We thank Reviewer 2 for his/her positive remarks, suggestions and hints, which allowed us to improve the overall quality of this study.

In the reply we used blue fonts for the reviewer's text, black fonts for the authors' reply and green italic fonts for the excerpts of the revised manuscript.

The manuscript by Bigi et al. attempts to bridge between in situ measurements (obtained through with multi-wavelength absorption photometry) and AERONET sunphotometer retrievals, in the air pollution hotspot of the Po Valley. Established modelling approaches are followed for the disaggregation of BC-BrC components and also for the apportionment of BC fossil fuel and biomass burning sources at urban traffic and background locations. The insights gained by the paper are important in terms of how much in-situ absorption monitoring translates into actual absorption aloft and whether it can be used for radiative forcing estimates. The characterization of the intra-urban variation of absorbing aerosol especially between different site types can lead also to useful inferences in the ambient exposure domain, thus, the authors are encouraged to expand towards the analysis of intersite associations/contrasts for in-situ source-specific BC and BrC. There should be also some attempt to discuss the transport of absorbing pollutants within or from outside the Po Valley. The paper is generally clearly written, with pertinent references and adequate discussion. It also recognizes uncertainties and limitations of the monitoring/modelling approaches used. It can be considered for publication after exploring the above mentioned directions and addressing the following specific comments.

Specific Comments

Introduction, 3rd paragraph: The health effects from exposure to BrC should also be mentioned, given that a substantial part of light-absorbing OC are of polycyclic aromatic nature and therefore linked to oxidative stress induction and carcinogenic effects.

We agree with the reviewer and in the Revised Manuscript (RM) we added the following statement (line 52): *BrC has also been shown to have detrimental health effects, enhanced because of its enrichment in organic compounds (Chowdhury et al., 2019; Offer et al., 2022), possibly related to aerosol aging (Li et al., 2022; Tuet et al., 2017; Weitekamp et al., 2020).*

l48-49: Given the existing ambiguity in the definition of eBC, this compilation should be recognized as a rather challenging task.

In the RM we changed these lines as follows (line 56): "*[...] the large uncertainty associated with source emission factors, PM speciation and eBC definition makes the implementation of systematic and harmonized emission estimates a challenging task.*"

l59-65: Why go into such details about these methods when they are not used in the present study?

While there are extra (and perhaps extraneous) sentences on the details about these methods, we consider it useful for the reader since there is quite a lot of active research on BC measurement techniques.

l101: Sources such as? Discuss.

In the RM the sentence was modified as follows (line 114): *Significant aerosol sources other than traffic remain present in the valley, e.g. biomass burning by domestic heating for several compounds including organic aerosols and BC, and farming for* NH_3*, a major PM precursor. Their role in PM levels was highlighted by the small decrease in PM across the basin (Ciarelli et al., 2021; Putaud et al., 2021) and in particle count in Modena (Shen et al., 2021) during the 2020 lockdown due to the SARS-CoV-2 pandemics.*

l132: Give some more details about the fuel types that are used for domestic heating and emit PM in the area. The impact of residential biomass burning should be introduced because BB aerosols are integral to this study.

In the RM more details regarding domestic heating emissions have been provided (line 153): *More specific to non-industrial combustion (SNAP 2), most of buildings use compressed natural gas for both heating and cooking; consistently 99.4% of* PM_{10} *emissions by SNAP 2 are estimated to be produced by biomass combustion for domestic heating (ARPAE, 2020).*

l134-136: This statement might be a bit ambitious, and taken out of context depending on how the reader understands the extent of the Po Valley. Moreover, the limited impact of industry that the authors claim for the Modena area, might not be the case in other more industrialized cities. I would suggest to tone this part down.

In the RM the sentence was changed according to the following (line 158): *Modena's setting is quite representative of several mid-size urban areas across the Po valley, particularly in terms of traffic and domestic emissions sources and topography.*

l142-143: Indicate the distance from the nearby road for the UB site, and the traffic intensity in the adjacent road for the UT site. It could be also useful to mention some inter-site differences observed for the regulatory pollutants, to illustrate how clear the UT-UB distinction is.

