations as a crucial indicator of urban pollution. Consequently, various studies have explored the fluctuations of BC mass concentrations at different urban locations (Savadkoohi et al., 2023; Grange et al., 2020). While these studies employed precise methods for measuring BC, their findings were restricted to specific monitoring sites. Therefore, they could not deliver continuous spatial data across the entire city, which is essential for comprehensive epidemiological studies. In addition to spatially resolved information, source apportionment is also essential for understanding the specific pollution sources contributing to atmospheric concentrations.

The present study aims to provide information on the levels of BC and to identify its main sources in the urban environment of Modena, a city of about 200,000 inhabitants located in the middle of the Po Valley. To accomplish this task, we conducted a two-sampling period winter campaign (February-March 2020 and December 2020-January 2021), using two filter-based mid-cost instruments named micro-aethalometers, at different urban locations, traffic and background sites. To support the identification of local and regional sources and to provide spatial information about BC concentrations, we employed a hybrid Eulerian-Lagrangian modelling system (Ghermandi et al., 2020; Veratti et al., 2020; 2021). By combining observed and modelled data, we aimed to improve our understanding of the main emission sectors contributing to total BC levels for the development of effective mitigation strategies and to provide valuable insights for addressing environmental and health concerns related to BC.

DESCRIPTION OF THE MEASUREMENT CAMPAIGN

The measurement campaign was carried out from 4 February to 7 March 2020 and from 26 December 2020 to 21 January 2021 using two MA200 micro-aethalometers (microAeth® MA Series, Aethlabs, USA) equipped with PTFE filter tapes. These instruments quantify the absorption of light-absorbing carbonaceous aerosols at five different wavelengths (375, 470, 528, 625 and 880 nm), allowing for the estimation of the equivalent BC (eBC) concentrations by employing wavelength-specific mass absorption efficiencies. Sampling was performed at two air quality monitoring stations in the city, one located in a public park, representing typical background conditions, and the other located in an urban traffic hotspot, serving as a representative location for areas of dense traffic and associated air pollution (for further details on the measurement locations see Bigi et al., 2023). The instruments operated at a flow rate of 100 ml min^{-1} in dual-spot sampling mode to compensate for filter loading effects and a constant correction factor of 1.3 was chosen to account for multiple light scattering within the filter fibres and between deposited particles and the filter, as suggested by the manufacturer. Absorption measurements from the MA200 devices were further partitioned into specific components, including BC, BrC and their respective sources, namely fossil fuel (FF) and biomass burning combustion (BB), using the Multi-Wavelength Absorption Analyzer model (Massabò et al., 2015). The Absorption Ångström Exponent (α) for BC and FF was set equal to 1, while α for BB was set equal to 2. These values correspond respectively to the midpoint of the α probability density function (PDF) during the morning peak hour at the urban traffic site and the upper tail of the α PDF calculated from the fit over the five wavelengths and applying a stringent filter of $r^2 > 0.99$.

DESCRIPTION OF THE HYBRID EULERIAN-LAGRANGIAN MODELLING SYSTEM

To obtain detailed spatial information on BC levels, a hybrid Eulerian-Lagrangian modeling system was implemented. The GRAMM-GRAL suite (Oettl et al., 2015) was chosen due to its capability to simulate emission dispersion within the study area, considering the influence of urban obstacles like buildings on flow field reconstruction. Additionally, the chemical transport model CHIMERE (Mailler et al., 2017) was essential for complementing GRAMM-GRAL's output by estimating background concentrations from external sources and evaluating regional impacts on BC levels. By leveraging the strengths of both models, we aimed to develop a thorough understanding of BC concentrations in the study area, accounting for the complex dynamics of BC transport in the urban environment.

The mesoscale meteorological simulations with GRAMM were carried out in a domain covering an area of 30 × 30 km^2 centred in Modena, with a horizontal resolution of 200 m. The simulation domain was chosen to be large enough to represent all topographical features potentially affecting the flow in the city. The domain extends vertically from the surface to 3000 m above ground, with 22 geometrically spaced layers varying in thickness from 12 m near the surface to 500 m in the free troposphere. GRAL dispersion simulations were performed at the urban scale on a smaller domain of 10.2 x 7.7 km^2, covering most of the urban area of Modena, with a horizontal resolution of 4 m. On the other hand, CHIMERE was set-up over a domain covering a significant part of the Po valley (from 8.78° E to 13.13° E longitude and from 43.57° N to 45.79° N latitude), with a fine horizontal resolution of 3 km. The vertical grid consisted of 15 levels, ranging from 997 hPa (25 m) for the first layer to 500 hPa. See Veratti et. al (2023) for more details on CHIMERE configurations.

