Author response regarding the manuscript “From direct emission factors to inverse and indirect impact factors of road traffic: Influence of emerging and unregulated pollutants” by Savolainen et al.

Dear editor,

Thank you for considering our manuscript to be published in the Atmospheric Chemistry and Physics. The reviewers gave us constructive feedback which helped us to improve the discussion and clarity of our study and results. Our responses to the reviewers’ comments as well as the changes made in the revised manuscript are collected below. We hope that our responses and changes are satisfactory.

With best regards,

Teemu Lepistö,
Lauri Savolainen,
and the co-authors.

Reviewer #1

Savolainen and co-authors present in-situ measurements of air pollutants in a street canyon in wintertime Helsinki and investigate road traffic emissions and their impact on air quality. Measurement data evaluated in this study are from a five-week campaign in 2022, and emission factors (EFs) of particulate and gaseous pollutants were calculated based on linear regressions between geometric means of air pollutant concentrations and binned CO_2 mixing ratios. In addition to regulated air pollutants typically associated with road traffic emissions, the authors also investigate additional compounds such as organic and inorganic chemical components of particulate matter (PM), volatile organic compounds (VOC), and even compounds such as ozone, which is not directly emitted by traffic. Field-based measurements of EFs from road traffic are extremely valuable in order to be able to compare estimates representing real-world conditions with bottom-up estimates and regulatory requirements. Thus, the topic is very important and well within the scope of ACP. However, interpretation of the calculated EF metric and the so-called impact factors is at times misleading and many statements should be presented more carefully. My major comments are summarized in the following:

We want to thank you for the constructive feedback on our manuscript. We agree that field-based measurements of EFs are highly valuable, and we are glad to hear these positive thoughts regarding the importance of our study. Your comments definitely helped us to improve the manuscript, including clarifications to our methodology, discussion of results, and conclusions. Our responses to your comments are collected point-by-point below:

1) The methodology to calculate EFs presented in section 2.3 is based on the assumption that road traffic is the major source of CO_2 and the measured pollutants. However, the authors themselves state "that the method is susceptible to non-vehicle influences [...], such as regional wood combustion and long-range transported pollution" (l. 183/184). Could the authors give a quantitative estimate of the contribution of other sources to CO_2 mixing ratios at the site under wintertime conditions?

- We understand this concern, which is also the main reason why we decided to separate the non-episodic and episodic pollution episodes in the EF analysis. During the non-episodic periods, the contribution of other pollution sources to the measurement data at the street canyon was considerably lower than road traffic's. For example, the average diurnal CO_2 concentration followed very well the pattern of traffic rate towards south on the street (the measurement site is located next to the lanes towards south). During wintertime, the most likely pollution sources affecting the data would be the mentioned residential wood combustion and long-range transported pollution. The effects of these sources should, however, be clearly visible especially in measured BC and $PM_{2.5}$ concentrations. During the non-episodic periods, mean $PM_{2.5}$ and BC were 3.1 μg m^{-3} and 0.41 μg m^{-3}, respectively. Hence, the contribution of these sources was likely very low. As a comparison, during the episodic periods, the mean concentrations were 10.3 μg m^{-3} and 1.04 μg m^{-3}, respectively. Also, during the episodic periods, the diurnal pattern of CO_2 concentration shows clear increase in evening, suggesting contribution of residential wood-combustion.
- However, we acknowledge that other pollution sources contributed to the measured concentrations also during the non-episodic period. We unfortunately cannot give an accurate quantitative estimate of this contribution. Still, we think that our data clearly shows that road traffic was clearly the dominating factor during the non-episodic periods, and the effect of other sources on the determined EFs is likely minimal.
- To support our approach, we added:
    - New Fig. S2, showing the diurnal pattern of CO_2 and road traffic rate on the street during the non-episodic and episodic periods. Here, it can be seen that CO_2 mixing rate followed the diurnal pattern of road traffic rate (especially towards south).
    - New Fig. S4, showing the diurnal patterns of PN, BC and $PM_{2.5}$ together with CO_2 during the non-episodic periods. The patterns of all the mentioned parameters are close to the CO_2's one, further supporting the idea that the road traffic was indeed clearly dominating the measured aerosol.
    - New Table S4, showing the average CO_2, PN, BC, $PM_{2.5}$ and NO_x concentration during the non-episodic and episodic periods. Especially the low BC and $PM_{2.5}$ concentrations during the non-episodic periods strongly indicate that residential wood combustion or long-range transported pollution did not contribute considerably to measured concentration during the non-episodic periods.
- Changes in the manuscript text:
    - In 2.3.: *"On the other hand, road traffic should clearly be the most significant local contributor to the measured concentrations at the supersite (e.g., Saarikoski et al., 2021). To focus on the effects of road traffic, the data measured during the mentioned pollution episodes (section 2.1) was not considered in sections 3.1-3.3.* ***The similar diurnal patterns (Fig. S2) of measured CO_2 concentration and traffic rate (towards south) support the assumption that the effect of other sources than road traffic on CO_2 during the non-episodic periods was minimal. For example, weather conditions (temperature and wind speed), did not follow similar diurnal pattern (Fig. S3). The diurnal patterns of PN, BC and $PM_{2.5}$ were similar to the CO_2's one (Fig. S4), and BC and $PM_{2.5}$ concentrations were low (3.1 μg m^{-3} and 0.41 μg m^{-3}) during the non-episodic periods (Table S4), further supporting that the contributions of e.g., residential wood combustion and long-range transported***

pollution were low during these periods. In general, it should be noted that the presented results represent only the average weather conditions during the campaign. *The effect of the pollution episodes is investigated in section 3.4."*

- In 3.4.: *"In this study, the non-episodic EFs and IFs identified can be considered to accurately represent the effects of wintertime road traffic in Helsinki central area,* ***as CO_2 was well connected with the road traffic rate****, the regional pollution levels during the non-episodic periods were low and the linkages of studied pollutants were strong with CO_2* ***(Fig. S2-4, Table S4). Still, it is good to note that other urban pollution sources can have slight effects on the results. Also, weather conditions during the campaign likely influenced especially the parameters that are strongly dependent on the atmospheric chemistry, such as O3. Hence, it should be noted that the results represent the situation at the studied location during the average conditions of the measurement campaign.****"*

