Response to comment

We would like to thank Reviewer 1 for the comments and corrections, which have helped us improve the quality of the manuscript. Please find below the point-to-point response to the questions raised.

1. While the paper presents the counting efficiency results (in different ways), it falls short of discussing in enough depth the "effect" of soot aerosols on the counting efficiency, as the title suggests, and "why" those effects are observed.

Answer: The Reviewer is right that an in-depth discussion on why the effects are observed is missing. The reason is that most manufacturers have not disclosed publicly any information on the design of their instrument. It is therefore impossible for us to link the observed measurements (i.e. counting efficiency) with the exact operation principle of each DC-based sensor.

2. It is not clear why the studied particle properties (e.g., EC/TC ratio or primary particle size) would affect the instrument counting efficiency. I understand that diffusion charging depends on the size of the particle and its morphology (if it is not spherical), but particle effective density (at one size), primary particle diameter, and EC/TC ratio do not provide meaningful insight into particle morphology.

Answer: We would argue that the EC/TC ratio can have an effect on the morphology of the soot particles. Soot particles formed in premixed flames (i.e. high EC/TC) exhibit a loose agglomerate structure where the primary particles are clearly distinguishable from one another, while soot generated in fuel-rich flames (high OC/TC) has a more compact structure and the primary particles tend to merge with each other (see Fig. 3 in Ess at al. https://doi.org/10.1080/02786826.2021.1901847).

We have amended the text in Subsection 3.1 as follows: "Particle number concentration measured by diffusion chargers depends on the average number of charges carried by each particle (Fierz et al., 2011). Particle size and morphology have been shown to have an effect on the number of charges carried by the particles and, thus, on the counting efficiency of diffusion charger based PN-PTI instruments (see (Dhaniyala et al., 2011; Vasilatou et al., 2023) and references therein). Soot particles form complex structures described by a fractal-like scaling law (Mandelbrot, 1982), and their mobility is influenced by their morphology (described by the fractal dimension and fractal pre-factor) and the momentum-transfer regime (Filippov et al., 2000; Melas et al., 2014; Sorensen, 2011). To characterise the soot particles produced by the different aerosol generators, the following aerosol properties were determined: particle size distribution, EC/TC ratio, primary particle size and fractal dimension. EC/TC ratio can also have an effect on the morphology of the soot particles. Soot particles formed in premixed flames (i.e. high EC/TC) exhibit a loose agglomerate structure where the primary particles are clearly distinguishable from one another, while soot generated in fuel-rich flames (high OC/TC) has a more compact structure and the primary particles tend to merge with each other (see Fig. 3 in (Ess et al., 2021b))."

Specific comments

- Why did the authors choose TSI NPET 3795 as the reference particle counter? The authors state that NPET was calibrated according to ISO 27891, so why didn't they use the same reference instrument used to calibrate NPET as the reference particle counter? According to the specifications of NPET, it has a counting efficiency of < 50% at 23 nm and > 50% at 41 nm. The relatively low detection efficiency of NPET at 23 nm and even at 41 nm can potentially lead to unknown counting efficiency of PTI instruments, as some of the (smaller) soot aerosols may not be measured by NPET.

Since this study is done by METAS, I suggest a reference CPC (with a lower d50 size) and a diluter with a known dilution factor be used as the reference particle counter.

Answer: This is a good point. We chose to use the NPET as a reference particle counter for the field campaign because it is a robust, portable instrument which combines a stable, reproducible aerosol dilution system and a CPC in a single unit. The NPET was calibrated in the laboratory against a CPC (with a cut-off at around 6 nm) which was equipped with a custom-made dilution system. This system is not designed for field measurement as it is difficult to transport and would require recalibration every day. In our experience, there are no commercial dilution units with a known dilution factor, i.e. the dilution factor does not necessarily remain stable when the instrument is moved from the lab to the field.

We have amended Section 2 as follows: "The NPET was selected as reference instrument for two reasons; i) it could be used in field measurements as it included a dilution system, a volatile particle remover and a particle counter, ii) during type examination portable PN-PTI instruments are typically used as reference".

During data analysis, the particle number concentration reported by the NPET was corrected with respect to the size-dependent counting efficiency, therefore we are confident that the data reported in this study are reliable. We have amended the text as follows: " NPET had been calibrated in a traceable manner according to the ISO 27891 standard, and showed a CE of 0.58 ± 0.02, 0.77 ± 0.02, 0.77 ± 0.01, 0.80 ± 0.01 and 0.79 ± 0.02 at a GMD_{mob} of 23 nm, 50 nm, 70 nm, 80 nm and 100 nm, respectively, and this counting efficiency was taken into account during data analysis".

- The authors have shown TEM images of different soot aerosols, which provide qualitative insight about particle morphology. For example, it is clear from these images that soot from MISG has a very compact structure, while soot from other sources are fractal aggregates. However, it is important to quantify the morphology of particles too to allow studying its effect on counting efficiency. This is typically done by determining the fractal dimension (e.g., through image analysis) or mass-mobility exponent (e.g., measuring particle mass or effective density over a range of particle sizes). The use of effective density at one particle size (100 nm) or primary particle diameter cannot give meaningful information about particle morphology.

Answer: Thank you for this comment. We have now calculated the fractal dimension and amended Table 1 (see last column).

We have also amended the text as follows: "The fractal dimension D_f of soot particles with a nominal GMD_{mob} of 100 nm was derived via image analysis of high-quality TEM-images using the FracLac feature of ImageJ 1.53e (ImageJ, National institutes of Health, USA). In a first step, the greyscale TEM-images were converted into binary images utilizing the auto-convert function of FracLac. In a second step, the D_f values were determined via the so-called box counting, averaging 12 rotations of each image. The D_f values summarised in Table 1 represent the average values obtained from at least 10 particles for each type of soot. These values agree well with those reported in previous studies for bare (i.e. freshly emitted) soot particles (Pang et al., 2022; Wang et al., 2017)".

- The recommendations given in Section 4 do not seem to be directly drawn based on the results of this study. Rather, some of the recommendations are generic and seem to be based on the results of other or previous studies.

Answer: Recommendations based in previous publications (https://doi.org/10.1016/j.jaerosci.2023.106182) were amended to account for the new findings of this study. For instance, Recommendation 1) highlights that the **same type** of combustion generator

should be used for the determination of CE during type-examination and verification. Recommendation 2) is new and Recommendation 3) highlights the need to verify that type-approval of PN-PTI instruments is harmonised in Europe. Up to now, it was believed that DC-sensors would respond similarly to all types of soot irrespective of the combustion generator. We now show that this not the case, thus type-examination procedures in different European countries might not be equivalent since the approval authorities use different types of soot generators.

- Figures S6 – S9 are not referenced in the main text of the paper.

Answer: Thank you for bringing this to our attention. We have amended subsection 3.2 with the following sentence: "The counting efficiency of the different PN-PTI counters as a function of time is shown in Figs. S6-S9 for a measurement duration of 2 min".

- Table S1: It seems that EC/TC ratio is typed mistakenly as “EC/OC ratio”. In any case, EC/TC ratio is also given in Table 1, so I suggest providing this information in one place only (either in Table 1 or S1).

Answer: Thank you for spotting this typo. It now reads "EC/TC mass fraction (%)". Although the EC/TC mass fraction is provided in the main manuscript, we wanted to provide in SI a comprehensive summary of the setpoints we used during the study, so that the readers can find all relevant information in one Table.