This information are now included in the RM (line 166): *The UT site faces a major road with two lanes per direction, with estimated median daily traffic counts of ~ 20 thousand vehicles, while the UB is within Modena's largest urban park at a distance of ~120 metres from the nearest road.* [...] *The daily* PM_{10} *median (10th, 90th quantiles) concentration at the UB site over the period 2017 – 2021 was 24 μg* m^{-3} *(13 μg* m^{-3} *, 57 μg* m^{-3}*), while at the UT sites the same statistics for* PM_{10} *resulted were 27 μg* m^{-3} *(14 μg* m^{-3} *, 63 μg* m^{-3}*). Consistently, over the same period, hourly* NO_2 *at the UB showed lower levels than at the UT site, with the two locations having a median (10th, 90th quantiles) of 23 μg* m^{-3} *(6 μg* m^{-3} *, 50 μg* m^{-3}*) and 35 μg* m^{-3} *(14 μg* m^{-3} *, 66 μg* m^{-3}*) respectively.*

l155: The aggregation procedure here is unclear. What is meant by "custom"?

We agree wth the reviewer that ‘custom’ is not a proper definition. The aggregation of the raw transmittance count from the MA200 is based on a transcription in R programming language of the dual-spot compensation algorithm presented by Drinovec et al. (2015). In the RM the text was changed as follows (line 179).

In order to compensate for the occasionally low absorption readings at the latter site, the 1-minute raw transmittance counts at UB were firstly aggregated to 5 minutes and then used to compute the corresponding σ_{ap} by a transcription in R programming language of the dual-spot compensation algorithm as described in Drinovec et al. (2015)

l157: A flow rate over 100cc is necessary to use the DualSpot compensation, but what factored in using different seasonal flow rates? Is there some recommendation by the authors?

The larger flow in summer is needed because of the lower atmospheric levels: if one wants to keep the same time resolution, flow needs to be increased to deposit sufficient material on the filter. In the RM the sentence was changed according to the following (line 182): *Flow was set to 100 ml min^{-1} in winter and increased to 125 ml min^{-1} in summer, because of the lower atmospheric concentrations.*

l210-211: Describe in brief the screening process.

We screened them referring to work done in section 2.4.1. Thus, as suggested also by reviewer 1, in the RM we now refer to this section regarding this screening issue (line 281).

Section 2.3: A separate uncertainty section is not necessary here. Mover this information to the respective sections.

In the RM the uncertainty of each dataset (in-situ observations, ERA5 model and columnar observations) is now included in the respective method section.

Section 2.4.1: I understand that the model has been presented already in literature, however, it is necessary to include a description of the procedure here and maybe some of the key equations. Otherwise, the reader will not understand how the different AAE values that are preselected are put into use.

In the RM the description of the in-situ and of the columnar absorption data is now expanded, as reported below.

line 295: *In-situ aerosol absorption coefficient $\sigma_{ap}(\lambda)$ was apportioned to species (i.e. Black Carbon, Brown Carbon, referred to as $\sigma_{ap,BC}(\lambda)$ and $\sigma_{ap,BrC}(\lambda)$, respectively) and sources (fossil fuel and biomass burning combustion, referred to as $\sigma_{ap,FF}(\lambda)$ and $\sigma_{ap,BB}(\lambda)$, respectively) using the Multi-Wavelength Absorption Analyzer model (MWAA model, Massabò et al., 2015; Bernardoni et al., 2017). This model assumes an equivalence between the Absorption Ångström Exponent (AAE, Moosmüller et al., 2009) of BC and that of fossil fuel ($AAE^{i}_{FF}=AAE^{i}_{BC}$), and it assumes biomass burning to be the only source of BrC. Under these hypotheses, the MWAA model assumes that both the following equations hold for the total $\sigma_{ap}(\lambda)$ at each wavelength:*

$$\sigma_{ap}(\lambda)= \sigma_{ap}\, BC(\lambda)+ \sigma_{ap}\, BrC(\lambda)= A\lambda^{-AAEiBC}+ B\lambda^{-AAEiBrC} \quad (2)$$

$$\sigma_{ap}\, (\lambda)= \sigma_{ap}\, FF(\lambda)+ \sigma_{ap}\, WB(\lambda)= A'\lambda^{-AAEiFF}+ B'\lambda^{-AAEiBB} \quad (3)$$

