We thank all reviewers and the editor for their comments. Our answers, in normal text, are presented below the comments, which are bold. Changes to the manuscript are in italic.

REFEREE #3

The authors of the manuscript (acp-2022-156) have well addressed most of my comments in the last round of revision. However, there remain two comments that I highly recommend the authors consider addressing before publication. Once the comments are addressed, in my opinion, the manuscript is suitable to be accepted by the journal.

1. I still think the current introduction section is too general; instead, it should include more explanation about the variabilities of soot from different sources (the first paragraph). This is very important to the readers to understand why the authors focus on comparing absorption, size, and mixing state of BC between the sites.

As suggested we have significantly reworked the introduction by expanding the sections on soot formation/transformation (which explains the variability from different sources), health and measurement techniques as well as added a few references.

The full section now reads as follows:

Virtually everywhere in the world, a fraction of the ambient aerosol consists of soot. Soot is formed by incomplete combustion of carbonaceous fuels at hot air-starved conditions, and commonly contains both highly absorbing graphitic-like black carbon (BC) and organic carbon. It has severe effects on climate and human health (e.g. Bond et al., 2013; WHO, 2021; IARC, 2014; IPCC, 2021). Therefore, ambient measurements of soot concentrations and properties are of great importance to constrain and model its effects. Soot measurements are complicated by the fact that the ambient aerosol is a dynamic mixture, and that soot from different sources may not possess the same properties in terms of chemical content and nanostructure (Vander Wal et al., 2010; Malmborg et al., 2019), light absorption (Sandradewi et al. 2008), size (Schwarz et al., 2008), and toxicity (Hakkarainen et al., 2022).

A soot particle survive in the atmosphere approximately 5-9 days (Textor et al., 2006). During its lifetime several processes affects its properties. Soot is formed in a chain of steps, starting with the inception of the first condensed-phase particles from gas-phase hydrocarbon soot precursor species (Michelsen et al 2020). These then grow rapidly by coagulation and gas-to-particle conversion and become more graphitic, also making them more light-absorbing. These near-spherical 10-30 nm primary particles coagulate to form agglomerates that are emitted from the source to the surrounding air, if not removed by oxidation already in the combustion process or by exhaust after-treatment (e.g. particle filters). The properties of the emitted soot particles depend on the combustion conditions and fuel used. For example, it is well known that the wavelength dependence of light absorption is stronger for biomass burning BC than for traffic emissions (Sandradewi et al., 2008).

Once in the atmosphere, the freshly formed soot particles evolve from their initial agglomerated state, each consisting of large numbers of primary soot particles, into compacted soot cores with significant coatings of inorganic and organic material (Corbin et al., 2023). During such atmospheric ageing, the soot particles will not only increase their size and effective density, but also their ability to absorb light will typically increase (Zhang et al., 2018). Further, the particles change from being nearly hydrophobic into hygroscopic particles that can act as CCN (cloud condensation nuclei; Swietlicki et al., 2008) which increases the likelihood for wet removal which is the main deposition process for BC in the atmosphere (Textor et al., 2006). This transformation depends on atmospheric conditions and constituents but can be on the order of hours under favourable conditions (Eriksson et al., 2017).

Already in 2012, the International Agency for Research on Cancer, which is part of the World Health Organization (WHO), classified diesel engine exhaust as carcinogenic to. WHO further concluded that there is sufficient evidence for an association between short-term BC levels and all-cause and cardiovascular mortality, and cardiopulmonary hospital admissions [ref: REVIHAAP, 2013].Drawing similar conclusions as the WHO, the US EPA (2019) summarizes the associations between several health effects and BC concentrations in their impact assessment (USEPA, 2019). In its latest update of the Air Quality Guidelines, WHO did not yet

recommend air quality guidelines for black carbon (WHO, 2021). Instead, they made a statement of good practice recommending systematic measurements of black and elemental carbon, and production of BC/EC emission inventories, exposure assessments and source apportionment.

