



Supplement of

Measurement report: Source attribution and estimation of black carbon levels in an urban hotspot of the central Po Valley – an integrated approach combining high-resolution dispersion modelling and micro-aethalometers

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Table S1. Comparison of MAE values used in this study with measurements from different European sites. The values in brackets represent the minimum and maximum MAE values observed during the reference period, while the mean MAE plus or minus the standard deviation is indicated.

| Reference | Location | Type of sampling site | Instrument for b_{abs} | Period | Wavelength (nm) | $\underset{(m^2g^{-1})}{\text{MAE}}$ |
|--------------------------|--|-----------------------------------|--------------------------|------------------------|---------------------------------|--|
| This study | - | - | - | - | 880 625 528 470 375 | 10.120 14.091 17.070 19.070 24.069 |
| | Athens (GR) | urban background | AE33 | 2017-2020 | 880 | 6.1 ± 4.0 |
| | Marseille (FR) | urban background | AE33 | 2014-2019 | 880 | 5.1 ± 1.7 |
| | Zurich (CH) | urban background | AE33 | 2012-2021 | 880 | 4.9 ± 2.3 |
| | Rome (IT) | urban background | AE33 | 2019-2022 | 880 | 8.8 ± 3.1 |
| | Elche (SP) | urban background | AE33 | 2022-2023 | 880 | 7.3 ± 2.1 |
| | Athens (GR) | suburban | AE33 | 2017-2019 | 880 | 10.1 ± 2.3 |
| | Bern (CH) | urban traffic | AE31-AE33 | 2015-2021 | 880 | 5.7 ± 3.5 |
| | Paris (FR) | suburban | AE31-AE33 | 2012-2021 | 880 | 5.5 ± 3.0 |
| | Payerne (CH) | rural background | AE31-AE33 | 2012-2021 | 880 | 12.4 ± 4.1 |
| | London (UK) | urban background | AE22 | 2009-2019 | 880 | 8.8 ± 2.6 |
| Savadkoohi et al. (2024) | London (UK) | urban traffic | AE22 | 2009-2019 | 880 | 9.7 ± 1.8 |
| , | Helsinki (FN) | urban traffic | MAAP | 2015-2019 | 637 | 12.4 ± 4.7 |
| | Leipzig (DE) | urban traffic | MAAP | 2011-2019 | 637 | 13.8 ± 4.6 |
| | Dresden (DE) | urban traffic | MAAP | 2017-2019 | 637 | 9.6 ± 2.6 |
| | Barcelona (SP) | urban background | MAAP | 2009-2021 | 637 | 9.6 ± 2.6 |
| | Granada (SP) | urban background | MAAP | 2006-2020 | 637 | 10.4 ± 3.3 |
| | Milon (JE) | urban background | MAAP | 2017-2019 | 637 | 10.4 ± 4.1 11.2 ± 6.5 |
| | $\frac{1}{2} \frac{1}{2} \frac{1}$ | urban baakground | MAAP | 2013-2021 | 637 | 11.2 ± 0.3 14.0 ± 2.7 |
| | Huytiala (CZ) | rural background | ΜΑΑΓ | 2021-2022 | 637 | 14.0 ± 3.7 73 ± 47 |
| | Ispra (IT) | rural background | ΜΔΔΡ | 2008-2020 | 637 | 10.8 ± 4.4 |
| | 13piu (11) | Turur buckground | 1017 11 11 | 2000 2020 | 037 | 10.0 ± 1.1 |
| | Kramolín (CZ) | | AE31 | 2017 | 950 | 4.14 ± 1.39 |
| | | rural background | | | 880 | 4.93 ± 1.96 |
| | | | | | 660 | 6.71 ± 2.28 |
| Mbengue et al. (2021) | | | | | 590 | 7.35 ± 2.57 |
| | | | | | 520 | 8.46 ± 3.00 |
| | | | | | 470 | 10.34 ± 3.71 |
| | | | | | 370 | 12.10 ± 4.72 |
| Yuan et al. (2021) | Melpitz (DE) | regional background | PAX | Feb 2017 | 870 | 7.4 (7.2-7.9) |
| Ciupek et al. (2021) | London (UK) | urban traffic urban background | AE22 | 2019 2018 | 880 | 7.8 (6.0-9.8) 7.8 (6.0-9.8) |
| Gilardoni et al. (2020) | Milan (IT) | urban background | AF22 | Feb 2016 | 880 | 9.8 ± 2.2 |
| | Motta Visconti (IT) | rural background | | Feb-Mar 2016 | | 10.5 ± 1.1 |
| Grange et al. (2020) | Switzerland | 4 rural and 2 urban sites | AE33 | 2008-2018 | 950 | 11.3 ± 2.9 ^a |
| Mousavi et al. (2019) | Milan (IT) Bareggio (IT) | urban background | AE31 AE51 | 2017-2018 2017-2018 | 880 | 9.5 ± 1.3 |
| | | urban background | | | 370 | 16.5 ± 0.6 |
| | | rural background | | | 880 | 8.5 ± 1.2 |
| | | | | | 370 | 16.2 ± 1.2 |
| Zanatta et al. (2016) | Aspyreten (SE) | regional background | PSAP | 2010-2011 | 637 | 8.38 + 1.22 |
| | Birkenes (NO) | regional background | PSAP | 2010-2011 | 637 | 8.09 ± 1.33 |
| | Finokalia (GR) | rural background | PSAP | 2008-2010 | 637 | 15.1 ± 1.75 |
| | Puy de Dôme (FR) | rural background | PSAP | 2010-2011 | 637 | 7.04 ± 1.87 |
| | Harwell (UK) | regional background | MAAP | 2010 | 637 | 8.87 ± 1.84 |
| | Ispra (IT) | rural background | MAAP | 2008-2011 | 637 | 9.31 ± 1.35 |
| | Melpitz (DE) | regional background | MAAP | 2008-2010 | 637 | 8.22 ± 1.47 |
| | Montseny (SP) | rural background | MAAP | 2008-2011 | 637 | 8.29 ± 1.82 |
| Laborde et al. (2013) | Paris (FR) | suburban 1 | AE31 | Jan-Feb 2010 | 880 | 8.6 (7.3-8.8) |