In Equations 2 and 3 $AAE^i_{BC}=AAE^i_{FF}=1$ was set, based on the AAE^i computed over 5 wavelengths at morning rush hour on winter weekdays at UT, consistent with fresh uncoated BC particles (e.g. Liu et al., 2018). AAE^i for BrC was determined by a preliminary non-linear fit of Equation 2, performed considering AAE^i_{BrC} as a free parameter (and resulting in an average $AAE^i_{BrC}=3.9$); $AAE^i_{bb}=2$ was set on literature data for the Po valley (Bernardoni et al., 2011, 2013; Vecchi et al., 2018; Costabile et al., 2017). A, B were then obtained for each sample by multi-wavelength fit of Equation 2 (after fixing AAE^i_{BrC}) and A', B' by multi-wavelength fit of Equation 3. It is noteworthy that MWAA hypotheses neglect possible contributions from mineral dust. To limit uncertainties resulting from this, the days with significant dust load were discarded prior the application of the MWAA model to the in-situ data, i.e. whenever the in-situ apportionment data is presented throughout the text, it is screened for dust.

line 321: *AAOD was apportioned to BC, BrC and mineral dust using the approach proposed in Bahadur et al. (2012), i.e. by directly solving the system of Ångström equations (see Appendix A) using the AERONET almucantar L1.5* retrievals. The system includes Equations A1, reporting the additive contribution of AAOD by each species to the total AAOD and Equations A2, reporting the exponential dependence of AAOD on the wavelength*.

l245: Do you consider that there are uncertainties around this assumption? For example, at a traffic impacted location, there could be some traffic-related BrC expected. This would also probably mean that AAE-BC and AAE-FF are not identical. Please discuss and recognize the limitations.

The model used to process data presented in this work is based on the hypothesis that $AAE_{BC}=AAE_{FF}=1$. The Referee is certainly raising an interesting and complex point, since it is possible that these two parameters have different values depending, for example, on the aerosol aging or even on the type of fossil fuel burned. However, the effect of their variation on both optical and source apportionments is of second order to the more critical parameters such as AAE_{BrC} and AAE_{BB}. The limitations related to the choice of these parameters (AAE_{BC}; AAE_{FF}) are the same connected to source apportionment methods based on optical properties. We strongly think that a sensitivity study able to evaluate the effects of the variation (and linked uncertainties) of these parameters is beyond the scope of the present work; a dedicated study is currently underway and will soon be proposed as an independent paper. It will also present and discuss a freeware version of a software that integrates an updated and extended version of the MWAA model.

l250: The selection of a fixed AAE-BrC value is critical for the calculations and it has to be justified better here. Provide more information on how the 3.9 value was derived (method, location, season, dominant sources etc.).

As now better explained at line 304 of the RM, 3.9 was the average value obtained on the dataset presented in this paper, running the MWAA model having AAE-BrC as a free parameter.

l292-301: I don't think that a whole paragraph introducing the Figures is necessary. You can

guide the reader through the presentation of the results.
We think that the dataset can be slightly tricky so a short recap paragraph is useful to present the analysis.

l302-303: Mean values are mentioned here, while in the Figure-Table, the medians are displayed. Maybe consider a homogenization of the presentation.
We agree with the reviewer, but since we compared the data to existing literature, depending on the referred article we had to compute the corresponding statistics on our dataset, in order to be comparable.

Figure 2: A couple of things stand out here and should be discussed. First, in the holidays the nighttime peak of BCff absorption at UB is comparable to the workdays, and also bigger than that at UT. Second, at the UT site, nighttime BCbb and BrC absorptions become larger in holidays than in workdays.
We agree with the reviewer that these are two noteworthy points and have been added in the RM at line 395: *More specifically, on weekday evenings* $\sigma_{ap,BC,ff}$ *peaks at 20:00 LT, one hour later than on holidays, at both UB and UT, with the former site recording* $\sigma_{ap,BC,ff}$ *levels higher in the evening that in the morning."* and lines 408: *"The weekly pattern for these two species is larger at the UT, with an increase during holidays in the overall median values of* $\sigma_{ap,BC,bb}$ *and* $\sigma_{ap,BrC}$ *of 22% and 35% respectively, along with an increase in their IQR of 16% and 28%. [...] The holiday increase in biomass burning aerosol is probably linked to the longer stay at home compared to weekdays and to a large recreational use of biomass burning in town*

l302-304: Is there an increasing interannual trend for absorption in cities of the Po valley? Discuss.
In the RM a note regarding Elemental Carbon trend is now included within the Introduction section (line 112). *Similarly, a drop of ~4% per year over the period 1997 – 2016 was recorded for the elemental carbon content in fog samples at the rural background site of San Pietro Capofiume (Gilardoni et al., 2020b).*

l321: It should be "BrC estimated".
In the RM that paragraph was rewritten and most of the text was moved to a table.