2) While it is not explicitly stated in the manuscript, the emission ratio ERp in equation 1 (l. 156/157) is probably derived from the slope of the linear regression fit between geometric mean values of pollutant concentrations and CO_2 mixing ratios in 5 ppm CO_2 mixing ratio bins. When averaging pollutant concentrations in CO_2 mixing ratio bins, the change in pollutant concentration per change in CO_2 mixing ratio is not only related to emissions from a common source but there could also be non-causal correlations, for example, simply due to related diurnal cycles of the pollutant concentrations and CO_2 mixing ratios. I expect that CO_2 mixing ratios exhibit a clear diurnal cycle, and one would probably obtain rather good linear fits with high R^2 values for many atmospheric variables not related to road traffic emissions, e.g. meteorological variables such as air temperature and wind speed. Therefore, R^2 values of the averaged linear fits close to 1 do not necessarily indicate that the pollutants are mainly derived from road traffic, as stated e.g. in l.209-211 ("R^2 values of the averaged linear fits were above 0.93, showing that the pollutants were mainly derived from road traffic at the studied site"). This is a major limitation of the study, and interpretation of the calculated EFs must be presented in a more careful way.

- It is true that conditions affect CO_2 mixing ratios. However, as mentioned in the response to the previous comment, the diurnal variation of CO_2 shows that road traffic was the dominating factor on CO_2 during the non-episodic periods. Temperature and wind speed did not follow similar diurnal pattern as CO_2 and the traffic rate (see new Fig. S2-3). Both, temperature and wind speed increased during the day, but much later (around noon) than CO_2 and traffic rate (6-7 am). Also, temperature and wind speed did not have similar drop between hours 10 and 16 as CO_2. Hence, strong correlations in the averaged linear fits should not be possible without a clear contribution of road traffic on the studied pollutant. For example, in Fig. S4, the PN, BC and $PM_{2.5}$ concentrations follow similar diurnal pattern as CO_2 (and road traffic).
- However, as mentioned, conditions affect the measured concentrations. Even though the impact of weather conditions on CO_2 and air pollutants can be similar in some cases (e.g., dilution), the weather conditions can also affect, e.g., chemical reactions, particle aging processes and, thus, change the determined EFs. So, in the revised text we want to emphasize that our results represent the average conditions during the non-episodic periods.

- Changes made:
    - It is now clarified that ER_p is derived from the slope of the averaged linear fit: *"The linear fit was applied for the averaged data to determine emission ratios for each pollutant **based on the slope of the fit** in the CO_2 range of 420–465 ppm, representing 99.2 % of the non-episodic data."*
    - Added Fig. S3, which shows the diurnal pattern of temperature and wind speed compared to CO_2. The new figure is also mentioned in the text (see response to previous comment)
    - We emphasize that the results represent only the average conditions during the campaign (see response to previous comment)
    - To further demonstrate the impact of traffic on the studied compounds, we added additional figures in the supplement, showing the diurnal patterns of PN, BC, $PM_{2.5}$ (Fig. S4, see the previous comment), NO_x and O_3 (Fig. S11, see later comments)

3) Related to my first two comments, there are several instances when the authors acknowledge that the CO_2-based EF calculation can be difficult to interpret, e.g.:

l.483/484: "Thus, even though CO_2 is generally a good tracer for road traffic, the CO_2-based EF calculation could be challenging if there are other major sources of the studied pollutants and CO_2 nearby."

l.487-489: Even though the results of this study show that CO_2 EF-calculation is a good method to observe the direct, and indirect, effects of road traffic, the mentioned uncertainty related to other contributing pollution sources should generally be better acknowledged."

l.491/492: "…based on the episodic data, one could misinterpret the data to conclude road traffic as a clear source of sulfate."

l.539/540: "…the data measured during the pollution episodes showed that the method is also vulnerable to misinterpretations if background concentrations or effects of other sources cannot be recognized."

However, the mentioned limitations are not considered throughout the manuscript when interpreting results and there are many instances when the authors discuss their results without acknowledging these challenges, e.g.:

- Thank you for this comprehensive comment. In our responses to comments 1-2 we have clarified the impact of road traffic on the measured CO_2 as well as discussed the contribution of other contributing factors. Hence, we are confident that these mentioned limitations do not considerably affect our results during the non-episodic periods. We have collected our point-by-point responses to the points below.

l.281/282: "Despite this challenge, strong connection between $LDSA^{al}$ and CO_2 shows the clear contribution of road traffic on the metric." - The good averaged linear fit does not necessarily indicate that particles contributing to $LDSA^{al}$ are mainly derived from road traffic.

- Regarding the contribution of road traffic to $LDSA^{al}$, the observations related to other particle metrics, such as PN and BC, strongly suggest that $LDSA^{al}$ is strongly contributed by road traffic, as $LDSA^{al}$ is known to be well connected with PN and BC concentrations. In the revised text, we have provided an additional explanation for this statement.
- Changes made:
    - "*Despite this challenge, strong connection between $LDSA^{al}$ and CO_2 shows the clear contribution of road traffic on the metric* ***which is also supported by the road traffic's contribution to PN and BC.***"
    - "*$LDSA^{al}$ is especially dependent on the ultrafine particles* ***(Lepistö et al., 2025)****, which deposit efficiently in the lung alveoli, as well as on BC* ***(Järvinen et al., 2019; Lepistö et al., 2022).***"

Järvinen, A., Timonen, H., Karjalainen, P., Bloss, M., Simonen, P., Saarikoski, S., Kuuluvainen, H., Kalliokoski, J., Dal Maso, M., Niemi, J. V., Keskinen, J., and Rönkkö, T.: Particle emissions of Euro VI, EEV and retrofitted EEV city buses in real traffic, Environmental Pollution, 250, 708–716, https://doi.org/10.1016/j.envpol.2019.04.033, 2019.

