Response to Interactive comment of Anonymous Referee #1

Comments and questions of the reviewers are in italics

Authors' answers are in regular typeface

Parts of the answers highlighted in yellow are inserted into the revised manuscript.

General Comments.

1. *Abstract: About 75% of the abstract is background (lines 15-23); the presentation of this work would be more impactful if more of the results were included in the abstract. I recommend more inclusion of the PAH and PM10 emission factors.*

We agree with the reviewer that emission factors should be included in the abstract; upon revision, we have added the following paragraph to the abstract:

It was found that the PM10 emission factors from the combustion of wood-based waste samples were about twice that of firewood, whereas EFs in the range of 11–82 mg g^{-1} were obtained for different types of plastic waste. The latter were also found to emit exceptionally high amounts of PAHs, by a factor of 50–750 more than upon the combustion of dry firewood under the same conditions. Since the more toxic 4–6 ring PAHs were predominant in the particulate emission from plastic waste burning, BaP equivalent toxicity was up to 4100 times higher than that from wood combustion.

2. *57 and results: At first this statement seemed overly broad to me, but upon my own search of the literature I was unable to find many articles looking at emission factors for specific plastic wastes. I still believe the manuscript would be improved if the authors provided an additional discussion of the emissions of PM and PAH from waste mixtures. Or even emissions of other hazardous components from these wastes. These might provide additional perspective into the relative importance of the emissions of the tested materials. In the results section, discussion of the EFs you observe relative to other wastes would also be interesting. Specifically, for the PAH results. Are there any EFs which exist for PAHs for municipal/household waste mixtures? Could we say anything about which materials might be the most important contributors to total PAH or toxicity?*

Besides PAHs various gaseous and particle-phase compounds are emitted into the atmosphere from the combustion of municipal waste. Even if their determination is outside the scope of the manuscript, these species have been mentioned in the revised manuscript with references.

(Lemieux et al., 2004; Estrellan and Lino 2010; Gullett et al., 2010; Wiedinmyer et al., 2014).

The observed EFs relative to other wastes as well as the relative toxicity of the PAHs have also been discussed in the results section.

3. *How the blanks/standards are treated is very important for accessing the accuracy of results in filter-based studies. I think this manuscript would benefit from additional detail regarding the methods. I've listed a few specific comments around this issue below. 137: What type of blanks are these? Are they just conditioned and unused filters? Or, are they a type of background measurement during times when the coal was being burned?*

We agree with the referee, the following part has been added to the manuscript:

The blanks represent background measurements during which only charcoal was burned. The sampling times of the blanks were comparable with those of the samples. For each waste type one blank sample was collected with combined air supply settings.

> *152: Is the PAH analytical method based on one found in literature or one that the authors developed? If it is from the literature, please include a citation. If it was developed by the authors, please include additional method validation data in the SI. 164: What about recovery from the filter? There is a relatively high potential for matrix effects in complex samples like those from combustion. The best way to account for these and get at the "true" recovery would be to spike a subset of filters (with particulate samples on them) with known concentrations of the target PAHs and examine the recovery including both filter extraction and any matrix effects. As performed, the QA/QC approach cannot account for these. If you are unable to perform these tests now, please provide a discussion of the actual recoveries that other studies have observed for similar filter extraction techniques and how these recoveries may impact your conclusions.*

The PAH analytical method is the adaptation of the standard method EN ISO 17993:2003. Concerning recovery the sample preparation can be divided into 2 parts:

1. Solvent extraction of PAHs from the aerosol particles collected on the filter. This step can be characterized by the extraction efficiency.

2. Once PAHs are dissolved in the extract purification and concentration of the extract will further influence the recovery of the target compounds.

The recovery of the second part can be determined by dissolving standard solution of PAHs in the extraction solvent (n-hexane in this case) and performing the extract purification and concentration steps. This has been performed and the recovery of the individual PAHs from the extract was determined accordingly.

The recovery of the first part i.e. extraction efficiency can be determined by two approaches:

a.) First the amount of PAHs is determined in a subset of a sample then another subset of the same sample is spiked with known amount of PAHs and then analysed again. From the difference of the two results the extraction efficiency can be determined. However, this approach involves the assumption that PAHs originally present in the sample and those added to the sample can be extracted with the same extraction efficiency which cannot be guaranteed.

b.) The extraction efficiency can be determined without need for the above assumption by performing consecutive extraction and quantifying PAHs in the extracts as long as they can be detected. By increasing the solvent volume the number of consecutive extraction steps necessary to achieve nearly quantitative extraction can be decreased. We applied this approach and analysed PAHs in two consecutive extracts. In the second extract the amount of PAHs was only 1–3% of that present in the first extract. These results evidenced that under the experimental conditions applied (solvent to filter ratio, time of extraction) 97–99% of PAHs were extracted in the first step. Therefore, the extraction was not continued and the sum of the amount of PAHs in the two extract was considered as 100%. Because of the high efficiency of the first extraction step the samples were extracted only once and the results were corrected with the extraction efficiency (97%–99%).

The manuscript has been complemented with a more detailed description of the sample preparation as follows:

The extraction efficiency was tested by sequential extraction of the same filter spots in two consecutive steps. In the second extract the amount of PAHs was found to be only 1–3% of that present in the first extract. These results evidenced that under the experimental conditions applied (solvent to filter ratio, time of extraction) 97–99% of PAHs were extracted in the first step. Therefore, the extraction was not continued and the sum of the amount of PAHs in the two extract was considered as 100%. Because of the high efficiency of the first extraction step the samples were extracted only once and the results were corrected with the extraction efficiency (97%–99%). Recovery of PAHs from the extract was also studied by performing the clean-up procedure on diluted PAH standard solutions. Average recovery of 98 % (SD=8.9 %) was obtained for the 12 PAHs investigated in this study. The analytical results were corrected for both extraction efficiency and recovery of the sample preparation.

4. *The EF data that the authors use is drawn from a diluted gas stream. This methodology makes sense given the high particle concentrations associated with combustion. That said, the authors should add additional discussion of how such a sampling method may affect their results. How do the authors think the cooling associated with dilution may affect the particulate mass and composition of the emitted particles? Is it reasonable to expect that this rate and magnitude of rapid cooling would also happen in the atmosphere? This process will affect gas to particle partitioning; do the authors believe this is important for the levels of PAHs or PM they observed?*

We agree with the referee, the following text has been added to the manuscript:

The physical processes took place during the applied sampling procedure were very similar to those occurring after the emission of smoke from the chimney into open air as in both cases the smoke cools and dilutes rapidly.

…and to sub-chapter 2.3:

The ambient temperature affects the gas-to-particle partitioning of the compounds especially those of the semi-volatile ones. During the sampling the ambient temperature varied between 16.1 °C and 26.0 °C, on average it was 20.3 °C, which is higher than typical ambient temperatures during the heating season. It implies that some semi-volatile species might be lost to the gas phase upon sampling, thus measured EFs for PM10 and total PAHs are likely lowerbound estimates for real-life conditions. It should be added that the standard procedure of the gravimetric PM_{10} measurements include the conditioning of the exposed filters for 48 hours at 20°C (and RH=50%) which might also result in some loss of semi-volatile compounds. Concerning PAHs, at the sampling temperatures some of the more volatile 2- and 3-ring compounds might have been lost to the gas phase as compared to the case at ambient temperatures in winter, and were likely to some extent underrepresented in the filter samples. These potential losses, however, do not influence the conclusions regarding PAH relative toxicity due to the very low toxicity of the 2-3-ring compounds.

5. *The Authors introduce abbreviations in section 2.1, but do not consistently use them throughout the text. For clarity, I recommend using them any time material is referenced after the abbreviation is introduced.*

We agree with the referee, the abbreviations have been updated in revised manuscript.

