Response to Dr. Zsófia Kertész's opinion on the PhD dissertation entitled "Ecotoxicological profiling of PM generated by urban heating and transport"

First, I am grateful to Dr Zsófia Kertész for giving her valuable comments, recommendations and questions related to my dissertation.

Reply to the general comments

- **Although the language of the thesis improved by a lot compared to the preliminary defense, there are still plenty of grammatical and linguistic errors, which sometimes makes it difficult to understand.**

I am really sorry for grammatical and typing errors.

- **There are plenty of errors with the units (e.g. 5.3 Limit of detection: 0.001 µ/L, and the same in Tables 3 and 4., p.66: 290 mg/mg and 8.87 mg/m3, units in section 6.5.1: µg m_3, etc)**

I have made a unit mistake as follows: (e.g. 5.3 Limit of detection: 0.001 µg/L, and the same in Tables 3 and 4. p.66: 290 mg/kg and 8.87 mg/m^3, units in section 6.5.1: µg m^{-3}, etc)

- **The results and discussion chapter is relatively short (38 pages) compared to the lengths of the literature review and the description of the methods. In addition, it goes into great detail on comparisons with the international literature data, while the presentation of own results is extremely concise. During the discussion in some cases, it was not even clear which research was referred to (own data or literature data). In my opinion, it would have been better if you had explained your own results in more detail.**

Thank you for your suggestion and recommendation. Discussing own results I followed the structure of published papers and included their content as much as possible.

Reply to the specific comments and questions

- **p.11 (wildfire): "Emissions are predominated by fine PM, which represent app. 92 % of the PM (Vicente et al., 2017). In regions of Europe, the Fire Inventory from NCAR version 1.0 12 (FINNv1) estimated that coarse and fine PM annual average emission from open combustion were 0.39 Tg yr-1 and 0.22 Tg yr-1 (Wiedinmyer et al., 2011)." These two statements contradict top each other.**

I should have put more emphasis on the differences between the Portuguese situation reported by Vicente et al. (2017) and European data reported by Wiedinmyer et al. (2011).

- **P.15: p.15: 15 "In addition, black carbon, mineral dust, and sea salt can be found in SIA." In the previous chapter (3.2) these were all described as primary aerosols.**

I apologize for that. This statement contains dubious information.

- **p.17: p. 17: "PAHs released from open-air combustion amounts to 61% of the total man-made air pollution".**

I am really sorry for the mistake, this sentence was meant to express the idea as follows - The amount of PAHs released from open-burning globally contributes to 61% of the total man-made PAH emission.

- **What does biologically available and unavailable means in the following statement (p. 35): "First, such organic solvents are used in many cases which extract the whole toxic fraction of the sample, both biologically available and biologically unavailable."?**

Bioavailable toxicants can be readily assimilated by biota while the biologically non-available fraction of chemicals are bound to particles in a way which does not favour their assimilation (e.g. Pereira et al. 2023).

Pereira, J. G., Raikar, S. S., Bhatti, A. G., Fatarpekar, P. G., & Nasnodkar, M. R. (2023). Metal bioavailability, bioaccumulation, and toxicity assessment through sediment and edible biota from intertidal regions of the Aghanashini Estuary, India. Marine Environmental Research, 191, 106172.

- **You say, that "Second, these solvents are not part of a natural exposure pathway" What solvents are? Which solvent represent best the natural systems? Which solvent do you think is the most suitable and why?**

In case inhalation exposure pathway is to be investigated, the best solvent could be the lymph produced in the upper and lower airways which is very difficult to reproduce and cannot be covered by our laboratory. On the other hand, a large portion of the biota (espacially plants) can be affected by wet deposition meaning that water extract can represent this exposure pathway.

- **How do you explain the big difference between the toxicity of the water extract and the direct contact test? (ch. 6.1)**

Characteristic contaminants in atmospheric PM are polycyclic aromatic hydrocarbons. Acute toxicity as well as carcinogenicity of PAHs are expected to increase in parallel with the increase of the molecular weight (Zhang et al. 2020), however, these PAHs are not water soluble.

Zhang, L., Yang, L., Zhou, Q., Zhang, X., Xing, W., Wei, Y., Hu, M., Zhao, L., Toriba, A., Hayakawa, K., Tang, N., 2020. Size distribution of particulate polycyclic aromatic hydrocarbons in fresh combustion smoke and ambient air: a review. J. Environ. Sci. 88, 370–384.