Lepistö, T., Kuuluvainen, H., Lintusaari, H., Kuittinen, N., Salo, L., Helin, A., Niemi, J. V., Manninen, H. E., Timonen, H., Jalava, P., Saarikoski, S., and Rönkkö, T.: Connection between lung deposited surface area (LDSA) and black carbon (BC) concentrations in road traffic and harbour environments, Atmospheric Environment, 272, 118931, https://doi.org/10.1016/j.atmosenv.2021.118931, 2022.

l.415/416: "...whereas the concentrations of intermediate ions (3-7 nm, Tammet, 1995) and larger ions increase as a function of increased traffic."

- Regarding the effects of road traffic on ions, we want to emphasize Fig. S13 (old Fig. S8), which shows that the changes in the size-resolved ion concentrations follow the traffic rate: The concentration of the smallest ions is the lowest during traffic rush hours (6-10) and (14-18) whereas the concentrations of the intermediate and larger ions are the highest during those periods. So, if this phenomenon would have been driven only by some other processes/pollution sources, we would not expect to see that clear agreement between the ion concentrations and the traffic rates. Hence, we believe that this statement is reasonable even though we understand the general uncertainties of ion measurement as well as our method (discussed in earlier comments).
- Changes made:
    - We rephrased the statement: "***..., whereas the intermediate ions (3-7 nm, Tammet, 1995) and larger ions have positive IFs, i.e., their concentrations increase as a function of increased traffic****"*
    - We mention the agreement between IFs and diurnal ion concentration trends: *"The road traffic's inverse contribution to small ions can also be seen when examining the*

*diurnal ion concentration variation (Fig. S13), where the concentration of small ions is reduced during rush hours compared to less trafficked hours**, supporting the observation of negative IFs**."*

l.430-432: "Overall, the analysis of the small ion concentrations as a function of CO_2 enables in-detail detection of the effects of road traffic as a sink for small ion concentration. By combining this analysis with size-resolved EF of particle number, the method could be potential for estimating the production and removal processes of ions in urban environments." - In my opinion, "enables in-detail detection of the effects of road traffic" is an overstatement.

- This is a good point, and we agree that "in-detail detection" is a too positive claim.
- Changes made: *"Overall, the analysis of the small ion concentrations as a function of CO_2 enables **estimation** ~~in-detail detection~~ of the effects of road traffic as a sink for small ion concentration."*

l.441/442: "However, the results suggest that terpenoids could be more directly linked to road traffic than previously considered." - I don't see any evidence for this statement.

- We changed the statement: *"However, the results suggest that terpenoids could be ~~more~~ directly linked to road traffic as well. ~~than previously considered~~"*

l.445/446: "As limonene is found in many cleanser products, the correlation with the CO_2 and road traffic could potentially be related to the number of recently washed vehicles on the street and the use of windshield washer fluid." - There are other, probably simpler explanations. For example, with wood combustion as a potential source, both terpenoids and CO_2 would be co-emitted.

- As discussed regarding the earlier comments, the effect of wood combustion was low during non-episodic periods. Even though occasional effects of wood combustion cannot be completely ruled out, we would not expect to see this strong a correlation in the averaged linear fits if the source of limonene would only be the wood combustion. We agree that there can be other potential (road traffic related) reasons behind the correlation between CO_2 and limonene, but we think that the car cleaning products could be one potential explanation, which is why it is mentioned.

l.446-449: "As the contribution of natural sources in this study should be minimal, it is likely that road traffic contributes directly also to the other terpenoids. Hence, the results indicate that road traffic could have previously unconsidered direct effects on terpenoid concentrations in urban environments." - I don't see evidence that road traffic "likely" contributes directly to emissions of terpenoids. Also, road traffic effects on terpenoid concentrations have been considered in previous studies (e.g. Borbon et al. 2023, JGR 128, e2022JD037566. https://doi.org/10.1029/2022JD037566).

- We modified the discussion related to the impacts of road traffic on terpenoids.

- Changes made: *"However, the results suggest that terpenoids* ***could be directly linked to road traffic as well****. In previous source apportionment studies, some fractions of terpenoids, especially isoprene and α-pinene, have been included in factors linked to traffic and combustion (Borbon et al., 2001; Hellén et al., 2012; Xiong et al., 2020; Zhang et al., 2020).* ***Also, Borbon et al. (2023) have reported the connection between road traffic and monoterpenes, but,*** *overall, the mechanism linking road traffic to terpenoid emissions remains poorly understood****, as, in addition to tailpipe emissions, other contributing factors are likely.*** *For example, as limonene is found in many cleanser products, the correlation with the CO_2 and road traffic could potentially be related to the number of recently washed vehicles on the street and the use of windshield washer fluid. As the contribution of natural sources in this study should be minimal****, the results indicate that road traffic has direct effects on terpenoid concentrations in urban environments. Further studies on the terpenoid IFs in other environments and conditions could, hence, be beneficial to understand the mechanisms and atmospheric implications of road traffic influenced terpenoids, as they can strongly contribute to the reactivity of anthropogenic VOC emissions in urban areas (Borbon et al., 2023).***

l.525: "For example, it was found that in cases of high CO_2 concentration, i.e. high traffic volume, NO2 concentration remained constant regardless of increasing CO_2 and the increase of NOx was almost completely contributed by NO. This phenomenon was linked to near-zero O_3 concentration which caused that NO was no further oxidized to NO_2. The approach could, hence, be useful when estimating NO/NO_2 concentrations in urban environments." - Again, in the first sentence, high CO_2 concentrations are equated with high traffic volume. However, elevated CO_2 concentrations might be observed in shallow boundary layers, for example at nighttime, when traffic volume is typically lower than during daytime. The observed phenomenon is consistent with the well-established diurnal cycle of nighttime ozone titration and daytime photochemical formation but I don't see an advantage in calculating the CO_2-based EFs to study the interactions between NO, NO_2 and O_3.