6. *The discussion of PAH's in the manuscript is limited to bulk EFs and estimated toxicity. One interesting finding that could be extracted from these data is the EFs of individual PAHs for specific substances. Although a complete discussion of these data may be beyond the author's scope, at least a cursory discussion of any patterns observed in PAH concentrations would be very interesting. For example, are there any specific PAHs that could be used as a tentative tracer of the burning of a specific waste? I also recommend adding a table of the emission factors of each PAH for each material, either in the supplement or main text. Such a table would add value to this text for the readers. This would also increase the usability of the data to those who are interested in applying it in future emissions studies.*

A table (S2) listing the EFs for each PAH and waste type has been added to revised the manuscript with a discussion of the results as follows:

The EFs of the individual PAHs (Table 2 in the SI) are different for wood and waste burning. While the combustion of WOOD resulted in the emission of primarily phenanthrene and 4-ring PAHs (fluoranthene, pyrene, benzo(a)anthracene and chrysene) and the contribution of 5- and 6-ring compounds was only a few per cents, the latter compounds accounted for 14–45% of the total PAH emission in the different waste types. This shift in the chemical profile has a considerable effect on the toxicity of the particles emitted as discussed later. The availability of PAH emission data from the burning of any given type of waste is rather limited in the literature. Tomsej et al. (2018) studied the co-combustion of wood and polyethylene plastics (PE and PET). They burned 93 w/w% beech log and 7 w/w% PE in a 20 kW boiler and obtained total PAH EFs of 11 mg kg^{-1} (SD 1.2 mg kg^{-1}) and 16 mg

kg^{-1} (SD 1.6 mg kg^{-1}), in the particulate phase for the 12 compounds investigated in our study under nominal and reduced output conditions, respectively. The corresponding values for the co-combustion of 93 w/w% beech log and 7 w/w% PET were 8.5 mg kg^{-1} (SD 1.0 mg kg^{-1}) and 19 mg kg^{-1} (SD 3.6 mg kg^{-1}). These results indicate the influence of operational conditions on the emission factors of PAHs. Furthermore, the total PAH EF values obtained for the mixtures of beech/PE and beech/PET by Tomsej et al. (2018) were between the total PAH EFs obtained for WOOD (0.34 mg kg^{-1}, SD 0.19 mg kg^{-1}) and PE (31 mg kg^{-1}, SD 23 mg kg^{-1}) or WOOD and PET (32 mg kg^{-1}, SD 14 mg kg^{-1}) in our study, respectively. Maasikmets et al. (2016) studied the EFs of 4 PAHs emitted from burning municipal solid waste mixed with wood in domestic heaters. They reported emission factors of 0.41 mg kg^{-1}, 0.18 mg kg^{-1}, 0.12 mg kg^{-1}, and 0.10 mg kg^{-1} for benzo(a)pyrene, benzo(b)fluoranthene, benzo(k)fluoranthene and indeno(1,2,3-cd)pyrene, respectively. As a comparison, Lemieux (1997) found EFs of 1.4 mg kg^{-1}, 1.86 mg kg^{-1}, 0.67 mg kg^{-1} and 1.27 mg kg^{-1} for the same compounds from barrel burning of household waste. These emission factors are comparable to those found in our experiments and summarised in Table S2.

7. *Results Section: I recommend adding SD values to any reported EF throughout the text. The only reason not to add these would be if there are too few measurements or that conditions were so different between tests that the experiments are essentially measuring different endpoints. If the latter, then these differences should be discussed in detail. Additionally, there are many points in the results the authors state that their results are similar to those in literature when at first look the values are quite different (see specific comments below). This manuscript could be improved by further delving into the sources of these differences.*

We agree with the referee, the SD values have been calculated and are shown with the measured EF values. The large SD values indicate the effects of the different combustion parameters. For discussions of the differences between our and literature results, see our answer to another question below.

Specific Comments

8. *Introduction: As written the introduction is all one paragraph. I recommend separating this section into at least 3 paragraphs to help with readability.*

We agree with the referee, in the revised manuscript the introduction has been split into 3 paragraphs.

47: Be sure to spell out abbreviations the first time they are introduced (OSB here) throughout the text.

We agree with the referee, the abbreviations have been updated in the revised manuscript.

> *49-51: This line repeats much of what is said in line 47.*

Agree, the sentence has been deleted.

> *51-52: This statement should be supported by references since it is essential to the motivation for this work.*

We agree with the referee, four new references have been added to the revised manuscript:

Burning any type of waste poses excessive risks to the health of people living in those areas since a plethora of toxic, carcinogenic or mutagenic compounds are emitted in immense quantities compared to the burning of authorised solid fuels such as dry fuel wood or high-quality coal (Lemieux et al., 2004, Estrellan and Lino 2010, Gullett et al., 2010, Wiedinmyer et al., 2014).

Estrellan, C. R., and Iino, F.: Toxic emissions from open burning, Chemosphere, 80, 193–207, 10.1016/j.chemosphere.2010.03.057, 2010.

Gullett, B. K., Wyrzykowska, B., Grandesso, E., Touati, A., Tabor, D. G., and Ochoa, G. S.: PCDD/F, PBDD/F, and PBDE Emissions from Open Burning of a Residential Waste Dump, Environmental Science & Technology, 44, 394–399, 10.1021/es902676w, 2010.

Lemieux, P. M., Lutes, C. C., and Santoianni, D. A.: Emissions of organic air toxics from open burning: a comprehensive review, Progress in Energy and Combustion Science, 30, 1–32, 10.1016/j.pecs.2003.08.001, 2004.

Wiedinmyer, C., Yokelson, R. J., and Gullett, B. K.: Global Emissions of Trace Gases, Particulate Matter, and Hazardous Air Pollutants from Open Burning of Domestic Waste, Environmental Science & Technology, 48, 9523–9530, 10.1021/es502250z, 2014.

> *53-55: Please clarify what specified means in this context. Do you mean that these emissions are not included in emissions inventories? If so, please provide examples and/or references.*

Yes we do. The following part has been added to the text:

Since burning any kinds of municipal waste in households is strictly prohibited all over Europe, understandably such emissions are not included in many emission inventories (Wiedinmyer et al., 2014).

> *55: Do these reports from NGOs provide any estimations of magnitude? If so, it would be interesting to see them reported here.*

The reports from the NGOs focused on the frequency of the waste burning and/or the type of the burned waste, the magnitude of the waste burning is not known.

> *53-55: This line is slightly contradictory since it states that emissions are not regulated but are illegal. I would interpret "illegal" as a form of regulation. Do you mean there's no enforcement?*

The referee is right, the original sentence was misleading. In the revised manuscript it has been rephrased as follows:

Since burning any kinds of municipal waste in households is strictly prohibited all over Europe, understandably such emissions are not included in many emission inventories (Wiedinmyer et al., 2014).

Yes, in Hungary basically there is no enforcement against this illegal activity.

> *64,65: Add citation for PAH carcinogenic and mutagenic properties.*

In the revised manuscript the following citations have been added for PAH carcinogenic and mutagenic properties:

PAHs are among the most hazardous combustion products due to their carcinogenic and mutagenic nature (Bjorseth, 1983; Kim et al., 2013).

Bjorseth, A.: Handbook of Polycyclic Aromatic Hydrocarbons, Marcel Dekker Inc; 1st edition, ISBN-10:0824718453, 1983.

Kim, K. H., Jahan, S. A., Kabir, E., and Brown, R. J. C.: A review of airborne polycyclic aromatic hydrocarbons (PAHs) and their human health effects, Environment International, 60, 71–80, 10.1016/j.envint.2013.07.019, 2013.

> *68-69: Please clarify what you mean here by a single study. Do you mean a single study reporting EFs of PM2.5 and PAHs or a single study with plastic waste in an industrial boiler? What about emissions factors from bulk household waste combustion?*

We meant that there is only one study dealing with the emission factor of particulate mass and that of the PAHs obtained for the co-combustion of specific wastes (PE and PET) mixed with wood. In the revised manuscript the text has been modified as follows:

To the best of our knowledge there has been only one study reporting PM and PAHs emission factors for co-combustion of PE and PET waste with beech logs (Tomsej et al., 2018).

> *74-102: Some abbreviations are introduced twice here. I would remove the first instance (starting at line 74) or use the abbreviations after they are introduced.*

We agree with the referee, the abbreviations have been revised and updated.

> *109: As written this sentence is slightly confusing. I believe the authors are trying to communicate that burn conditions were variable, but later they report relatively consistent burning temperatures.*

We agree with the referee, the sentence has been modified as follows:

Each type of waste (except PAP) was burned at high, combined and low air supply resulting in different temperatures and conditions inside the oven, but it was largely independent from the type of waste combusted in the stove at any given air supply ratio.