^a Average value of six different sites, four of rural background (Payerne, Magadino-Cadenazzo, San Vittore and Rigi-Seebodenalp), one of urban background (Zurich-Kaserne) and one of urban traffic (Bern-Bollwerk).

S1 Analysis of the absorption Ångström exponent

Combustion sources such as wood burning and traffic emissions have distinct wavelength-dependent light absorption signatures, particularly in the ultraviolet and lower visible range. This characteristic fingerprint allows their contribution to be identified from the total measured absorption Zotter et al. (2017); Sandradewi et al. (2008); Massabò et al. (2015); Bernardoni

- 5 et al. (2017). The absorption Ångström exponent (α) serves as a single parameter to quantify the source-specific dependence of light absorption coefficient on wavelength. In the absence of validation measurements, the α frequency distribution can guide the estimation of suitable α values for the source apportionment model. Figure S1 shows the probability density function of α for the hourly aggregated measurements during the two periods analysed in the main text, i.e. from 4 February to 7 March 2020 and from 26 December 2020 to 21 January 2021 at the traffic site, and from 4 February to 7 March 2020 and
- 10 from 26 December 2020 to 7 January 2021 at the background site. Data reported with light blue colour are calculated as follow:

 $\alpha = -\frac{\log(b_{880}/b_{375})}{\log(880/375)}$

Where b_{880} and b_{375} are the absorption coefficients at 880 nm and 375 nm respectively. While the data shown in grey are obtained by fitting the absorption coefficient as a function of the five wavelengths (375, 470, 528, 625 and 880 nm) with a filter of $r^2 > 0.99$ to the fit. In addition, a subplot for the traffic site has been incorporated under its facet, showing α values only for the morning rush hour (08:00 GMT time) on working days, i.e. when the traffic contribution is expected to be at its peak. For traffic, the source-specific α value is set to 1, which corresponds to the centre of the probability density function (PDF) distribution for the morning rush hour on working days at the traffic site. Conversely, the α value for biomass burning is set

20 to 2, representing the upper tail of the stringent-filtered PDF distribution for both traffic and background sites, as suggested by Tobler et al. (2021).



Figure S1. The absorption Ångström exponent (α) probability density function calculated from the ratio of the 375 nm and 880 nm wavelengths and from the fit off all wavelengths from 375 nm to 880 nm and filtered for fit $r^2 > 0.99$.