- As discussed earlier, we are confident that CO_2 was dominated by road traffic during the non-episodic periods. It is true that weather conditions related to the boundary layer height likely affect e.g., the dispersion and dilution of pollutants. So, it is good to note that our results represent only the situation during the average conditions and in this particular location, as mentioned earlier. Regarding O_3, we want to emphasise, that the diurnal variation of O_3 concentration, together with the strong correlation with CO_2 in the averaged fits, shows that road traffic indeed was the main (inverse) contributor to O_3 concentration. This average diurnal pattern of O_3 is shown in new Fig. S11. So, we disagree that this phenomenon would represent only the diurnal cycle of nighttime ozone titration and daytime photochemical formation.
- Regarding the benefits of our method, see our responses to later comments.
- Changes made: In 3.2.1.: *"In Fig. S11, the average diurnal pattern of O_3 further supports the idea of inverse contribution of road traffic on O_3 as the hours with lowest concentrations match well with the traffic rush hours."*

l.530-532: "Even though terpenoid VOC concentrations have been earlier connected with human activity, the results indicate that road traffic could also have direct effects on these compounds,

which is not generally well understood." - In my opinion, the presented data support several previous studies but it is not really additional evidence for direct road traffic emission of these compounds. For example, wood combustion would co-emit terpenoid VOCs and CO_2 as well.

- As mentioned earlier, the effect of wood combustion was low during the non-episodic periods. Hence, we are confident that road traffic was indeed the major contributor on the VOCs that had a strong correlation with CO_2 in the averaged linear fits.
- Also, as mentioned earlier, we modified the discussion related to the terpenoids in section 3.3.3.
- Changes made: We modified the mentioned lines in Conclusions: *"Furthermore, the contribution of road traffic to compounds that generally have not been considered to be traffic-influenced was found. Positive IFs of road traffic were found for terpenoid VOCs, especially α-pinene and limonene.* ***Terpenoid VOC concentrations have been earlier connected with human activity and traffic, but the connection is not completely understood. Hence, the results further support the idea that road traffic could have direct effects on these compounds.****"*

4) The concept of the so-called impact factor (IF) used for assessing indirect effects of road traffic on air quality is somewhat confusing. While the authors acknowledge that the concept of $LDSA^{al}$ EF is tricky (l.278), it becomes even more difficult when looking at ozone IF. Clearly, a change in O_3 mixing ratios is not dependent only on the kg of burned fuel but also on solar radiation and on mixing ratios of NO, NO_2 and VOCs. In that sense, the IF indicates the relationship between ozone and CO_2 (as a proxy for road traffic) while averaging all other influencing factors. This should be clearly stated. Also, the authors should carefully revise the manuscript to change instances when they refer to EFs for ozone (e.g. l.86/87, l.91/92).

- This is a good point. As discussed earlier, we now mention that the results represent only the situation during the average conditions and location of these measurements.
- We carefully checked that EF term is not used when discussing results related to O_3 or small ions.
- Changes made: In 3.4., we further emphasise the fact that weather conditions etc., affect the results: "***Also, weather conditions during the campaign likely influence especially the parameters that are strongly dependent on the atmospheric chemistry, such as*** O_3***. Hence, it should be noted that the results represent the situation at the studied location during the average conditions of the measurement campaign.****"*

Regarding the statements "In addition to direct EFs, we show that CO_2-based EF determination enables detection of inverse and indirect effects of road traffic" (l.24/25) and "Also, the findings related to the terpenoid VOCs, together with the observed inverse IFs on O_3 and small ions, show that the CO_2-based EF calculation is an effective approach to investigate not only the direct emissions but also the indirect and unconventional effect of road traffic on urban air" (l.534-537), the authors must clearly show and better explain what the advantage of the CO_2-based EF/IF calculation is. What are the main advantages of this particular approach?

- Regarding the first statement in the abstract, we decided not to do changes as we think that the manuscript text (especially conclusions) explain the reasoning behind it.
- About the second one, we want to emphasize that the method enables better estimation of the magnitude of road traffic's effect on these inverse effects. Previous studies have shown, e.g. that concentrations of O_3 and small ions tend to drop close to traffic sites. Our method further enables the estimation these effects directly linked to road traffic rates, i.e., per kg of fuel burnt. That information is relevant, e.g., regarding air quality modelling, where concentrations of air pollutants can be modelled based on traffic rate data among other data. We have clarified this reasoning in Conclusions.
- Changes made: "*In addition to direct EFs, we showed that the CO_2-based EF-calculation* ***can be beneficial*** *when investigating inversely proportional impact factors (IF) related to road traffic. Concentrations of O_3 and small ions decreased linearly as a function of increasing CO_2. Even though the negative contributions of road traffic to these metrics have been reported before, the utilized approach can be fruitful in terms of air quality modelling* ***and when estimating the magnitude of these effects with respect to traffic rates*** *as well as formation and removal rates of atmospheric gases and ions. For example, it was found that in cases of high CO_2 concentration… …The approach could, hence, be useful when estimating and modelling NO/NO_2 concentrations in urban environments.* ***As the inverse effects of road traffic are likely location and condition dependent, the presented approach to calculate IFs would be a rather simple method to estimate these effects in different circumstances***"

5) In the beginning of section 2.3 in l.150-152, the statement that the "EF for each pollutant metric was calculated […] with a time resolution of 1 min" is misleading. Later in l.173, it is explained that in "case of the DMPS, NAIS, SP-AMS, and VOC sampling, the measurement intervals were longer than 1 min". It is important to keep in mind different averaging intervals when interpreting results throughout the manuscript. Indeed, the time resolution defines whether or not dynamic emission processes can be resolved, and especially in urban environments, high time resolution is required. For example, Lamprecht et al. (2025; Atmos. Meas. Tech., 18, 5003–5016, 2025, https://doi.org/10.5194/amt-18-5003-2025) show that normalized enhancement ratios based on excess mixing ratios of NOx and CO_2 can strongly differ for different averaging intervals.