> *140: Which parameters?*

In the revised manuscript the sentence has been modified to 'the RH and temperature data were recorded.'

> *186: For clarity, I recommend using the abbreviations you introduced in your methods throughout the results section. Also, include the EFs for RAG here since they have been included for the other materials.*

In the revised manuscript the EF of RAG has been added. The abbreviations have also been revised and updated.

The EF of burning RAG (8.7 mg g^{-1}, SD 0.22 mg g^{-1})…

> *186-187: If they exist, also report the standard deviation of all measured EF values.*

In the revised manuscript the SD values are reported.

> *190: Please explain how these values are similar. As written, it looks like the values reported here are ~2x the values found in the literature.*

The sentences have been modified as follows:

The only available EFs reported for PET co-combustion in a boiler are between 1.8 ± 0.3 and 5.8 ± 0.5 mg g^{-1} depending on the combustion conditions (Tomsej et al., 2018). The differences between our and the reported EF values can be explained by the vastly different burning conditions (20 and 5 kW nominal power output for the boiler and the stove, respectively, and no fuel wood was co-fired in our experiments as against in those reported by Tomsej et al.).

Here we note that if we estimate the EF of the PET from the data reported by Tomsej et al. (2018) by taking into account the mass ratio of the burned materials (wood to plastic ratio was 93:7) and assuming linear combination of the EFs (which might be loaded with large uncertainties), the EF derived for combustion of 'pure' PET turns out to be identical (11 mg g^{-1}) to that obtained in the present study.

208: Capitalize RAG

Agree, done.

212: Your values do not seem similar to the literature values as you stated. Please comment on the differences.

Since the EFs depend on several parameters and refer to the specific experimental conditions, we believe that even a factor of 2 difference can be interpreted as a relatively good agreement. The relatively high SD values obtained in our study for a given plastic type indicate the effects of some key combustion parameters on the EFs. Nevertheless, the term 'similar' has been removed from the text and the differences have been commented as follows:

The differences between our and the reported EF values can be explained by the vastly different burning conditions (20 and 5 kW nominal power output for the boiler and the stove, respectively, and no fuel wood was co-fired in our experiments as against in those reported by Tomsej et al.).

219-220: I think these are referring to the emissions relative to wood. Using the word "increased" here is confusing because it makes it sound like something is causing the change. I recommend modifying this sentence for clarity.

Thes sentence has been modified as suggested:

The relative PAH emissions were higher by more than a factor of 50 in case of burning RAG, PE, TR and PET, and were well over a factor of 100 for PVC, PU, PP, PS and ABS.

227-232: add SDs to the reported PAH emission factors.

As suggested, SDs have been added to PAH EFs.

246: This is a very interesting result, I recommend highlighting it in the abstract.

In the revised manuscript the relative toxicity of PAHs has been highlighted in the abstract.

Since the more toxic 4–6 ring PAHs were predominant in the particulate emission from plastic waste burning, BaP equivalent toxicity was up to 4100 times higher than that from wood combustion.

> *Conclusion: Again, add SDs to all reported EFs.*

As suggested, SDs have been added to PAH EFs.

> *Figure 2: Clarify which axis is for which CO2 measure. Although this can be deduced by the reader, it should be explicitly stated. Additionally, it would be interesting the highlight the filtering times on this figure.*

Figure 2 has been modified to clarify which axis is for which CO_2 measurement.

> *Tables 1-3: It would not be too difficult, and would increase the clarity of the presentation, to combine all three of these tables into one. This would give the reader a single point where they can access all the results of the study as opposed to the current presentation where they need to jump between 3 different tables.*

As suggested, in the revised manuscript Tables 1-3 has been merged into a single Table for better readability.

Response to Anonymous Referee #2

Major comments:

> *In general, I find that there is insufficient context to the discussion. The manuscript still leaves open the question of importance, which I am sure any reader would ask. The only quantitative evidence I can find in the manuscript is a survey of Hungary, which found that 2-10% of participants are engaged in some sort of household waste combustion activity. So how much is there? All the results here are reported in emission factor ("emission" per "mass burned"), but there are virtually no data on "mass burned". Even an order of magnitude or a relative estimate would be useful. For example, for an average household who burn household waste, what is the ratio of mass of wood burned to mass of plastic burned?*

The primary focus of the manuscript was to determine the emission factors of PM10 and PAHs from the burning of different household wastes in stoves. An assessment of the relative contribution of waste burning to air pollution in any region was outside the scope of the manuscript. The polls conducted in Hungary focused on the time frequency of waste burning as well as on the relative fraction of the population who admittedly burn wastes at their home, but there was not any single question about the amount of waste burned. Since waste burning is an illegal activity but law enforcement is very rare, there are no estimates for the potential amount of waste burned in households in any regions. Using a tracer methodology, we plan to address this issue in another paper in the future.

> *I am also curious why plastic materials yield much higher PM and PAHs than wood materials. Is the composition of the material itself, or the mechanism of combustion? Both are organic polymers, but the size of the macromolecules may be different. Wood is generally made up of carbon, hydrogen and oxygen, where in plastics and rubber, nitrogen and chlorine are also present. The PM emissions seem to be somewhat linearly additive, as evidenced by the emission factors of RAG (a mixture of PET and wood) to be in between PET and wood.*

We found higher PM_{10} and PAH emission factors for those plastic materials which contain styrene units in the polymer chain (PS and ABS, which are made up of carbon, hydrogen and oxygen). This result indicates that the chemical composition affect the magnitude of the PAH formation and the PM10 emission factors. Beside the chemical composition the burning parameters, i.e. the mechanism of combustion may also affect the emission factors and the formation rates of PAHs. Indeed the emission factor of the RAG is between those of its constituents, but this might not be universally valid.

> *This manuscript also makes no mention of all the other toxic compounds that are known to be emitted from plastic burning. Two of the most studied ones are dioxins and furans. There may also be a lot of metals involved, such as from tyre materials. That is an important body of knowledge that is highly relevant to this discussion.*

Besides PAHs various gaseous and particle-phase compounds are emitted into the atmosphere from the combustion of municipal waste. Even if their determination is outside the scope of the manuscript, their occurence has been mentioned in the revised manuscript with references. (See also the response to Reviewer#1)

> *I would also note that the introduction, results and discussions are focused on the “illegal” burning of waste, but I am not sure why this aspect is important. What is it about “illegal” burning that needs special treatment? (It is even in the title.) To me, this is just combustion of different materials, and some materials yield much more PAHs and PM than others. Are “illegal” combustion conditions any different? If so, it might be useful to highlight. This issue is not just one of semantics – it is important for understanding the context of these results too. These emission factors may also apply to “legal” waste combustion, such as incineration facilities. For example, the high emission factors from plastics may provide additional incentives to separate them from other waste prior to incineration. Another potential area of concern is the increasing prevalence of wildland-urban interface fires. More manmade materials and household items would be burned in these fires, and these results have potential implications on the PM emissions and toxicity. My opinion is that the manuscript in its current form does not draw any connections to many other relevant areas.*

In our opinion, illegal and legal burning of waste should be treated separately. The legal waste burning includes waste combustion in municipal solid waste incinerators and/or in power plants where the emission of the pollutants and the burning processes are strictly controlled and regulated. This means that the emission factors obtained in the present study cannot be applied for waste incineration plants, as for them much lower emission factors are expected due to the more efficient combustion conditions (much higher temperatures) and the use of high-efficiency scrubbers, electrostatic precipitators, and filters. In contrast, illegal waste burnings are performed household stoves (or in open air) without any emission control to reduce the emissions. In this study we determined the EFs from waste burning in a commercial household stove under different and potentially relevant burning conditions. Burning of municipal waste in residential stoves is strictly prohibited in most countries that is why the word “illegal” is in the title of the manuscript.

> *There are also some major questions about the methods:*
>
> *First is that based on the pictures in Figure 1, the materials appear to be prepared such that they are all roughly the same size. How does the material preparation affect the emissions? Would large pieces combust differently and emit different amounts of pollutants?*

The size of the specimens of any given waste type was determined in preliminary test burns with the objective that similar filter loads be obtained in each combustion test. We agree with the referee that large differences in the size of the specimens might affect the burning mechanism and thus the measured emission factors. In our case relatively good agreement was found with available EF data in the literature (in the case of wood and PET) implying that the method may be acceptable.