A myriad of BC measurement techniques exist, and the terminology is based on the measured property (Petzold et al., 2013). From light absorption techniques, the equivalent black carbon (eBC) mass concentration is obtained from the ratio of measured light absorption coefficients to the corresponding mass absorption cross-section (MAC). More recently, techniques based on laser-induced incandescence (LII) have been developed for measuring the refractory black carbon (rBC) mass concentration. For instance, the single particle soot photometer (SP2) (Schwarz et al., 2006; Stephens et al., 2003) deploys LII to measure the single particle rBC mass. In addition, the SP2 can be used to retrieve the rBC core size, mass-size distribution and provide a an estimation of the coating thickness of rBC particles(Moteki & Kondo, 2007).

Although BC mass concentrations are routinely estimated from optical methods, BC number concentrations, size distributions and mixing state are rarely measured. In a previous study it was shown that global aerosol microphysics models underestimate the BC particle size, by a factor of ~2-3, while overestimating the number concentrations, by more than a factor of 3, compared to airborne measurements using the SP2 (Reddington et al., 2013). BC from different sources have different size distributions (Schwarz et al., 2008; Laborde et al., 2013; Saarikoski et al., 2021), that affects both transport (lifetime) and light interactions (Hinds, 2012), as well as deposited dose (Alfoldy et al., 2009; Rissler et al., 2012). Recent studies have pointed to the importance of BC mixing state, governed by emission source and atmospheric ageing, in understanding the light absorbing properties, and hence climatic impacts, of BC containing particles (e.g. Liu et al., 2017; Liu et al., 2019; Fierce et al., 2020; Yuan et al., 2020). These properties of BC, that are crucial to understand both health and climate impacts, are not measured by the BC measurement techniques commonly used by monitoring networks.

The aim of this study was to compare BC particle properties in nearby urban and rural settings, and to investigate the influence of urban emissions on the rural background air, utilizing both filter-based absorption measurements and single particle LII. We compare BC mass and number concentrations, size distributions and mixing state. Furthermore, the relation to total particle number concentrations and chemically resolved PM1 are assessed. Trajectory analysis was used to assess the influence of long- and short-range transport.

New references added:

Corbin, J. C., R. L. Modini and M. Gysel-Beer: Mechanisms of soot-aggregate restructuring and compaction, Aerosol Science and Technology, 57, 89-111, 2023.

Michelsen, H. A., M. B. Colket, P. E. Bengtsson, A. D'Anna, P. Desgroux, B. S. Haynes, J. H. Miller, G. J. Nathan, H. Pitsch and H. Wang: A Review of Terminology Used to Describe Soot Formation and Evolution under Combustion and Pyrolytic Conditions, Acs Nano, 14, 12470-12490, 2020.

Petzold, A., J. A. Ogren, M. Fiebig, P. Laj, S. M. Li, U. Baltensperger, T. Holzer-Popp, S. Kinne, G. Pappalardo, N. Sugimoto, C. Wehrli, A. Wiedensohler and X. Y. Zhang: Recommendations for reporting "black carbon" measurements, Atmospheric Chemistry and Physics, 13, 8365-8379, 2013.

Textor, C., M. Schulz, S. Guibert, S. Kinne, Y. Balkanski, S. Bauer, T. Berntsen, T. Berglen, O. Boucher, M. Chin, F. Dentener, T. Diehl, R. Easter, H. Feichter, D. Fillmore, S. Ghan, P. Ginoux, S. Gong, J. E. Kristjansson, M. Krol, A. Lauer, J. F. Lamarque, X. Liu, V. Montanaro, G. Myhre, J. Penner, G. Pitari, S. Reddy, O. Seland, P. Stier, T. Takemura and X. Tie: Analysis and quantification of the diversities of aerosol life cycles within AeroCom, Atmospheric Chemistry and Physics, 6, 1777-1813, 2006.