- Thank you for this note, we see that the statement in l. 150-152 was misleading. We agree that the measurement time-resolution is very important when measuring at a road traffic site due to the rapid changes in pollutant concentrations. However, because of measurement methodology, 1 min resolution is not possible with DMPS, NAIS, SP-AMS and VOC data.
- Changes made in 2.3:
    - "*EF for each pollutant metric was calculated by comparing the measured pollutant concentration with the concurrently measured CO_2 with a time resolution of 1 min* ***as a default***"
    - ***"The longer measurement intervals should be noted when comparing the measurements between different instruments as the instruments may not detect the rapidly varying concentrations caused by the road traffic (e.g., Lamprecht et al., 2025)."***

6) When comparing the calculated PN EF of approximately 2.1E14 #/km and the EU EURO standards for primary particle emissions (> 23 nm) of 6.0E11 #/km, the discrepancy is briefly explained as "likely related to the emissions of (semi-)volatile compounds" (l.227). I recommend to clarify that the EURO emission standards are defined for solid, non-volatile particles, while the ambient measurements include both volatile and non-volatile particles. In addition, the PN measurements in this study include particles > 5.4 nm. Only with a measurement setup separating volatile and non-volatile particles (e.g. using a thermodenuder), and covering the diameter range > 23 nm (e.g. from the DMPS data), a direct comparison would be reasonable.

- We have further clarified that the EURO standards are defined only for primary emission particles. We want to emphasize that this comparison is only indicative, and the purpose is to point out that unregulated (semi-)volatile particles (including the < 23 nm ones) have significant effect on PN in urban areas.
- Changes made: "*...the PN EF would be approx. 2.1 · 10^{14} # km^{-1}, representing considerably higher EF than the limit for primary emission particles (> 23 nm) utilized by the EU in the EURO standards (6.0 · 10^{11} # km^{-1}).* ***It is important to note that this difference*** *is likely related to the emissions of (semi-)volatile compounds* ***and particles smaller than 23 nm, which are not regulated in the EURO standards."***

7) When looking at the size-resolved PN EFs of the DMPS and NAIS instruments in Figure 2, I consider the "shapes of the EF size distributions determined from the DMPS and NAIS data seemed to agree well" (l.235/236) an overstatement. Also, why is the observed difference for particles < 30 nm expected to be due to the "different scanning times of the measurements" in particular (l.238)?

- We decided to rephrase this statement. We think that the different scanning times is likely one partial reason for the differences between the instruments as the higher time resolution of NAIS measurement likely detects better the rapid changed of particle concentrations contributed by road traffic.
- Changes made: *"The EF size distributions determined from the DMPS and NAIS* ***data both show the high contribution of particles smaller than 20 nm, even though the EFs from the NAIS data were higher compared to the DMPS.*** *This difference is likely related to the varying measurement methods* ***as well as the different scanning times of the measurements: The higher time resolution of NAIS measurement likely detects better the rapidly varying concentrations caused by the road traffic."*

8) In l.500-503, the authors state: "In this study, the non-episodic EFs and IFs identified can be considered to accurately represent the effects of wintertime road traffic in Helsinki central area, as the regional pollution levels during the non-episodic periods were low and the linkages of studied pollutants were strong with CO_2." - I agree that non-episodic periods were selected when regional pollution levels were low but as mentioned before, good correlation with CO_2 does not necessarily indicate road traffic as the dominant source. Obviously, the differences in magnitude of EFs derived during pollution periods and non-episodic periods suggest that factors other than road traffic can affect the results. Why should non-episodic periods be representative for road traffic only?

- As mentioned earlier, the general low concentrations of many pollutants during the non-episodic periods indicate that the effect of non-traffic related sources was low during these periods. Furthermore, diurnal pattern of CO_2 strongly followed the diurnal pattern of road traffic (as did the studied pollutants as well). Hence, we are confident that the results during the non-episodic period indeed represent mainly the effects of road traffic. Of course, in an urban area, we cannot completely rule out other pollution sources, but the results strongly indicate that the road traffic was clearly the dominating factor during the non-episodic periods. Please, check our responses to earlier comments for details (and made changes).

9) In l.521/522, the authors state: "In addition to direct EFs, we showed that the CO_2-based EF-calculation is suitable when investigating inversely proportional impact factors (IF) related to road traffic." How exactly did the authors show the suitability? They further state in l.524/525 : "...the approach utilized can be fruitful in terms of air quality modelling and when estimating the formation and removal processes of atmospheric gases and ions." How exactly could this approach be utilized? Do the authors suggest to use the calculated IFs in model parameterizations? I doubt that the calculated IFs will be transferrable to other seasons and conditions or even other sites.

- In the same paragraph we have mentioned *"Concentrations of O_3 and small ions decreased linearly as a function of increasing CO_2"*. We think that this example shows the benefits of using CO_2-based EF calculation for these metrics as well. As mentioned in an earlier response, it has been known that road traffic can have inverse effects on the concentrations of O_3 and small ions, but the magnitude of this effect as a function of traffic rate could not have been directly determined based only on the changes in the concentrations. The CO_2-based EF calculation directly links these concentration changes with road traffic and fuel consumption. This information could be highly beneficial in air quality modelling as mentioned in the text.
- We definitely agree that the presented IFs are not likely directly transferrable to other seasons, conditions or sites. To accurately implement IFs in models, more data is needed. Our point here is that the method itself is novel and beneficial, and it is rather easy to implement with already existing air quality monitoring data. Hence, the data required by the models should be achievable by utilizing our approach.
- Changes made: *"In addition to direct EFs, we showed that the CO_2-based EF-calculation* ***can be beneficial*** *when investigating inversely proportional impact factors (IF) related to road traffic. Concentrations of O_3 and small ions decreased linearly as a function of increasing CO_2. Even though the negative contributions of road traffic to these metrics have been reported before, the approach utilized can be fruitful in terms of air quality modelling and when estimating* ***the magnitude of these effects with respect to traffic rates as well as*** *formation and removal rates of atmospheric gases and ions. For example, it was found that in cases of high CO_2 concentration... ...The approach could, hence, be useful when estimating and modelling NO/NO_2 concentrations in urban environments.* ***As the inverse effects of road traffic are likely location and condition dependent, the presented approach to calculate IFs would be a rather simple method to estimate these effects in different circumstances.***"

Additional minor comments:

In l. 59/60, the authors state that BTEX group VOCs (benzene, toluene, ethyl benzene, and xylenes) have been recognized as carcinogenic compounds. Indeed, benzene and ethylbenzene are classified as carcinogenic by the International Agency for Research on Cancer (IARC), while toluene and xylene are categorized in IARC Group 3: Not classifiable as to its carcinogenicity to humans.