To estimate traffic emissions at the city scale, we used a comprehensive bottom-up approach that integrated activity data with the VERT traffic emissions model (Veratti et al., 2024). VERT implements the Tier 3 and Tier 2 methodologies outlined in the European EMEP/EEA guidelines to estimate exhaust and non-exhaust emission factors. In addition, VERT takes into account additional data such as local fleet composition, road gradient and average driving speed to improve the reliability of the calculations. In order to fully assess the overall impact of traffic, the resuspension emissions were also included in the calculation.

Anthropogenic emissions for other source sectors were taken from the regional emission inventory for the Emilia-Romagna region, which is regularly compiled by the local environmental agency Arpae. As emissions from biomass burning are subject to large uncertainties, we simulated five different emission scenarios based on different assumptions about the location of sources and the speciation conversion factors from total $PM_{2.5}$ emissions, reported in the local regional inventory, to BC emissions. To maintain consistency in the simulation, the same emission inventory was also used to perform simulations at the regional scale with the CHIMERE model.

RESULTS

Results from the measurement campaign showed that BrC concentrations were consistent and remarkably low throughout the day in both the analysed periods (around 0.05 μg m^{-3} in the first, 0.1 μg m^{-3} in the second). On the other hand, FF, a marker for traffic-related emissions, showed distinct diurnal peaks during rush hours, in the morning (07:00 - 08:00 GMT) and in the evening (18:00 - 20:00 GMT) for both periods, with values up to 18.5 μg m^{-3} at the urban traffic site. In contrast, BB showed less diurnal variability, with a minimum in the early afternoon (15:00 - 17:00 GMT) and a peak in the evening (20:00 - 21:00 GMT), and generally lower concentrations compared to FF. A comparison of the two periods showed significant differences. The period from 26 December 2020 to 21 January 2021 had higher FF and BB concentrations, likely due to different meteorological conditions such as lower wind speeds and increased atmospheric stability, accentuated by frequent thermal inversions occurring even during daylight hours. In addition, lower temperatures likely led to more biomass burning for heating, which increased eBC concentrations.

In the second part of the study, the hybrid modelling system was used to provide spatially resolved BC concentration maps over the urban area of Modena and to elucidate the contribution of different sources. Figure 1 shows the spatial distribution of the average BC concentrations derived from the hourly maps during the two simulated periods. The hybrid modelling system identified hotspots within the urban area exhibiting significantly elevated BC concentrations, reaching up to 6.72 μg m^{-3}. These findings highlight the critical role of spatial modelling in capturing the heterogeneity of pollution levels, which may be missed by relying solely on discrete monitoring station data.

Model validation involved comparing simulated concentrations from the hybrid system with observations from MA200 instruments. During February-March 2020, the model showed good performance, with a mean bias (MB) of 0.02 μg m^{-3} (+2%) at the urban background site and -0.12 μg m^{-3} (-5%) at the traffic site. The linear correlation coefficients were also satisfactory (0.51 and 0.62 respectively). In particular, daily concentration peaks were accurately captured under different meteorological conditions, including high wind speed episodes. However, the period December 2020 - January 2021 showed contrasting behaviour between stations. Limited background data coincided with a tendency to overestimate biomass burning emissions, especially in the morning. This effect, similar to the first period, was amplified by the holiday influence (7 measurement days out of 13 were holidays). Here the MB increased to +15% and the correlation coefficient decreased to 0.38. Conversely, complete traffic station data (covering the entire period) coincided with favourable conditions for pollutant accumulation (high pressure and thermal inversions). Under these circumstances, the model faced challenges in capturing the complex meteorological dynamics, resulting in a lower MB (-22%) and correlation coefficient (0.34).