Table 2: The vehicle count parameter should be expressed in vehicles-per-day units.
We agree with the reviewer and in the RM Table 2 reports the statistics regarding the total number of vehicles per day.

l350-355: Not much new in this paragraph and not in the core of the study. I suggest omitting it or condensing it to a sentence.
As correctly suggested by the reviewer, we condensed this paragraph into the following sentence (line 419) *O_3 exhibits a 'weekend effect' (Cleveland et al., 1974), common to most urban areas in Europe having a VOC–limited regime, i.e. on holidays ozone rises earlier in the morning due to the lower NO_x levels, leading to a more efficient photocatalytic cycle, and drops later in the evening due to the (later) increase in NO_x.*

l380-387: The enhancement of BrC for winds of the southern direction should be explained, since in this study BrC is considered as a source-specific variable (BB-related). It can be observed that the winds related to the increase are only moderate. So it should be examined if there is indeed a BB source area or it is a low-wind stagnation effect during nighttime when the highest BrC levels are expected (it should be also noted that it is observed only in holidays).

It is very challenging to detect precisely the location of the source causing the increase in BrC at the UT site on holidays. The increase occurs on Sundays or holidays, consistently with the hypothesis of biomass burning from domestic heating for recreational use, and it occurred only in the evening/night and not during daytime: so the combination of domestic heating and the nighttime stagnation probably are both responsible for the increase in BrC. In the RM a short note about this was added (line 453): *Also at the UT $\sigma_{ap,BrC}$ is higher during holidays and under southerly winds. This latter increase occurs during evening/night hours (not shown), consistently with biomass burning from domestic heating for recreational use, with the increase probably enhanced by nighttime atmospheric stagnation.*

l396-398: Did you consider compensating for the wavelength discrepancy by adjusting in situ absorptions by the calculated absorption AAE?

We preferred not to compensate for relatively small wavelength differences, since the uncertainty in the AAE might introduce a larger error than the error proceeding from the direct comparison of two slightly different wavelengths.

Conclusions: The section repeats numerical results from the previous parts of the manuscript. Some more implications of the findings, regarding atmospheric absorption research and urban BC exposure should be added.

In the RM a further point regarding implications and outlooks was added (line 587): *An improved knowledge of the role by the in-urban emissions of LAA is critical to control local air quality, urban heat island effects and climate forcing and an apportionment of LAA based on their atmospheric levels, as presented here, contributes towards this goal. This study provides important insights on the role of the in-situ absorption monitoring in estimating the actual absorption aloft and whether it can be used for radiative forcing estimates. Moreover the characterization of the intra-urban variation of absorbing aerosol based on different site types contributes in the ambient exposure domain. Towards this latter outcome, a more in depth investigation of the contribution of urban areas to atmospheric LAA can be gained by the application of specific atmospheric dispersion tools, and this represents one of the major study outlooks. More specifically, Lagrangian particle dispersion models would provide information on atmospheric levels across the urban area at a fine spatial resolution, supporting advanced exposure studies, and, further, would give an estimate of the spatial- and time-resolved emission factors for LAA in the urban area.*

Technical corrections

l31: Delete “over the Earth”
In the RM the text was changed accordingly.

l33: Delete “depending…specifics”
In the RM the text was changed accordingly.

l43” That should be “increased eBC concentrations”. Same at next line
In the RM the text was changed accordingly.

l314: “good agreement”
In the RM the text was changed accordingly.

l400: “overestimation”
In the RM the text was changed accordingly.

l400: “by some concurrent conditions: (a)”
In the RM the text was changed accordingly.

l402: “at the rural…”
In the RM the text was changed accordingly.

l405: “contribution”
In the RM the text was changed accordingly.

l450: “mean absolute deviation”
We actually meant the “median absolute deviation”, the median of the absolute deviations from the data's median. We used this as a robust estimate of the variable dispersion, i.e. a robust

l479: “with Ispra and Modena”
In the RM the text was changed accordingly.