- Thank you for pointing this out. We have clarified the statement in the revised text.
- Changes made: "*Some of the VOCs are also considered directly harmful**: For example, benzene is classified as a carcinogenic compound by the International Agency for Research on Cancer (IARC, 2026).**"*

In l. 105, regarding the dilution of pollutants at the measurement site, it would be interesting to add information about the orientation of the street, e.g. with respect to the main wind direction.

- Thank you for this good suggestion. We added map of the measurement site together with a wind rose plot in the supplement (new Fig S1).
- Changes made: In 2.1.: *"The map of the measurement street together with a wind rose plot during the campaign is shown in Fig. S1."*

In Table 1 (l. 215), the EF values of NO and NO_2 indicate emission of 1.98 g NO and 2.41 g NO_2 per kg fuel, i.e. a total of 4.39 g for the sum of NO and NO_2, while the NOx EF indicates emission of 5.59 g NOx per kg fuel. Please discuss this discrepancy.

- This discrepancy can be explained by the air quality monitoring guidelines where NOx concentration is reported based on the molecular weight of NO_2. Hence, to obtain NOx mass concentration from NO and NO_2 measurements, the NO mass concentration needs to be multiplied by the factor of 1.53 (the ratio between molar masses of NO_2 and NO: 46/30). Hence, the NOx equivalent sum of NO and NO_2 EFs is: 1.98 g/kg_{fuel} * 1.53 + 2.41 g/kg_{fuel} = 5.44 g/kg_{fuel}, which is close to the reported NOx EF. This slight difference can be explained by the EF calculation method, where the fits for each pollutant are calculated separately.
- Changes made: We added explanation for this result: ***"It should be noted that the NO_x EF in Table 1 has been calculated based on the NO_2 molecular weight, i.e., NO is converted into NO_2 (according to air quality standards), explaining the difference in comparison with the summed EFs of NO and NO_2."***

In l.224, for conversion of the EF values from units "per kg fuel to "per km", fuel consumption of 11.0 l/100 km is assumed. What is the reasoning for this particular estimate?

- This conversion is used for indicative estimation of the obtained EFs in the unit of "per km" which is used in regulations. In the original text, it was discussed in 3.1.2, that the estimation is reasonable, because the bottom-up emission inventory-based EFs agreed well with the obtained EFs with this estimate. But, indeed, this estimation is only for indicative comparison purposes and the real average fuel consumption on the street cannot be accurately

determined. The text in 3.1.2: *"The bottom-up emission inventory-based NO_x EF was 0.545 g km^{-1} at the studied street canyon. This result is close to the obtained CO_2 based EF, which was 0.522 g km^{-1} with the fuel consumption of 11 L per 100 km. Together with the emission inventory-based $PM_{2.5}$ EF, the results show that the CO_2 based fleet EF determination agreed well with the emission inventory-based calculations. Also, the results suggest that the estimated average fuel consumption of 11 L per 100 km is reasonable at the studied street canyon."*

- Changes made: We clarify that the estimation is indicative: *"**For an indicative estimation**, if fuel consumption of 11.0 L per 100 km was assumed (i.e., 10.7 kg km^{-1}, with estimated fuel density of 0.85 L kg^{-1}), the PN EF would be approx. 2.1 · 10^{14} # km^{-1}, representing considerably higher EF than the limit for primary emission particles (> 23 nm) utilized by the EU in the EURO standards (6.0 · 10^{11} # km^{-1}) ... **It should be noted that the average fuel consumption on the street cannot be accurately determined, but the assumed 11.0 L per 100 km is a reasonable estimation based on the comparison of obtained EFs and the bottom-up emission inventory-based EFs (see 3.1.2)."***

In l.261 it is stated that the "obtained PM10 EF was 2.81 times higher than the PM2.5 EF, suggesting contribution by non-exhaust emissions". Is the reasoning for this statement that non-exhaust emissions contribute mainly to the coarse fraction > 2.5 µm? Please clarify, and also give a reason why the diurnal pattern of PN EF is different from PM2.5/PM10 EF (Figure S2).

- Yes, the difference between PM10 and PM2.5 EFs is very likely related to non-exhaust emissions because they mainly contribute to coarse fraction. Also, the exhaust emissions typically don't contribute to coarse PM. This reasoning is now mentioned in the revised text.
- The diurnal difference between PN and PM_{10} is an interesting point. Based on our data, we cannot unambiguously explain the main reason behind this result. We, however, think that it could be related to the (semi-)volatile emissions, influencing the PN emissions in the morning weather conditions. PM mass emissions are likely more related to primary and non-exhaust emissions, hence the increase in emissions cannot be observed with $PM_{2.5}$ or PM_{10}.
- Changes made:
    - *"**As non-exhaust emissions contribute mainly to coarse PM, the obtained PM_{10} EF, which was 2.81 times higher than the $PM_{2.5}$ EF, suggests** contribution by non-exhaust emissions even though winter conditions typically limit the effects of non-exhaust emissions compared e.g., to spring (Kupiainen et al., 2016; Stojiljkovic et al., 2019)."*
    - *"**Regarding the diurnal variation of EFs (Fig. S6), interestingly, the EF of PN considerably increased during the morning rush hours compared to other hours. This same phenomenon was not observed with $PM_{2.5}$ or PM_{10}. This result could potentially be related to (semi-)volatile emissions influenced by weather conditions, as in contrast, $PM_{2.5}$ and PM_{10} are more likely derived from primary and non-exhaust emissions."***

In l.277/278, it is stated that "$LDSA^{al}$ is especially dependent on the ultrafine particles, which deposit efficiently in the lung alveoli, as well as on BC". Why is $LDSA^{al}$ considered to be dependent on BC?