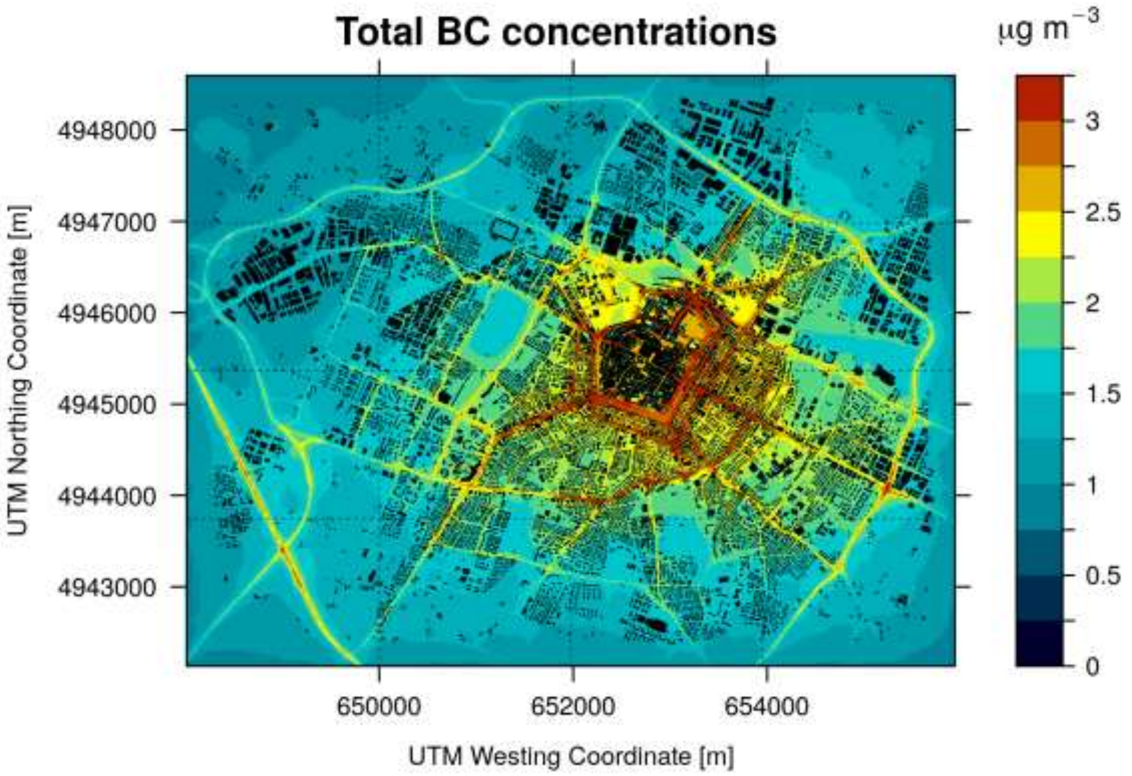

Figure 1. Spatial distribution of average BC concentrations for both the analysed periods (February - March 2020 and December 2020 - January 2021).

In addition, potential nearby high emission sources may have contributed to elevated BC concentrations at the traffic site. Despite the observed decrease in performance in the second period, all statistical metrics employed (including factor of two, normalised mean difference, fractional bias, and normalised mean square error) remained within established acceptance criteria for urban dispersion modelling (Hanna and Chang, 2012).

The analysis of model results revealed that local sources were responsible for 52% ± 10% of the total average BC, with background sources contributing the remaining 48% ± 10%. A more detailed breakdown of the local contributions identified biomass burning (BB) as the main source (35% ± 15%), followed by fossil fuel burning (FF) with 9% ± 4%. Non-exhaust emissions and re-suspension each contributed about 4% (4% ± 2% and 4% ± 3% respectively). It is noteworthy that traffic was the dominant contributor to BC emissions, while other FF sources had a minimal impact. In contrast, BB emissions came equally from open fireplaces, conventional stoves, high efficiency stoves and modern stoves as reported in the Arpae local emission inventory. Long-distance transport contributed to BB (28% ± 1%), FF (15% ± 2%), non-exhaust (3% ± 2%) and re-suspension (2% ± 2%) emissions.

The main source of traffic-related BC was exhaust emissions (50%), with Euro 4 diesel cars being the largest contributor (19% of total BC). The remaining 50% of traffic-related BC came equally from non-exhaust emissions and re-suspension (25% each).

CONCLUSIONS

This study investigated BC concentrations in Modena, Italy, through a measurement campaign and a hybrid Eulerian-Lagrangian modelling system. The measurement campaign revealed distinct diurnal variations in traffic-related BC with morning and evening peaks, while biomass burning showed less diurnal variability. The modelling system effectively captured the spatial distribution of BC, identifying hotspots within the city and highlighting the limitations of relying solely on monitoring stations. The modelling system achieved acceptable performance criteria when compared to observations, even under challenging winter meteorological conditions. The source apportionment analysis revealed that local sources were responsible for more than half of the BC, with BB as the main contributor (35%), followed by traffic (9%) and non-exhaust emissions and re-suspension (around 4% each). Euro 4 diesel cars were identified as the largest single source of traffic-related BC. These results can serve as a valuable basis for the development of more robust air quality management strategies in Modena and similar cities in the Po Valley.

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