US EPA (2019), Integrated Science Assessment for Particulate Matter, US Environmental Protection Agency, US EPA/600/R-19/188, Dec 2019.

Vander Wal, R. L., V. M. Bryg and M. D. Hays: Fingerprinting soot (towards source identification): Physical structure and chemical composition, Journal of Aerosol Science, 41, 108-117, 2010.

2. Add a time-series figure of BC and C4H9+ concentrations: The authors agreed that this could be a nice analysis, but decided not to add it to the current manuscript. Please add such a figure or explain why not adding. The figures can help readers understand the temporal variability of BC and HOA during a traffic plume. Adding the figure to the SI is fine.

We have added the figure suggested by the referee and caption below to help the readers understand how the plume analysis was performed. This is now referenced in the methods section.

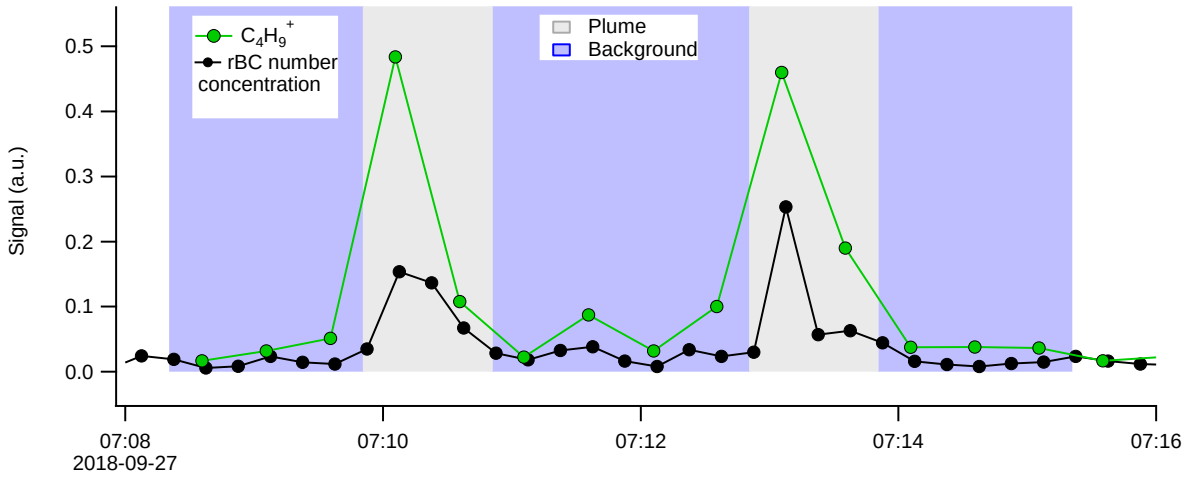

Figure S2. An example of typical plumes of rBC number concentration (measured by SP2) and signal of the HOA proxy mass fragment C4H9+ (measured by SP-AMS). Units are arbitrary. Plume mass spectra were isolated by subtracting the mass spectra of the background before and after the plumes.

REFEREE #5

The manuscript reports the particulate matter measurements in two sites, a rural and an urban, located in Sweden. The authors discuss the importance of the long range PM transportation. A large suite of instrumentation was used for the measurements of eBC, rBC, the chemical characterization of non-refractory PM and their size distributions. The authors also presented trajectory analysis results.

I believe that the authors have adequately addressed the comments made by the reviewers

Minor comment:

1) in several figures the y-axis needs to be corrected. More specifically in Figures 2, 5, 9

We have modified the figures as follows.

The error bars in Figure 2 was previously cut to better show the diurnal cycle. Since the std is symmetrical around the average all values can still be deduced from the original figure. However we have changed this now. New figure below.