- BC particles are typically in a size range from about 50 nm to 200 nm. Compared to smallest ultrafine particles, BC particles have a higher surface area (also due to the fractal structure). They also deposit rather efficiently in the lung alveoli (even though not as efficiently as the smallest ultrafine particles). Hence, $LDSA^{al}$ is typically strongly related to BC concentration which can be also seen in the typical $LDSA^{al}$ size distributions measured in urban environments.
- Changes made (see the earlier comment): We added citations to Järvinen et al. (2019) and Lepistö et al. (2022).

In l.323-325, I recommend to change "Organics had also a strong R^2 of the averaged linear fit (0.977), whereas other compounds, i.e., sulfate, ammonium, nitrate and chloride, had larger confidence intervals" to "R^2 of the averaged linear fit for organics was 0.977, whereas other compounds, i.e., sulfate, ammonium, nitrate and chloride, had R^2 values < 0.8".

- We changed this sentence accordingly.

When discussing R^2 values presented in Table 3, I recommend to point out the lower R^2 values of 1,3-diethylbenzene and acenaphthylene in l.350, and add that "most studied alkane hydrocarbons except for heptane and octane" correlated well with CO_2 in l.351. For 1,3-diethylbenzene, Fig. S3 in the supplementary material shows R^2 = 0.961, not 0.761. Which one is the right R^2 value?

- Thank you for pointing this out. The R^2 of 1,3-diethylbenzene should be 0.961 in Table 3. Hence, all aromatic hydrocarbons had R^2 > 0.92. It is true that the link between acenaphthylene and CO_2 is rather weak (EF is close to zero and R^2 is lower compared to naphthalene).
- Changes made: We corrected the text and Table 3: "*All 18 studied aromatic hydrocarbon compounds **as well as naphthalene** (Table 3) had a strong correlation with CO_2 (R^2 of the averaged linear fit **> 0.92**).*"

In l.418/419 it is stated that "recombination could affect the inverse IFs of the small ions which could partially explain why the impact of CO_2 is more significant with the positive ions in Figure 4." Please explain your reasoning.

- The idea here was to discuss potential reasons behind the IF differences of positive and negative small ions. Earlier, the inverse IFs of small ions were discussed to be related to coagulation. However, the differences between the positive and negative ions suggest that some other processes happen as well. We decided to restructure and clarify this paragraph.
- Changes made: *"As shown in Section 3.1, increasing CO_2 concentration increased particle concentrations, which, in principle, should increase the rate at which ions collide with other particles. **The road traffic's inverse contribution to small ions can also be seen when examining the diurnal ion concentration variation (Fig. S13), where the concentration of small ions is reduced during rush hours compared to less trafficked hours, supporting the observation of negative IFs. The negative effect of traffic on small ion concentrations has been observed before, e.g., by Hirsikko et al. (2007). On the other hand, direct engine exhaust measurements have indicated that road traffic is a source of these smallest ions***

(Lähde et al., 2009). Thus, the results show a paradox: road traffic emits small sub-2 nm ions but, at the same time, increased road traffic decreases the concentration of these smallest ions in ambient air. Interestingly, the inverse effect of increased CO_2 on small ions is more significant with positive ions. Hence, in addition to coagulation, other processes such as recombination could potentially affect the inverse IF of small ions. For example, braking is a more significant source of negatively charged particles compared to positively charged (Thomas et al., 2024). Overall, the concentrations of positive ions were generally higher (Fig. S13), which could affect the result as well."

In l.424, regarding the emission of small ions derived from engine exhaust measurements, also automotive braking as a source of highly charged aerosol particles could be mentioned (e.g. Thomas et al., 2024, PNAS 121, e2313897121, https://doi.org/10.1073/pnas.2313897121).

- This is a very good point. We have now mentioned braking as a source of highly charged particles. See our response to the previous comment. Also, braking is now mentioned in the beginning of 3.2.2.
- Changes made: "Also, traffic is known to produce significant amounts of ions via vehicle exhaust (Yu et al., 2004; Jung and Kittelson, 2005; Maricq, 2006; Hirsikko et al., 2007; Ling et al., 2010) **and braking (Thomas et al., 2024)**."

Technical comments

l.50: Please change "indications that even majority of" to "indications that even the majority of".

l.101: Please change "The station located at" to "The station is located at".

l.109: Please change "Kuuluvainen et al., (2018) and Barreira et al., (2021)" to "Kuuluvainen et al. (2018) and Barreira et al. (2021)".

l.118: Please change "Teinilä et al., (2025)" to "Teinilä et al. (2025)"

- We made the above four corrections accordingly.

l.188: I recommend to simplify the statement "some of the studied pollutant metrics had inverse trends as a function of increasing CO_2 concentration", for example, to "some of the studied pollutant concentrations decreased with increasing CO_2 concentration".

- We corrected the statement accordingly.

l.209: I suggest to change "The concentrations of PN, BC, PM10, PM2.5 and $LDSA^{al}$ were strongly connected with the CO_2 concentration" to "Average concentrations of PN, BC, PM10, PM2.5 and $LDSA^{al}$ were statistically related to the average CO_2 concentration".

- We changed the statement to: *"According to the averaged linear fits, the average concentrations of PN, BC, PM_{10}, $PM_{2.5}$ and $LDSA^{al}$ were strongly connected with the average*

CO_2 concentration. The R^2 values of the averaged fits were above 0.93, indicating that the pollutants were mainly derived from road traffic at the studied site"

l.251: Please change "and median and" to "and median".

l.369: Please change "Naphtalene" and "Acenaphtylene" to "Naphthalene" and "Acenaphthylene" in Table 3, in Figure S5 (supplementary material) and in Table S5 (supplementary material).

- We made the above two corrections accordingly.

l.410: In Figure 4, is the mean diameter of the largest ion fraction shown in blue 38.5 nm as stated in the figure caption?

- We corrected the figure caption: *"Figure 4: Left: Concentrations of selected positive ions **in the size ranges of 0.8–1.5 nm, 1.5–5.0 nm and 5.0–38.5 nm** as a function of CO_2 concentration..."*

l.436: Instead of referring to Table S5, where also p-cymene shows poor correlation during pollution events, I recommend to refer to Table 4.