- **5.1.3 "The blank samples was also sampled for each type of waste combustion" Please explain, what is called blank sample in this case, and how was it sampled?**

Blank samples were collected for seven waste types. In this case, the blanks represent background measurements during which only charcoal was burned. The quartz filters were conditioned at a temperature of 20 ± 1 °C and relative humidity (RH) of 45 %–50 % for 3 d. Filters were weighed in an isolated weighing room before and after the aerosol samplings according to the European standard (MSZ EN 12341:2014). The parameters (RH, temperature) were measured. The weighted filters were stored in glass Petri dishes (preheated at 450 °C) before sampling, whereas the exposed filters were stored in the freezer in glass Petri dishes wrapped in aluminum foil until conditioning and measurements (Hoffer et al., 2020).

In the each blank samples, direct contact test preparation was used to prepare an extract, after that all suspension were tested using kinetic Vibrio bioassay. Toxic unit (TU) indicated ranged from 0.12 to 0.61, which means that all blank samples were below 1.0 and it indicated that all blank samples were non-toxic. Kováts *et al.*(2012) who mentioned that the blank quartz fibre filter demonstrated completely inert and showed absolutely no toxicity.

Hoffer, A., Jancsek-Turóczi, B., Tóth, Á., Kiss, G., Naghiu, A., Andrea Levei, E. (2020) 'Emission factors for PM10 and polycyclic aromatic hydrocarbons (PAHs) from illegal burning of different types of municipal waste in households', Atmospheric Chemistry and Physics, 20(24), pp. 16135 16144. doi:10.5194/acp-20-16135-2020.

Kováts, N., Ács, A., Kovács, A., Ferincz, Á., Turóczi, B and Gelencsér, A. (2012) 'Direct contact test for estimating the ecotoxicity of aerosol samples', Environmental Toxicology and Pharmacology. Elsevier B.V., 33(2), pp. 284– 287. doi: 10.1016/j.etap.2011.12.021.

- **Is there an estimate of the possible contamination or loss of sample that could result from sample preparation for the direct contact test (e.g. grinding and transferring the sample)?**

In order to avoid contamination, agate mortar and other equipment are always cleaned using ethanol before using them for a new sample. In a Flash test normally we do not use the whole quantity of the prepared suspension, therefore any influence of potential sample loss is negligible.

- **Figures 19 and 21: it is difficult to read the figures, color figures would have been better. The captions of Figures 21 and 22 are exchanged.**

I apologize for figures. Fig 19 and 21 were published in *Kovats, N., Hubai, K., Sainnokhoi, TA., Eck-Varanka, B., Hoffer, A., Toth, A., and Teke, G. (2023): Ecotoxicity of PM10 emissions generated by open burning of waste PET. Environmental Toxicology and Pharmacology* and *Sainnokhoi et al. 2022. Characteristics of particle-bound polycyclic aromatic hydrocarbons*

(PAHs) in indoor PM2. 5 of households in the Southwest part of Ulaanbaatar capital, Mongolia. Environ. Monit. Assess. 194 (9), 1–21. https://doi.org/10.1007/s10661-022-10297- 0). These two published format were used in my dissertation. In the fig 21 and 22, I am really sorry for that.

- **p.66 6.4.1 is the 8.87 mg/m3 concentration is correct? "Based on evidence reports on human health, antimony occurring in higher concentration than 8.87 mg/m3 seem to cause respiratory problems"**

This sentence was trying to write the occurrence of high amounts of antimony poisoning among occupational workers in heavy industry and its correct form is: Chronic exposure to antimony trioxide dust from smelting plant (8.87 mg antimony/m3 or greater) was seen to cause pneumoconiosis, which is lung disease (black or brown lung). Therefore it is known as an occupational disease. However, it is important to note that the generally accepted threshold limit value is 0.5 mg/m^3 an 8-hour time-weighted average (https://www.epa.gov/sites/default/files/2016-09/documents/antimony-compounds.pdf). It is set by Occupational safety and Health Administration.