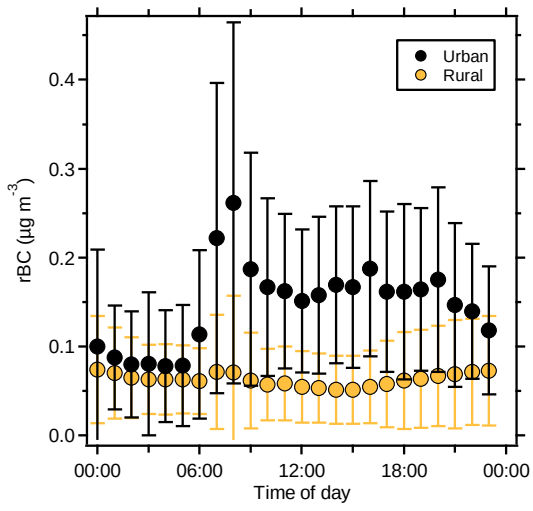

Figure 2.

In Figure 5 we clarified that the y-axis is the number concentration, and arbitrary units.

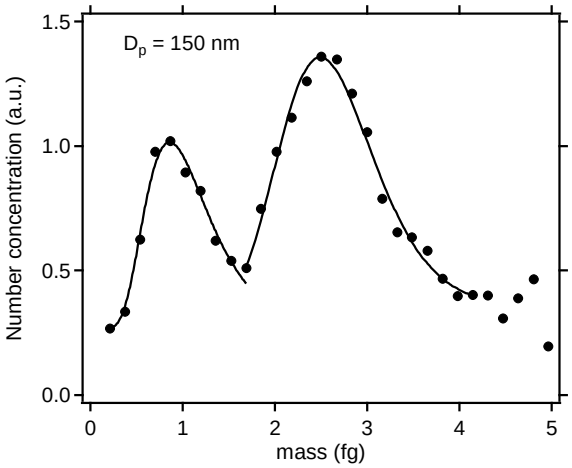

Figure 5.

In figure 9 we adjusted the y-axis of the insets to match and clarified that the AMS concentrations are normalized:

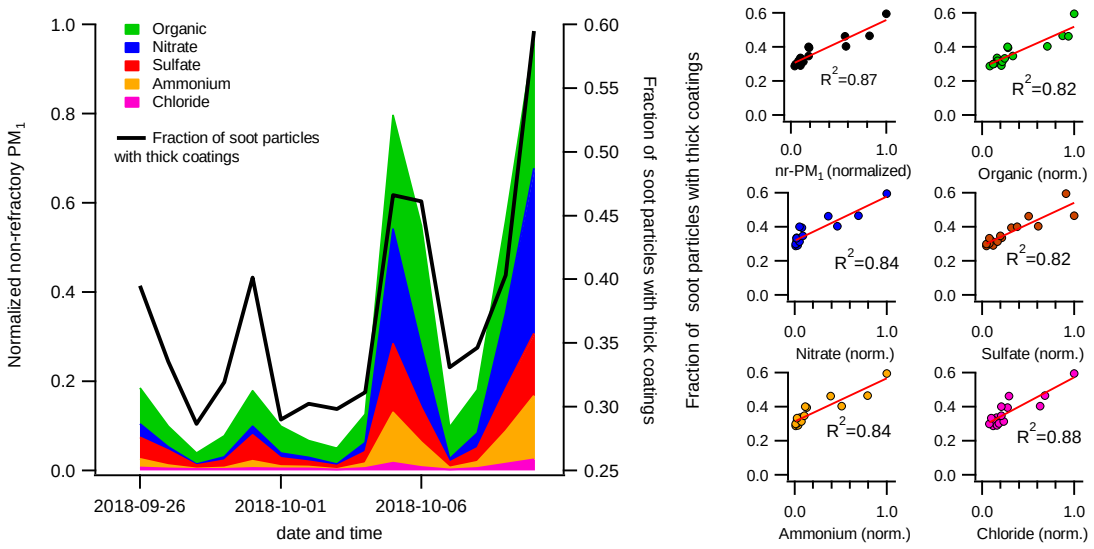

Figure 9.