- We changed the reference to Table 4.

Reviewer 2:

The authors present a study using observations in a street canyon in Helsinki, Finland, to determine emissions factors (EFs) of the wintertime vehicle fleet, using CO2 as a tracer for fuel consumption. EFs are reported for a number of particle and trace gas observations, as well as inorganic and organic components of PM and VOCs, including aromatic hydrocarbons, alkanes, and PAHs. Their findings show that older fleet vehicles are contributing significantly to the emissions, and exceed recent (2014) emissions standards. Further, they investigate the emissions impact on O3 (inverse), and introduce impact factor (IF) to describe the direct and inverse impacts of on-road emissions.

I agree with Anonymous Referee #1 that the topic is important and that it is within the scope of ACP, but I also agree that there is problematic terminology and oversimplification in both the approach to separating episodic and non-episodic data, and in the definition of IF which does not take into account all the other factors that would be contributing to ozone impacts beyond simple CO2 comparisons. The authors should address these limitations on their methodology and provide bounds for the general usefulness of IF applicability to other study regions.

- Thank you for reviewing our manuscript. We have carefully considered your thoughts together with the comments made by the Reviewer #1. Our point-by-point responses to your comments are collected below:

Major comments:

There are several instances where the authors describe emission factors for ozone, which needs to be corrected as ozone is not emitted (e.g., lines 92, 378, Figure S6). By definition, an emission factor cannot be negative, so the terminology is problematic regarding negative EFs. Species may have an inverse relationship, but using EF is not correct. Even qualifying it as an inverse effect, i.e., negative EF (line 26), is not the right terminology.

- We agree that EF is not correct term in the context of ozone. To discuss the effects of road traffics on parameters that are not emitted by traffic, we introduce the term "impact factor" (IF) in the manuscript. Hence, the linkage between traffic and ozone should be discussed as an IF rather than EF. Thanks to this comment, we noticed that in some cases, we accidentally use wrong terminology in the manuscript. We have now carefully checked the text that EFs and IFs are used correctly (as pointed out by reviewer #1 as well).

The binary episodic/non-episodic seems problematic – assuming that there are other influences (e.g., long-range and regional biomass burning emissions, an inversion triggering a build-up of pollutants) that impact the canyon site significantly on occasion, one can only deduce that there is the potential for these impacts to contribute to the background levels during the non-episodic times. Thus, conclusions attributing findings during the non-episodic time periods to "on-road traffic" alone need to either (a) provide sufficient evidence that they are not influenced by other factors, or (b) quantify the possible extent of the other factors in the findings.

- This is an important comment. As mentioned in our responses to the comments by Reviewer #1, during the non-episodic periods, the pollutant concentrations were generally very low, indicating that the effect of other pollution sources on the results would be low. Furthermore, the average diurnal pattern of CO_2 (and the studied pollutants) followed strongly the diurnal pattern of road traffic rate. Hence, we are confident that the results during the non-episodic period indeed represent mainly the effects of road traffic. It needs to be noted that, in an urban area, we cannot completely rule out the effect other pollution sources, but the results strongly indicate that the road traffic was clearly the dominating factor during the non-episodic periods. Please, check our responses to comments by reviewer #1 for details (and made changes in the manuscript).

The resolution of several figures is too low, including Figures 3, S3, S4, S7. Some others are acceptable but not great, while others are very high resolution, i.e., S1 and S2. Recommend using whatever methodology was used for the latter for the remainder of the figures.

- All article figures in the manuscript have been made with same methodology. The figure files itself have high resolutions, but with some figures the resolution drops when the figure is attached in the .docx-file. We will upload high-resolution files for all figures separately, which should solve this problem. We also checked the resolution of the figures in the supplement.

Technical Comments

Line 101 – The station "is/was" located… (and consider using present tense when describing the measurement site that is likely still there with the same features – for this whole paragraph.)

Line 101 and throughout – per the style guide, dates should be in [dd month yyyy] notation.

Line 102 – "similarly, coordinates need a degree sign and a space when naming the direction (e.g. 30° N, 25° E)."

Line 109 (and throughout) – the commas after “et al.” are unneeded when preceding a parenthetical date.

Lines 119-120 – The times need a qualifying “local time, LT” or otherwise.

- We corrected these above-mentioned points in section 2.1 (and throughout the manuscript).

Line 158 (and throughout) – per the style guide, units should be written with negative exponents, not in the format J/(K mol), or km/h, etc.

- We corrected the units throughout the manuscript.

Lines 186-187, 189, 194 (and throughout) – “The abbreviation "Sect." should be used when it appears in running text and should be followed by a number unless it comes at the beginning of a sentence.” Similarly, Fig. should be used for figures not at the beginning of sentences.

- Checked and corrected throughout the manuscript.

Line 196 – remove the space before “-based EF calculation.”

Line 206 – “Figs. 6 and S9” and “Figs. S3-S5”

Lines 224, 225, 251, 297, etc. – the abbreviation for litres should be a capital L

Lines 231 and 360 – “in downtown Toronto”.

Line 273 – likely better to say “diesel city buses” (as in line 305)

Line 295 – perhaps “road traffic is no longer a significant…”

Line 319 – “so-called super emitters”

Line 338 – “to the authors’ knowledge”

Line 367 – consider uniformly capitalizing the VOCs in Table 3 (i.e., o-Xylene, 3-Ethyltoluene, 2-Ethyl-p-xylene, etc.)

Line 391 – the x in NOx should be subscripted

Line 392 – the 2 in NO2 should be subscripted

Line 404 – radon should not be capitalized

Line 423 – add commas before and after “e.g.”: “before, e.g., Hirskko et al…”

Line 425 – “paradox: road traffic” (road should not be capitalized).

Line 515 – “In the case of PN…”

- We checked corrected all the above-mentioned points accordingly

Tables S1, S2 and S4, S6, and also Tables 3 and 4 – I recommend not having the data in the table in italics for better readability.

- Italics is no longer used in tables.