Response to Report #2 of referee #3

We would like to thank Referee #3 for his continued support and comments.

We respond to the specific comments in the following:

1) Although the authors tried to response the comments, the manuscript were poorly revised. For example, it was suggested to summarize the problems of previous instruments and then state the advances of this study in the revised introduction and conclusion, rather than only response.

In view of this comment of referee #3, the authors further revised the manuscript (1) to summarize the problems of presently available impactors when using TXRF analysis of aerosol metal content and (2) to emphasize the advances of the newly developed cascade impactor in the correspondingly revised introduction;

see section 1, line 56 and 57 of the downloaded preprint manuscript:

> "…the classifying nozzles deposit the particles is usually significantly larger than the area analysed by TXRF. *As a result, only a fraction of the particles collected by the impactor are actually analysed, while a significant proportion of the impacted particles remain inaccessible for the TXRF analysis and consequently, the overall sensitivity is reduced. To take…*"

and section 1, line 65 of the downloaded preprint manuscript:

> *"…provide low blank values and minimum cross-contamination between subsequent sampling periods. The impactor nozzles of the new cascade impactor are arranged in such a way that all particles impacted on the respective sample carrier contribute to the TXRF analysis, thus increasing the overall sensitivity. This enables shorter sampling times, which in turn opens up new possibilities for identifying pollutant sources. The corresponding arrangement of the impactor nozzles could not be achieved simply by compressing a previous deposition pattern, but rather the number and lateral arrangement of the impactor nozzles have to be recalculated as well as the diameters of the impactor nozzles. As a result of these considerations, the new cascade impactor is designed for a reduced gas mass flow compared to commercially available impactors, which in turn enables the use of smaller pumps, and thus portable and mobile battery-powered operation of the impactor in the field. We will first present the…"*

In our opinion, the Conclusions section (section 5) already reflects all relevant aspects of the progress achieved by the new impactor presented in the manuscript.

2) The authors responded that "A quantitative comparison of chemical composition using TXRF would be difficult because the deposition patterns of these impactors have lateral dimensions in some stages that lie outside the excitation range and/or the detection range of the TXRF spectrometer used in the present study." How can the authors ensure the accuracy of this new instrument? If the quantitative comparison can not be conducted, how can the instrument be applied worldwide? I think parallel filters can be sampled and analyzed to conduct the quantitative comparison.

Quantification of impactor samples by TXRF is inherently difficult and still a topic of active research. For example, Seeger et al. (2021) show the potential and limitations of the quantification of element mass concentrations in ambient aerosol samples using a commercial cascade impactor and TXRF in a comprehensive study. Vigna et al. (2022) investigate the influence of impactor deposition patterns on TXRF analysis in a related study. Further examples of current studies on quantitative (T)XRF analysis are included in the manuscript, e.g. Hönicke et al. (2019).

Our manuscript focuses on the development of a new impactor design in combination with established TXRF analysis and first applications. We show that the quantitative results for lead and nickel in PM10 are consistent with annual mean values of PM10 concentrations of lead and nickel in the state monitoring network. We acknowledge that this is not a direct comparison of impactor and filter samples as suggested by the referee. A direct comparison of the new impactor design with commercial impactors would be difficult because of the influence of different impactor deposition patterns on the TXRF analysis. Filter samples, as suggestes, cannot be directly analyzed with the TXRF used in our study. A direct quantitative comparison of our impactor samples and filter samples would require extraction and subsequent analysis, for example by ICP-MS, which is beyond the scope of our study.

Hönicke, P., Andrle, A., Kayser, Y., Nikoaev, K.V., Probst, J., Scholze, F., Soltwisch, V., Weimann, T., & Beckhoff, B. (2020). Grazing incidence-x-ray fluorescence for a dimensional and compositional characterization of well-ordered 2D and 3D nanostructures. Nanotechnology, 31(50), 505709, https://doi.org/10.1088/1361-6528/abb557

Seeger, S., Osan, J., Czömpöly, O., Gross, A., Stosnach, H., Stabile, L., ... & Beckhoff, B. (2021). Quantification of element mass concentrations in ambient aerosols by combination of cascade impactor sampling and mobile total reflection X-ray fluorescence spectroscopy. *Atmosphere*, *12*(3), 309.

Vigna, L., Gottschalk, M., Cacocciola, N., Verna, A., Marasso, S.L., Seeger, S., Pirri, C.F., Cocuzza, M. (2022) Flexible and reusable parylene C mask technology for applications in cascade impactor air quality monitoring systems. Micro and Nano Engineering 14, 100108.