| Emission Sector | Emission activity | Fuel | BC/PM _{2.5} (%) | BC EF |
|------------------------|---|--------------------------------------|-----------------------------|---|
| Energy production | all | liquid | 56 | $1.081 (0.05 - 5.04) \mathrm{g Gj^{-1}}$ |
| Energy production | all | biomass and agricultural residues | 6.4 | $4.389(2.1788.778)\mathrm{gGj^{-1}}$ |
| Energy production | all | CNG | 4 | $0.0056 \mathrm{~g~Gj^{-1}}$ |
| Domestic heating | open fireplaces | biomass | 7 | $57.4 (28.7 - 114.8) \text{ g Gj}^{-1}$ |
| Domestic heating | conventional stoves burning | biomass | 10 | $74 (37-148) \text{ g Gj}^{-1}$ |
| Domestic heating | high-efficiency stoves | biomass | 16 | 59.2 (45.6–118.4) g Gj^{-1} |
| Domestic heating | advanced/ecolabelled stoves and boilers | biomass | 28 | 44.8 (22.4–89.6) g $\rm Gj^{-1}$ |
| Domestic heating | pellet stoves and boilers | biomass | 15 | 9 (4.5–18) g Gj ^{-1} |
| Domestic heating | all | CNG | 5.4 | $0.119 (0.07 - 0.167) \text{ g Gj}^{-1}$ |
| Industry | all | solid fuel | 6.4 | $6.912 (3.84 - 14.08) \text{ g Gj}^{-1}$ |
| Industry | all | CNG | 4 | $0.0312 (0.019 - 0.014) \mathrm{g G j^{-1}}$ |
| Industry | all | liquid | 56 | $11.2 (6.72 - 15.68) \text{ g Gj}^{-1}$ |
| Other mobile machinery | all | liquid | 70 | $0.78 (1.540 - 0.002) \text{ g kWh}^{-1}$ |
| Waste management | incinerator | all | 20 | $0.105~(0.0380.29)~\mathrm{g~Gj^{-1}}$ |

Table S2. BC speciation factors from $PM_{2.5}$ emissions and BC emission factors (EF) expressed as g of BC per GJ or kWh net thermal input (with related 95% confidence intervals).

S3 Model evaluation

To evaluate the performance of the models in reproducing BC concentrations and wind speed, several statistical indicators were

25 considered. These indicators were derived by considering M as the modelled values, O as the observations, n as the count of model-observation pairs,

$$\bar{M} = \frac{1}{n} \sum_{i=1}^{n} M_i$$

as the average modelled value, and

$$\bar{O} = \frac{1}{n} \sum_{i=1}^{n} O_i$$

30 as the average observed value. The following metrics were employed for evaluation:

$$MB = \frac{1}{n} \sum_{i=1}^{n} (M_i - O_i)$$

$$NMB = \frac{1}{n} \sum_{i=1}^{n} \frac{(M_i - O_i)}{O_i}$$

$$r = \frac{\sum_{i=1}^{n} (M_i - \bar{M})(O_i - \bar{O})}{\sqrt{\sum_{i=1}^{n} (M_i - \bar{M})^2 \sum_{i=1}^{n} (O_i - \bar{O})^2}}$$

$$FAC2=\mbox{Fraction}$$
 of data where $0.5 \leq \frac{M_i}{O_i} \leq 2$

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$$NMSE = \frac{\overline{(O-M)^2}}{\overline{O} \cdot \overline{M}}$$

$$FB = \frac{\overline{O - M}}{0.5 \cdot (\overline{O} + \overline{M})}$$

$$NAD = \frac{\overline{|O - M|}}{(\bar{O} + \bar{M})}$$

$$RMSE = \sqrt{\frac{1}{n}\sum_{i=1}^{n}(M_i - O_i)^2}$$



Figure S2. Hourly time series of measured (temperature, pressure, wind direction, relative humidity, rainfall) and estimated (stability class) meteorological parameters during the first period (4 February - 7 March 2020) at OSS station.



Figure S3. Hourly time series of measured (temperature, pressure, wind direction, relative humidity, rainfall) and estimated (stability class) meteorological parameters during the second period (26 December 2020 - 21 January 2021) at OSS station.



Figure S4. Comparison of modelled (mod) and observed (obs) wind roses at CMP, OSS, DEX and POL stations for the two simulation periods: (a) first period (4 February - 7 March 2020) and (b) second period (26 December 2020 - 21 January 2021). Wind direction data from DEX station during the first period were excluded from the analysis due to a significant data gap and observed biases in the sensor readings.



Figure S5. Comparison of the estimated PBL height from soundings (blue) and CHIMERE simulations (orange) at the SPC station for the first simulated period (4 February - 7 March 2020) at 00:00 GMT. For qualitative comparison, the PBL height simulated by GRAL over the urban area of Modena is also included (green).



Figure S6. Comparison of the estimated PBL height from soundings (blue) and CHIMERE simulations (orange) at the SPC station for the second simulated period (26 December 2020 - 21 January 2021) at 00:00 and 12:00 GMT. For qualitative comparison, the PBL height simulated by GRAL over the urban area of Modena is also included (green).