> *Second, how are the emissions normalized to calculate EFs? What is the mass for normalization? Is it the mass of materials put into the stove, or the mass loss from combustion?*

The EFs were calculated using the weight of the waste specimens put into the stove. Here we note that the ash content of the plastics reported by Zevenhoven et al. (1997) (LDPE, HDPE, PP, PVC, PS) is below 3%.

Zevenhoven, R., Karlsson, M., Hupa, M., and Frankenhaeuser, M.: Combustion and gasification properties of plastics particles, Journal of the Air & Waste Management Association, 47, 861-870, 10.1080/10473289.1997.10464461, 1997.

> *Third, what is done about positive and negative artifacts for sampling semivolailte organic compounds and PAHs on quartz filters?*

The temperature of sampling does influence the partitioning of PAHs between gas and particulate phase as a consequence of the temperature dependency of the vapour pressure of these compounds. See also our answer to reviewer #1. We have not applied any method (e.g. dual filter trains, denuders etc.) to correct for potential sampling artifacts. Although this may add to some uncertainties to our results, especially with regard to the most volatile 2- and 3-ring PAHs, the main conclusions of our manuscript remain valid and robust.

> *How are the experiments repeated? I see there are uncertainty bars (and S.D.) reported in the figures. Are they replicate samples, or samples from different burns?*

Each type of waste was burned in separate test burns under different combustion conditions (see Figure S1 in the Supplement). SD values were calculated from the different test burns for each waste type.

Other comments

> *Title: the choice of the word "illegal" is questionable here. At first I thought this was a field study, sampling actual illegal combustion in households. Rather this work is done in a highly controlled environment. Therefore I find the title misleading and inaccurate.*

See also our corresponding answer above. The reviewer is partly right, we are not after illegal waste combustion in settlements. However, we did mimic illegal solid waste combustion in households in controlled laboratory experiments, therefore we believe that the use of the word "illegal" in the title can be justified. An additional argument is that by using our results the magnitude of the problem of illegal waste burning in settlements can be assessed knowing the amount of waste burned under such conditions.

> *Abstract: The abstract should mention more of the results and significance of this work. In this work, the major finding is that EF of PM10 and PAHs for plastics are much higher than expected, and toxicity equivalent factor is much higher. There are also some unsubstantiated claims about waste burning ("unprecedented health hazards") that should be moderated.*

In the revised manuscript more results has been added to the abstract (see the answer to reviewer #1). We agree with the reviewer that the word "unprecedented" is unsubstantiated. It has been replaced with "significant".

with the exception of a few media reports or court cases. Even though particulate emissions from illegal waste burning pose significant hazard to human health due to the combination of excessive emission factors (EFs) and uncontrolled chemical composition, there is scarce information on the specific EFs for PM_{10} and polycyclic aromatic hydrocarbons (PAHs) in the scientific literature. In this work, controlled combustion tests were performed with 12 different types of municipal solid waste and particulate emissions were measured and collected for chemical analysis. Absolute EFs for PM_{10} and PAHs as well as the benzo(a)pyrene toxicity equivalent of the latter are reported for the first time for the indoor combustion of 12 common types of municipal solid waste that are frequently burned in households worldwide. It was found that the PM_{10} emission factors from the combustion of wood-based waste samples were about twice that of firewood, whereas EFs in the range of 11–82 mg g^{-1} were obtained for different types of plastic waste. The latter were also found to emit exceptionally high amounts of PAHs, by a factor of 50–750 more than upon the combustion of dry firewood under the same conditions. Since the more toxic 4–6 ring PAHs were predominant in the particulate emission from plastic waste burning, BaP equivalent toxicity was up to 4100 times higher than that from wood combustion.

> *Abstract: the synonym EF is defined here but not used again in the abstract.*

As suggested, in the revised manuscript it has been corrected.

> *Introduction: There are instances where the background discussions reflect a Eurocentric bias and invoke negative stereotypes. For example, the first sentence mentions "In developing countries, more than three billion people use solid fuels. . .". The fact that developing countries are burning solid fuels is irrelevant to this work and mentioning "developing countries" is unnecessary, since this work seems to focus on illegal waste burning in European countries. Futhermore in a later paragraph, ". . .the illegal burning of municipal solid waste in households in several countries of the world, even in Europe". The way this sentence is constructed reflects an assumption that illegal activities are not expected in Europe and perpetuates a stereotype. I urge the authors to use more inclusive language and consider more diverse perspectives in their writing.*

We agree with the referee and have rephrased the objected sections as follows:

Billions of people use solid fuels (wood, coal or agricultural waste) as the main source of household energy worldwide (Anenberg et al., 2013).

On top of the significant emissions from the burning of solid fuels in households there is another existing yet largely unknown source of atmospheric pollution, the illegal burning of municipal solid waste in households in several countries of the world.

> *Page 2, lines 58-59: these papers should be cited, since they represent the state of knowledge on this topic.*

As suggested, the following references have been added to the revised manuscript.

Gu, Z. P., Feng, J. L., Han, W. L., Wu, M. H., Fu, J. M., and Sheng, G. Y.: Characteristics of organic matter in PM2.5 from an e-waste dismantling area in Taizhou, China, Chemosphere, 80, 800–806, 10.1016/j.chemosphere.2010.04.078, 2010.

Kumar, S., Aggarwal, S. G., Gupta, P. K., and Kawamura, K.: Investigation of the tracers for plastic-enriched waste burning aerosols, Atmospheric Environment, 108, 49–58, 10.1016/j.atmosenv.2015.02.066, 2015.

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Page 2, lines 65: again, references for the toxicity of these compounds should be cited.

As suggested, references have been added as follows:

PAHs are among the most hazardous combustion products due to their carcinogenic and mutagenic nature (Bjorseth, 1983; Kim et al., 2013).

Page 3, line 109: what does high/low air supply ratio mean? This is qualitative and not really explained.

As suggested, the following explanation has been added to the text:

The stove used was a commercially available model which allowed setting the air supply entering the combustion chamber of the stove through an adjustable slit. There were two endpoints of this slit, thus either larger (high air supply ratio) or smaller volume of fresh air (low air supply ratio) was allowed into the combustion zone.

Page 3, line 119: how much of the CO2 comes from the background smouldering of coal?

As Figure 2 illustrates the CO_2 concentration of the background case (between peaks when no waste specimen is being burned) is a few per cent (between about 4% and 5%).

Page 5, line 190: what does significantly higher nominal power mean? Why is that relevant to emission factor? It might be more direct to say how the combustion conditions differ. Also, I believe "though" should be "through", but I am not sure.

Indeed, it is difference between the combustion conditions that matters. The objected part has been modified as follows:

The only available EFs reported for PET co-combustion in a boiler are between 1.8 ± 0.3 and 5.8 ± 0.5 mg g^{-1} depending on the combustion conditions (Tomsej et al., 2018). The differences between our and the reported EF values can be explained by the vastly different burning conditions (20 and 5 kW nominal power output for the boiler and the stove, respectively, and no fuel wood was co-fired in our experiments as against in those reported by Tomsej et al.).

Page 6, line 196: tyres contain a lot of metals and their emissions are potentially toxic too. This paper examines PM and PAHs, but metals and organic compounds could be redox active and cause oxidative stress. This may be a good topic for discussion in the conclusions section.

We agree with the referee, but given that the topic was outside the scope of our study and no relevant measurements were carried out, we did not discuss it in the conclusion section.

> *Page 6, Line 200: ABS is very interesting. Since acetonitrile is involved, there may potentially be nitrogen-containing compounds in the PM as well. Again, there is lack of discussion of why the PM emission factors (and PAH toxicity equivalents) are so different, and no attempts to examine the link between PM/PAH and the fuel itself.*

The referee indeed raised very challenging questions. As this is a pioneering study and rather limited in its extent (limited number of test burns, burning conditions, etc.), it is not possible to answer to any of these questions in a responsible way from the results. Further dedicated studies are clearly warranted to give scientifically sound explanations. We just hypothesize that the propagation of flame and heat inside the plastic sample specimens might be responsible for the observed high EFs.

> *Page 6, Line 208: lower case "rag" was used here, but upper case "RAG" was used earlier.*

Done.