- **Same question: p. 66. 6.4.2 "Total PAHs content of both PET waste samples varied in the samples and ranged from 290 mg/mg to 425 mg/kg."**

I have made a unit mistake, the sentence was meant to be written as follows: "Total PAHs content of both PET waste samples varied in the samples and ranged from 290 mg/kg to 425 mg/kg"

- **What do you think what caused the large differences in $PM_{2.5}$ concentrations between households?**

Many factors are influenced to indoor air quality such as household age, construction quality and size, etc. (So et al., 2019). PM concentrations were significantly higher in HH-2 (324 μg/m3), followed by HH-8 (246 μg/m3), and HH-7 (214 μg/m3). Whereas, lowest PM concentration was found in HH-9 (63.2 μg/m3), followed by HH-5 (3.6 μg/m3), and HH-1 (107.2 μg/m3). We also compared the construction age, room size of household, people activities, and heater types. Difference was found between in HH-2, HH-7, and HH-8 to other households. Room size of these three households ranged from 18 to 27 m^2. The ages of construction were varied and all three were above 10 years as well as heater type is stove. In the family members, it ranged from 6-7 people. In the contrary, the age of HH-5 was 4.5 year, household area in HH-1, HH-9, and HH-5 ranged from 32 to 104 m2. Of the family members, all were above 6 people. In HH-5, heater type is stove and electric usage. Others had only stove. In another study, during heating season in urban area, northwest China, highest PM concentration (119± 64 μg/m3) found in small room (bed room) (Li *et al.*, 2016). Lim et al. (2018) determined the indoor PM2.5 concentration in ger with coal stoves during winter period around the non-connected heating system area, Ulaanbaatar capital. The

result showed that the average 24-h PM2.5 concentration was high, and also mentioned the combustion method of the stoves.

Tianxin, L.,Suzhen,C.,Delong, D., Zhang, Y., Wang, B., Zhao, X., Leaderer, P. B., Shen, G., Zhang, Y., and Duan, X. (2016) 'Household concentrations and personal exposure of PM2.5 among urban residents using different cooking fuels', Science of the Total Environment. Elsevier B.V., 548–549, pp. 6–12. doi: 10.1016/j.scitotenv.2016.01.038.

Lim, M., Myagmarchuluun, S., & Ban, H. (2018). Characteristics of indoor PM2.5 concentration in gers using coal stoves in ulaanbaatar, Mongolia. International Journal of Environment Research Public Health, 15(11). https://doi. org/10.3390/ijerph15112524

So, Y., Lee, N., & Kim, S. (2019). Characteristics of lifestyle and living environment of Ger District Residents in Ulaanbaatar, Mongolia. Korean Journal Public Health, 55, 12–21. https://doi.org/10.17262/kjph.2018.12.55.2.12

- **p. 75: what is the two numbers in the brackets in this sentence: "4-ring PAHs in indoor PM2.5 in each sample were abundant as well, in decreasing order: HH-5 (28%; 17.58%), HH-7 (21.71%; 6.42%), HH-8 (20.87%; 18.33%), HH-2 (18.75%; 8.57%)"?**

I apologize for the typing mistake. The sentence was meant to read as follows: 4-ring and 3-ring PAHs in indoor PM2.5 in each sample were abundant as well, in decreasing order: HH-5 (28%; 17.58%), HH-7 (21.71%; 6.42%), HH-8 (20.87%; 18.33%), HH-2 (18.75%; 8.57%).

- **6.5.5. I do not see the TU value of HH5 to be 5.5 in Figure 25. It is more near to 4 ("In HH-5, ratio of HMW PAHs amounted to 42.2%, TU was 5.5 respectively.") In figure 8 of the publica-tion (Sainnokhoi, TA., Kováts, N., Gelencsér, A. *et al.* Characteristics of particle-bound polycy-clic aromatic hydrocarbons (PAHs) in indoor PM2.5 of households in the Southwest part of Ulaanbaatar capital, Mongolia. *Environ Monit Assess* 194, 665 (2022). https://doi.org/10.1007/s10661-022-10297-0) and on Fig. 25 the TU values corresponding to the households HH1-HH10 differs significantly. What is the explanation to this? They should be the same.**

Thank you for the thorough comparison. Unfortunately, the above mentioned most possibly typing error cannot be corrected. However, values are correct in the final version of the dissertation and are compatible with the figure.

- **6.5.5. The cluster analysis of only 10 samples is rather forced (normally much more samples are needed for such an anylsis), and thus it yielded the result which was already known from the data. Therefore, I do not see the need for it.**

The cluster analysis was meant to visualise differences and similarities in the selected households.

- 6.5.5: Last sentence: **"In our study, the kinetic diagram of indoor PM2.5 extracted samples of households were higher than typical curves of each control samples,**

however, these diagram of samples are regarded as far less toxic." Please explain this statement. (e.g. less toxic than what?)

I am sorry for the dubious sentence. It was meant: In our study, the kinetic diagram of indoor PM2.5 extracted samples of households showed higher inhibition in the highest concentrations than curves of each control samples, however, these diagrams can be regarded as moderately toxic.

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