Figure S7. Daily time series of measured PM_{10} and $PM_{2.5}$ concentrations at urban traffic and background sites for the first period, panel (a), and the second period, panel (b).



Figure S8. Skew-T Log-P plot from S. Pietro Capofiume station on 3 January 2021 on the first row (panels a and b) and on 4 January 2021 on the second row (panels c and d), at 00:00 GMT on the left, and at 12:00 GMT on the right. The red line represents the temperature of the atmosphere, while the green line represents the dewpoint.



Figure S9. Skew-T Log-P plot from S. Pietro Capofiume station on 13 January 2021 on the first row (panels a and b) and on 14 January 2021 on the second row (panels c and d), at 00:00 GMT on the left, and at 12:00 GMT on the right. The red line represents the temperature of the atmosphere, while the green line represents the dewpoint. 13



Figure S10. Skew-T Log-P plot from S. Pietro Capofiume station on 18 January 2021 on the first row (panels a and b) and on 19 January 2021 on the second row (panels c and d), at 00:00 GMT on the left, and at 12:00 GMT on the right. The red line represents the temperature of the atmosphere, while the green line represents the dewpoint.



Figure S11. Hourly time series of modelled Planetary Boundary Layer height by GRAL over the urban area of Modena for the second period.



Figure S12. Daily modulation profile of BC emission sources on working days (on the left) and holidays (on the right).

S6 Linear regression analysis between modelled and measured BC concentrations



Figure S13. Linear regression between modelled and measured BC concentrations for Biomass Burning-derived BC (left panels), Fossil Fuel-derived BC (central panels) and total BC concentrations (right panels). Data include concentrations for both time periods simulated in this study.

| $\begin{array}{c}\text{MAE at 880 nm}\\(\text{m}^2\text{g}^{-1})\end{array}$ | site | source | slope | $\underset{(\mu gm^{-3})}{RMSE}$ | $\underset{(\mu gm^{-3})}{MB}$ | NMB (%) | FAC2 (%) |
|--|------------------|--------|-------|----------------------------------|--------------------------------|------------|-------------|
| 10.12 | Urban traffic | FF+BB | 0.65 | 1.72 | -0.52 | -16 | 72 |
| | Urban traffic | FF | 0.56 | 1.22 | -0.68 | -28 | 59 |
| | Urban traffic | BB | 0.77 | 1.03 | 0.24 | 22 | 51 |
| | Urban background | FF+BB | 0.83 | 1.00 | 0.11 | 7 | 68 |
| | Urban background | FF | 0.49 | 0.63 | -0.32 | -28 | 58 |
| | Urban background | BB | 0.98 | 0.70 | 0.31 | 43 | 53 |
| 8.096 | Urban traffic | FF+BB | 0.81 | 2.15 | 0.18 | 5 | 72 |
| | Urban traffic | FF | 0.70 | 1.52 | -0.24 | -10 | 61 |
| | Urban traffic | BB | 0.96 | 1.28 | 0.58 | 53 | 46 |
| | Urban background | FF+BB | 1.03 | 1.26 | 0.52 | 34 | 62 |
| | Urban background | FF | 0.61 | 0.78 | -0.12 | -10 | 61 |
| | Urban background | BB | 1.23 | 0.88 | 0.57 | 78 | 45 |
| 12.144 | Urban traffic | FF+BB | 0.54 | 1.44 | -0.98 | -30 | 67 |
| | Urban traffic | FF | 0.47 | 1.01 | -0.96 | -40 | 51 |
| | Urban traffic | BB | 0.64 | 0.86 | 0.02 | 2 | 53 |
| | Urban background | FF+BB | 0.69 | 0.84 | -0.16 | -11 | 71 |
| | Urban background | FF | 0.41 | 0.52 | -0.45 | -40 | 55 |
| | Urban background | BB | 0.82 | 0.58 | 0.14 | 19 | 59 |

Table S3. Results of the linear regression (slope and RMSE) for the three scenarios analysed and statistical scores (MB, NMB and FAC2) from the comparison between modelled and measured BC concentrations.

S7 Fleet composition and BC traffic emission factors



Figure S14. Local Fleet Composition. The number of registered vehicles is normalised taking into account the estimated total kilometres travelled by each vehicle category per year.



Figure S15. BC emission factors (EF) of diesel and petrol passenger cars as a function of travelling speed, as implemented in the VERT R package used in this study to estimate exhaust vehicle emissions. EF function for Petrol Conventional and Euro 1 overlaps with that of Euro 2. The same is for the curves of Petrol Euro 3 and Euro 4.

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