> *Page 6, line 232: What does "reactive share of PAHs" mean? It is not clear what reactivity the authors are referring to.*

It is a typo "Reactive share" should be "relative share".

> *Page 7, lines 236-261: I wonder if the difference has to do with volatility. What do the volatility distribution of PAH look like between the samples? Is it because in general plastics produce "heavier" PAHs, and the other samples generate "lighter" PAHs? (I understand the toxicity discussion and it is very important, but I am looking for some more information that would tell us how these PAHs are generated.)*

This issue is closely connected to the question raised above (Page 6, Line 200). See our answers there. Combustion conditions inside plastic specimens should be unique and seem to produce markedly different volatility distributions of PAHs. But further studies are clearly warranted.

> *Conclusions: I don't really see the point of repeating all the numbers if they are already shown in the table and in the figures.*

In the conclusion just a few exemplary numbers are given for illustrating the main conclusions.

> *Figure 2: what does O2 rise and CO2 drop at some times? Is this when the combustion chamber is open to place the sample?*

The peak in the CO_2 concentration represents the combustion of a small specimen of a given waste. During the sampling several waste specimens were combusted successively. Indeed, the concentration of O_2 increased when a specimen was placed into the combustion chamber.

> *I don't really see why it is necessary to have both a table and a figure showing essentially the same data. (Table 1 and Figure 3, Table 2 and Figure 4)*

The figures illustrate and compare the magnitude of the relative (to wood) emission factors of the different waste types, whereas the tables list absolute emission factors (per unit mass burned) which may be directly used in compiling emission inventories and air quality modeling.

Emission factors for PM_{10} and PAHs from illegal burning of different types of municipal waste in households

András Hoffer[1], Beatrix Jancsek-Turóczi[2], Ádám Tóth[2], Gyula Kiss[1,2], Anca Naghiu[3], Erika Andrea Levei[3], Luminita Marmureanu[4], Attila Machon[5], András Gelencsér[1,2]

[1] MTA-PE Air Chemistry Research Group, 10 Egyetem Street, H-8200 Veszprém, Hungary

[2] Institute of Environmental Sciences, University of Pannonia, 10 Egyetem Street, H-8200 Veszprém, Hungary

[3] Research Institute for Analytical Instrumentation Subsidiary, National Institute for R&D for Optoelectronics, 67 Donath Str., RO-400293 Cluj-Napoca, Romania

[4] Remote Sensing Department, National Institute of R&D for Optoelectronics, 409 Atomistilor Str., Magurele, Ilfov, Romania

[5] Air Quality Reference Centre, Hungarian Meteorological Service, 39 Gilice Square, H-1181 Budapest, Hungary

Correspondence to: András Gelencsér (gelencs@almos.uni-pannon.hu)

Abstract. It is a common practice in the developing countries and in some regions of Europe that solid wastes generated in the households (e.g. plastic beverage packaging and other plastic wastes, textile wastes, fibreboards, furniture, tyres, and coloured paper waste) are burned in wood- or coal-fired stoves during the winter months. In Europe, the types and volume of municipal waste burned in households is virtually unknown because these activities are illegal and not recorded, with the exception of a few media reports or court cases. Even though particulate emissions from illegal waste burning pose significant hazard to human health due to the combination of excessive emission factors (EFs) and uncontrolled chemical composition, there is scarce information on the specific EFs for PM_{10} and polycyclic aromatic hydrocarbons (PAHs) in the scientific literature. In this work, controlled combustion tests were performed with 12 different types of municipal solid waste and particulate emissions were measured and collected for chemical analysis. Absolute EFs for PM_{10} and PAHs as well as the benzo(a)pyrene toxicity equivalent of the latter are reported for the first time for the indoor combustion of 12 common types of municipal solid waste that are frequently burned in households worldwide. It was found that the PM_{10} emission factors from the combustion of wood-based waste samples were about twice that of firewood, whereas EFs in the range of 11–82 mg g^{-1} were obtained for different types of plastic waste. The latter were also found to emit exceptionally high amounts of PAHs, by a factor of 50–750 more than upon the combustion of dry firewood under the same conditions. Since the more toxic 4–6 ring PAHs were predominant in the particulate emission from plastic waste burning, BaP equivalent toxicity was up to 4100 times higher than that from wood combustion.

1 Introduction

Billions of people use solid fuels (wood, coal or agricultural waste) as the main source of household energy worldwide (Anenberg et al., 2013). It is well known that fossil fuel combustion and biomass burning are the two most important sources of fine particulate matter in the atmosphere (Simoneit et al., 2002). Karagulian et al.

(2015) estimated that domestic fuel burning (wood, coal and gas) might contribute up to 32% of $PM_{2.5}$ emissions, and up to 45% of PM_{10} emissions in Central and Eastern Europe (globally 20% and 15%, respectively). In Europe solid fuels (primarily wood but in some countries also coal) are extensively used for home heating. Residential wood combustion was found to be the main emission source of fine particles all over Europe in winter (Tissari et al., 2008; Puxbaum et al., 2007; Gelencsér et al., 2007; Marmureanu et al., 2020). Emissions from wood burning were estimated to contribute to about 30% of the $PM_{2.5}$ fraction in Portugal (Goncalves et al., 2012). In Lombardia somewhat lower contributions (5−25% to PM_{10}) were estimated (Pastorello et al., 2011), even in the city of Milan (Piazzalunga et al., 2011). Based on recent studies fine particles from wood combustion may cause severe health effects such as lung cancer, chronic lung and heart diseases (Rafael et al., 2015; Bai et al., 2015), contribute to visibility reduction (Pipal and Satsangi, 2015), and even modify synoptic conditions (Tiwari et al., 2015).

On top of the significant emissions from the burning of solid fuels in households there is another existing yet largely unknown source of atmospheric pollution, the illegal burning of municipal solid waste in households in several countries of the world. In Hungary, two independent polls have recently revealed that 2−10% of the population burn their wastes in stoves on a regular basis (Kantar Hoffman LTD., 2017; Századvég Foundation, 2018). The major types of waste burned were treated wood (furniture, oriented strand board (OSB), hardboard, painted wood, plywood), clothes, plastics, tyres, and used oil. In many villages in Romania the burning of household waste is an everyday practice due to the lack of organised waste collection system. Burning any type of waste poses excessive risks to the health of people living in those areas since a plethora of toxic, carcinogenic or mutagenic compounds are emitted in immense quantities compared to the burning of authorised solid fuels such as dry fuel wood or high-quality coal (Lemieux et al., 2004; Estrellan and Lino 2010; Gullett et al., 2010; Wiedinmyer et al., 2014). Since burning any kinds of municipal waste in households is strictly prohibited all over Europe, understandably such emissions are not included in many emission inventories (Wiedinmyer et al., 2014). Being an illegal activity, even its magnitude is mostly unknown apart from a few public reports of NGOs, media coverages, and documented court cases. More interestingly, there has been no single systematic study on the EFs for burning abundant types of solid waste in households in the scientific literature. There are only a small number of scientific papers on the burning of different types of (mostly plastic) waste with the specific focus of finding organic tracers for their tracking in atmospheric particulate matter (Simoneit et al., 2005; Tomsej et al., 2018; Gu et al., 2010; Kumar et al., 2015). This is surprising in the light of the fact that worldwide solid waste burning is a known and important source of particulate pollution. Among, there are only a handful of studies that report EFs of $PM_{2.5}$ for the open burning of municipal mixed solid waste, in the order of 10 g kg^{-1} (Christian et al., 2010; Park et al., 2013; Jayarathne et al., 2018).

PAHs are among the most hazardous combustion products due to their carcinogenic and mutagenic nature (Bjorseth, 1983; Kim et al., 2013). Among them 5-ring PAHs such as benzo(a)pyrene, benzofluoranthenes, dibenzo(a,h)antracene are known as highly carcinogenic compounds. The EFs of PAHs may vary for different waste types, but such data are scarcely available in the scientific literature though absolute EFs would be essential to assess the impact of residential waste burning on air quality. To the best of our knowledge there has been only one study reporting PM and PAHs emission factors for co-combustion of PE and PET waste with beech logs (Tomsej et al., 2018). In this study EFs for total PAH and PM_{10} were determined for 12 waste types and compared to those of wood burning.

2 Material and methods

2.1 Waste samples for combustion tests

The types of solid waste specimens for the combustion tests were selected based on their abundance in households and available information on illegal waste burning practices in Hungary. Polyethylene terephthalate (PET), Polystyrene (PS), Polypropylene (PP), Polyethylene (PE), Polyvinyl chloride (PVC) Polyurethane (PU), Acrylonitrile Butadiene Styrene (ABS), tyre (TR), OSB, laminated Melamine Low-Density Fibreboard (LDF), rag (RAG), paper (PAP), and firewood (WOOD) were selected for waste combustion tests (Figure 1).

Figure 1 Solid waste specimens prepared for combustion tests.

The PET waste samples were prepared from beverage plastic bottles (volume 1.5 and 2 L) without cap and label. The PS waste samples were hard cups/pots of dairy products (yoghurt and pudding) without aluminium foil cap, and pieces of expanded PS insulation board (80 mm × ~100 mm × ~100 mm). The hard and expanded PS waste sample specimens were burned separately. The PP waste sample specimens were the mixture of plastic cups/pots of dairy products (sour cream and pudding) without paper labels and aluminium foils and quartered plastic tray of meat. The PE waste specimens were prepared from the mixture of high and low density polyethylene (HDPE and LDPE). The HDPE and LDPE fractions consisted of plastic caps of beverage bottles and pieces of various foils and plastic bags, respectively. The PVC waste samples consisted of soft packaging, small pieces of vinyl flooring, and hard plastic water pipe. The PU waste samples consisted of pieces of packing sponge (average size: 120 mm × 100 mm × 15 mm). The ABS samples were shredded pieces of stands of computer monitors (average size: 40 mm × 40 mm × 20 mm). The OSB samples consisted of slices of OSB material of different brands (average size: 120 mm × 100 mm × 20 mm). The LDF samples were pieces (average size: 130 mm × 100 mm × 20 mm) of different fibreboards including coloured laminated coating and plastic border. The TR sample specimens consisted of pieces of a new and old tyre of a van and a passenger car, respectively (average size: 80 mm × 40 mm × 15 mm). The RAG samples consisted of a mixture of cotton, polyester, and polyamide fabrics from different clothes. Ball-shaped specimens (average weight: ~70 g) of two types of PAP samples (colourful glossy coated paper and uncoated paper from advertising flyers and newspaper) were burned separately. The WOOD samples consisted of pieces of logs (average weight: ~130 g) of Turkey oak (*Quercus cerris*) and black locust (*Robinia pseudoacacia*). All sample specimens were weighted with an analytical balance before the combustion tests. Authorisation for conducting controlled waste burning tests was granted by the Department of Environmental Protection and Nature Conversation of Veszprém District.

2.2 Experimental conditions of the combustion tests

Combustion tests were carried out in a commercially available cast iron stove (type: Servant S114, heating power: 5 kW). The stove was heated up with smouldering charcoal for ~1 hour prior to the start of combustion tests which produced very low particulate emission baseline throughout the duration of the measurements. During each combustion test 1–10 aliquots of solid waste were burned depending on the emission characteristics of the given waste type. The mass of each sample aliquot was measured with an analytical balance and was recorded. The stove used was a commercially available model which allowed setting the air supply entering the combustion chamber of the stove through an adjustable slit. There were two endpoints of this slit, thus either larger (high air supply ratio) or smaller volume of fresh air (low air supply ratio) was allowed into the combustion zone. Each type of waste (except PAP) was burned at high, combined and low air supply resulting in different temperatures and conditions inside the oven, but it was largely independent from the type of waste combusted in the stove at any given air supply ratio. The temperature of flue gas was measured before and after each measurement by a K-type thermocouple thermometer (maximum temperature 1000 °C; Testo 925) in the stack 11 cm above the exhaust opening of the stove, respectively. Between the different sample runs the stove and the stack were heated up to above 700 °C for a minimum of 10 minutes to minimise cross-contamination between combustion tests with different waste types. The temperature of the flue gas served as an indicator for the experimental conditions of each combustion test run. The temperature values of the flue gas during the experiments were stable as the mean temperature values were 299 °C (SD 11 °C) and 233 °C (SD 10 °C) at high and low air supply, respectively (see Fig. S1).

The mixing ratios of CO_2 and O_2 in the flue gas were measured with a CO_2−O_2 analyser (Servomex) at the end of the stack. At the end of the stack a small amount of the flue gas was introduced through a brass tubing (ø 5 mm) into a dilution unit of a volume of 80 L and was diluted with ambient air. The concentration of CO_2 in the diluted flue gas was also monitored with a CO_2 analyser (SensAir) at the inlet of the filter sampling head. The dilution ratio was determined from the ratio of the measured concentrations of CO_2 at the end of the stack and in the dilution unit (taking into account the ambient CO_2 concentration as well). The mean dilution ratio was about 81.2 (SD 12) (see Fig. S2) which is independent of the air supply but may depend on the progress of the burning process. The physical processes took place during the applied sampling procedure were very similar to those occurring after the emission of smoke from the chimney into open air as in both cases the smoke cools and dilutes rapidly.

Figure 2 shows the variation of the CO_2 and O_2 mixing ratios at the end of the stack and the CO_2 mixing ratio in the diluted flue gas for a PS sample. Each peak corresponds to the burning of a single aliquot of the solid waste. The collections of each PM_{10} aerosol sample were started after reaching stabile baselines of CO_2 and O_2 concentration values and were finished after the return of stable baselines at the end of combustion of all aliquots of solid wastes.

Figure 2 Example (PS-F9 sample) for the variations of the measured CO_2 and O_2 mixing ratios at the end of the stack and the CO_2 mixing ratio in the diluted flue gas during combustions tests.

PM_{10} aerosol samples were collected on quartz filters of 150 mm in diameter (Advantec QR-100 quartz fiber, binder free) with a high-volume aerosol sampler (flow rate 32 m^3 h^{-1}; Kalman System Co., Hungary) at the dilution unit. Blank samples were also collected for each waste type. The blanks represent background measurements during which only charcoal was burned. The sampling times of the blanks were comparable with those of the samples. For each waste type one blank sample was collected with combined air supply settings. The quartz filters were conditioned at a temperature of 20 ± 1 °C and relative humidity (RH) of 45−50% for three days and 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 and collected by a data acquisition system. The weighted filters were stored in glass petri dishes (preheated at 450 °C) prior to sampling, whereas the exposed filters were stored in the freezer in glass petri dishes wrapped into aluminium foil until conditioning and measurements.

Table S1 lists the key parameters of the combustion tests, including the type and mass of sample specimens, the air supply settings, the number of test burns (by air supply settings), and the measured blank corrected PM_{10} mass on each filter. The mass of the waste sample specimens burned was optimised in preliminary tests to yield PM_{10} concentrations of about the same magnitude in each combustion test. Since different waste types yielded vastly different particulate emissions upon burning (e.g. PS, PP, PE, PVC, PU, ABS, and TR were superemitters compared to PET, OSB, LDF, RAG, PAP and WOOD), this step was necessary to avoid massive overloading of the filters and the measuring instrumentation. Thus the measurements were comparable and the measured mass of PM_{10} on filters was kept in the range of 5.4 mg and 37.2 mg for all combustion tests.

2.3 Analysis of PAHs in the filter samples

The amount of PAHs in the filter samples was determined by analysing a filter spot with a diameter of 1.4 cm. First PAHs were extracted with 4.5 mL hexane in an ultrasonic bath for 15 minutes then the extract was filtered through ashless quantitative (Grade 44, Whatman, UK) filter paper and cleaned on 0.5 g Florisil adsorbent. The cleaned extract was gently evaporated to dryness and re-dissolved in 1.0 mL acetonitrile. The concentration of 15 PAHs out of the 16 EPA priority pollutant PAHs was determined in the final aliquot by HPLC (200 Series, Perkin Elmer, Shelton, CT, USA) on a Inertsil ODS-P 5µm, 4.6 × 150 mm (GL Sciences Inc. Tokyo, Japan) column by using water-acetonitrile binary gradient elution and time-programmed fluorescence detection.

The ambient temperature affects the gas-to-particle partitioning of the compounds especially those of the semi-volatile ones. During the sampling the ambient temperature varied between 16.1 °C and 26.0 °C, on average it was 20.3 °C, which is higher than typical ambient temperatures during the heating season. It implies that some semi-volatile species might be lost to the gas phase upon sampling, thus measured EFs for PM_{10} and total PAHs are likely lowerbound estimates for real-life conditions. It should be added that the standard procedure of the gravimetric PM10 measurements include the conditioning of the exposed filters for 48 hours at 20°C (and RH=50%) which might also result in some loss of semi-volatile compounds. Concerning PAHs, at the sampling temperatures some of the more volatile 2- and 3-ring compounds might have been lost to the gas phase as compared to the case at ambient temperatures in winter, and were likely to some extent underrepresented in the filter samples. These potential losses, however, do not influence the conclusions regarding PAH relative toxicity due to the very low toxicity of the 2-3-ring compounds.

The extraction efficiency was tested by sequential extraction of the same filter spots in two consecutive steps. In the second extract the amount of PAHs was found to be only 1–3% of that present in the first extract. These results evidenced that under the experimental conditions applied (solvent to filter ratio, time of extraction) 97–99% of PAHs were extracted in the first step. Therefore, the extraction was not continued and the sum of the amount of PAHs in the two extract was considered as 100%. Because of the high efficiency of the first extraction step the samples were extracted only once and the results were corrected with the extraction efficiency (97%–99%). Recovery of PAHs from the extract was also studied by performing the clean-up procedure on diluted PAH standard solutions. Average recovery of 98 % (SD=8.9 %) was obtained for the 12 PAHs investigated in this study. The analytical results were corrected for both extraction efficiency and recovery of the sample preparation.

3 Results and discussion

3.1 Emission factors for PM_{10} from waste burning

The PM_{10} EFs of the fuel wood and different solid wastes were found to be highly variable (from ~2.1 to ~81.5 mg g^{-1}), the lowest values being representative for the wood burning. In the literature, the $PM_{2.5}$ EF of wood burning in traditional woodstoves varies between ~2.8 and ~13.3 mg g^{-1}, whereas the PM_{10} EF from a chimney-type (eco-labelled) woodstove is smaller (~1.1−2.9 mg g^{-1}, Querol et al., 2016). Kistler et al., (2012) investigated the PM_{10} EF of 12 Central European wood types from a chimney-type wood stove and found that it varied between 0.3 and 3.6 mg g^{-1}. The values for turkey oak and black locust were 1.0 and 1.1 mg g^{-1}, respectively. For the burning of oak logs in two different stoves Schmidl et al., (2011) reported PM_{10} EF of 1.2−1.4 mg g^{-1}. In our measurements the PM_{10} EF from the burning of mixed turkey oak and black locust varied between 1.3 and 3.2 (on average 2.1 mg g^{-1}, SD 0.7 mg g^{-1}), indicating that the obtained EFs agree quite well with those reported in recent studies.

Compared to the EFs of burning plastics, the PM_{10} EFs of burning wood-based materials were found to be largely similar to each other, though the burning of OSB and LDF (both contain glue, the LDF surface coating and edge tape as well) release twice as much PM_{10} (~3.2 mg g^{-1}, SD 1.3 mg g^{-1} for LDF, and 5.2 mg g^{-1}, SD 1.4 mg g^{-1} for OSB) into the atmosphere as dry firewood under similar burning conditions (Fig. 3).

Figure 3 PM_{10} emission factors for burning of different waste types relative to the burning of dry fuel wood with a fixed experimental setup.

The EF of burning RAG (8.7 mg g^{-1}, SD 0.22 mg g^{-1}) is between the EF of PET and wood-based materials. This is not surprising as on average 46% of the weight of the burned RAG was cotton and 43% was PET-based material. The EF for PET (on average 11 mg g^{-1}, SD 1.6 mg g^{-1}) is roughly five times higher than that of the WOOD. The only available EFs reported for PET co-combustion in a boiler are between 1.8 ± 0.3 and 5.8 ± 0.5 mg g^{-1} depending on the combustion conditions (Tomsej et al., 2018). The differences between our and the reported EF values can be explained by the vastly different burning conditions (20 and 5 kW nominal power output for the boiler and the stove, respectively, and no fuel wood was co-fired in our experiments as against in those reported by Tomsej et al.). Our findings indicate that the burning of a given mass of PE and/or PU release on average 1 order of magnitude more PM_{10} into the atmosphere than wood burning. The combustion of PP, tyre and PVC is even more polluting as 16 times more PM_{10} is released into the atmosphere when burned under similar conditions. Somewhat surprisingly, our results also agree well with the few available EFs (for PM2.5) for the open air burning of mixed (presumably mostly plastic) waste reported in the literature (10.5 and 7.37 mg g^{-1}) (Christian et al., 2010 and Jayarathne et al., 2018, respectively). The tyres already contain soot and inorganic fillers which might contribute to the increased PM_{10} emission of burning. The styrene containing materials yielded the highest EF among the investigated components. The PM_{10} EF for the burning of PS was on average 53.1 mg g^{-1} (SD 15 mg g^{-1}) and that of the expanded polystyrene was even higher (81.5 mg g^{-1}, SD 27 mg g^{-1}). On average the highest PM_{10} EF was obtained for the copolymer of styrene with butadiene and acetonitrile. The absolute EFs for all waste types are summarised in Table 1.

Table 1 The absolute emission factors of PM_{10} (mg g^{-1}), total PAHs (mg PAHs kg^{-1} fuel) and total PAHs expressed in BaP toxicity equivalent (mg kg^{-1} fuel) from wood burning and residential waste burning.

3.2 Emission factors of PAHs from waste burning

The EFs of total PAHs (sum of 12 EPA priority pollutant PAHs from 3-ring phenanthrene to 6-ring indeno(1,2,3-cd)pyrene) during the combustion of different waste types were determined (Table 1). Both wood and wood-based waste types were characterised by EFs below 5 mg kg^{-1}. The lowest EF was obtained for WOOD followed by PAP, LDF and OSB. At least an order of magnitude higher EFs were measured for the other waste types investigated in this study. The average values covered a wide range from 21 mg kg^{-1} (SD 19 mg kg^{-1}) for RAG to 257 mg kg^{-1} (SD 103 mg kg^{-1}) for ABS. The average total PAH EFs were similar for RAG, PE, TR and PET while it was higher for PVC, PU and PP but remained under 100 mg kg^{-1} for all of these waste types. The highest EFs were measured during the combustion of PS and ABS exceeding 100 mg kg^{-1}. The relatively high SD values are the consequence of varying conditions (air supply) applied during the burning experiments as described in section 2.2.

The EFs of the individual PAHs (Table 2 in the SI) are different for wood and waste burning. While the combustion of WOOD resulted in the emission of primarily phenanthrene and 4-ring PAHs (fluoranthene, pyrene, benzo(a)anthracene and chrysene) and the contribution of 5- and 6-ring compounds was only a few per cents, the latter compounds accounted for 14–45% of the total PAH emission in the different waste types. This shift in the chemical profile has a considerable effect on the toxicity of the particles emitted as discussed later. The availability of PAH emission data from the burning of any given type of waste is rather limited in the literature. Tomsej et al. (2018) studied the co-combustion of wood and polyethylene plastics (PE and PET). They burned 93 w/w% beech log and 7 w/w% PE in a 20 kW boiler and obtained total PAH EFs of 11 mg kg^{-1} (SD 1.2 mg kg^{-1}) and 16 mg kg^{-1} (SD 1.6 mg kg^{-1}), in the particulate phase for the 12 compounds investigated in our study under nominal and reduced output conditions, respectively. The corresponding values for the co-combustion of 93 w/w% beech log and 7 w/w% PET were 8.5 mg kg^{-1} (SD 1.0 mg kg^{-1}) and 19 mg kg^{-1} (SD 3.6 mg kg^{-1}). These results indicate the influence of operational conditions on the emission factors of PAHs. Furthermore, the total PAH EF values obtained for the mixtures of beech/PE and beech/PET by Tomsej et al. (2018) were between the total PAH EFs obtained for WOOD (0.34 mg kg^{-1}, SD 0.19 mg kg^{-1}) and PE (31 mg kg^{-1}, SD 23 mg kg^{-1}) or WOOD and PET (32 mg kg^{-1}, SD 14 mg kg^{-1}) in our study, respectively. Maasikmets et al. (2016) studied the EFs of 4 PAHs emitted from burning municipal solid waste mixed with wood in domestic heaters. They reported emission factors of 0.41 mg kg^{-1}, 0.18 mg kg^{-1}, 0.12 mg kg^{-1}, and 0.10 mg kg^{-1} for benzo(a)pyrene, benzo(b)fluoranthene, benzo(k)fluoranthene and indeno(1,2,3-cd)pyrene, respectively. As a comparison, Lemieux (1997) found EFs of 1.4 mg kg^{-1}, 1.86 mg kg^{-1}, 0.67 mg kg^{-1} and 1.27 mg kg^{-1} for the same compounds from barrel burning of household waste. These emission factors are comparable to those found in our experiments and summarised in Table S2.

In Figure 4 the EFs for total PAHs are shown for different waste types relative to wood burning. It is clearly visible that even the combustion of wood-based waste types (PAP, LDF, OSB) generated considerably more (by a factor of 3–8) PAHs than the burning of wood. The relative PAH emissions were higher by more than a factor of 50 in case of burning RAG, PE, TR and PET, and were well over a factor of 100 for PVC, PU, PP, PS and ABS. These extreme emission factors underline the severe hazard associated with the illegal burning of solid wastes in households.

Figure 4 Emission factors of total PAHs for different waste types relative to the burning of dry fuel wood with a fixed experimental setup.

From the PM_{10} mass collected and the amount of total PAHs determined on the filter the PAH content of the PM_{10} fraction generated from each combustion experiment was calculated. The lowest mean PAH content of the PM_{10} aerosol was obtained for WOOD burning (0.16 µg mg^{-1}, SD 0.08 µg mg^{-1}) followed by the wood-based wastes OSB (0.52 µg mg^{-1}, SD 0.20 µg mg^{-1}), PAP (0.53 µg mg^{-1}, SD 0.40 µg mg^{-1}) and LDF (0.53 µg mg^{-1}, SD 0.12 µg mg^{-1}). For the other waste types the mean PAH content increased from 0.70 µg mg^{-1} (SD 0.16 µg mg^{-1}) (TR) through 1.1 µg mg^{-1} (SD 0.52 µg mg^{-1}) (PE), 1.4 µg mg^{-1} (SD 0.44 µg mg^{-1}) (PVC), 1.8 µg mg^{-1} (SD 2.0 µg mg^{-1}) (RAG) to 2.5 µg mg^{-1} (SD 1.8 µg mg^{-1}) (PS). The highest PAH content was observed when burning PET (3.0 µg mg^{-1}, SD 1.5 µg mg^{-1}), PU (3.0 µg mg^{-1}, SD 0.92 µg mg^{-1}), ABS (3.1 µg mg^{-1}, SD 0.86 µg mg^{-1}) and PP (3.1 µg mg^{-1}, SD 1.5 µg mg^{-1}). It is clearly visible that the relative share of PAHs in PM_{10} emitted is typically an order of magnitude higher in plastic combustion as compared to wood burning. The relatively high PM_{10} but low PAH emission factors of tyre combustion (as shown in Table 1) can be explained by the presence of inorganic components in the material of tyres (e.g. inorganic fillers).

It is important to note that in addition to the total PAH emission factors the emission profile (i.e. the relative contribution of individual PAHs) should also be investigated when the health effects are studied since the toxicity of PAHs varies from compound to compound. Among the 16 EPA priority pollutant PAHs 5-ring compounds such as benzo(a)pyrene, dibenzo(a,h)anthracene, benzofluoranthenes, the 4-ring benzo(a)anthracene and the 6-ring indeno(1,2,3-cd)pyrene were found to be the most carcinogenic compounds (Nisbet and LaGoy, 1992; Safe, 1998 and references therein). In order to compare the toxicity of different samples toxic equivalency factors have been defined for PAHs. In this scale benzo(a)pyrene (BaP) has a value of 1 and the toxicity of the other 15 EPA priority pollutant PAHs varies from 0 to 1 (Nisbet and LaGoy, 1992, Safe, 1998 and references therein, EPA, 2007). There are numerous such toxicity scales but for the sake of comparability the toxic equivalency factors of the EPA (2007) were applied in our study similarly to the work by Tomsej et al. (2018). In this scale benzo(a)pyrene is taken into account with a toxicity equivalency factor of 1, benzo(a)anthracene, benzofluoranthenes, dibenzo(a,h)anthracene and indeno(1,2,3-cd)pyrene with a factor of 0.1 and chrysene with a factor of 0.01 and the toxicity of the other PAHs is neglected. By applying the BaP toxicity equivalency factors the overall toxicity of the PAHs emitted during wood burning and the combustion of different wastes was compared (Table 1). The EFs of total PAHs as expressed in benzo(a)pyrene toxicity equivalent covered a very wide range of more than three orders of magnitude. The burning of 1 kg wood (oak) produced PAHs with total toxicity equivalent of 4.2 µg of BaP (SD 5.5 µg). The combustion of the same amount of wood-based wastes led to the emission of PAHs equivalent of about 30 times more BaP. The combustion of PE, RAG, TR, PET and PU resulted in the emission of PAHs 280−800 more toxic than the PAHs released from the burning of the same amount of wood. The toxicity equivalent EFs for PE and PET reported in Table 1 (1.2 mg kg^{-1}, SD 1.1 mg kg^{-1} and 2.2 mg kg^{-1}, SD 1.4 mg kg^{-1}, respectively) were similar to those found by Tomsej et al. (2018) for mixtures of the above plastics with beech. They reported toxicity equivalent EFs of 2.3±0.5 mg kg^{-1} and 1.1±0.2 mg kg^{-1} for the mixture of PE and beech under nominal output and reduced output conditions, while the corresponding values for the mixture of PET and beech were 1.8 ± 0.6 mg kg^{-1} and 1.8 ± 0.3 mg kg^{-1}, respectively. Toxic PAHs in the highest amount were emitted from the combustion of PVC, PS, PP and ABS. The emission factors of total PAHs expressed in BaP toxicity equivalent were more than 3 orders of magnitude (!) higher for the combustion of these plastic wastes than the value obtained for wood burning. The very high emission of toxic

PAHs from the combustion of plastic wastes as compared to wood burning may follow from the synergic combination of three factors:

1) On a per mass basis significantly more PM$_{10}$ is emitted from the combustion of plastic waste than from the burning of dry fuel wood (see Figure 3, Table 1);
2) PM$_{10}$ released from plastic waste combustion contains more PAHs per unit mass than PM$_{10}$ generated from wood burning;
3) The distributions of PAHs emitted from plastic waste burning differ markedly from that produced in wood burning. The combustion of plastic wastes results in the formation of the significantly more carcinogenic 4–6-ring compounds.

It should be added, however, that the burning conditions affect the profile and total amount of PAHs considerably. The low amount of 5-ring and 6-ring PAHs in the particles emitted from wood burning resulted in very low toxicity equivalent EFs and, consequently, led to high relative toxicity of the other wastes as compared to wood.

4 Conclusion

The PM$_{10}$ emission factors, total PAH emission factors and benzo(a)pyrene toxicity equivalent total PAH emission factors were determined for waste burning under controlled combustion conditions for the first time. The EF values were established for combustion of wood, paper, LDF, OSB, rag, PET, PE, PU, PP, tyre, PVC, PS and ABS which are all abundant waste types frequently burned in households and open fires in the developing countries and even in Europe. The determined PM$_{10}$ EF for wood burning was about 2.1 mg g^{-1} (SD 0.7 mg g^{-1}) which shows good correlation with the results of recent studies. The PM$_{10}$ EFs from the burning of other wood-based materials (PAP, LDF and OSB) were only slightly higher, while from the combustions of RAG and PET were about five times higher than the EF of wood burning. The EFs for combustions of PE and PU were about 18 mg g^{-1} (SD 7.2 mg g^{-1}) and 22 mg g^{-1} (SD 8.4 mg g^{-1}) indicating nearly tenfold PM$_{10}$ emission from burning of these waste types. Even higher PM$_{10}$ EFs were determined during the combustion of PP, TR and PVC which were 33 mg g^{-1} (SD 18 mg g^{-1}), 35 mg g^{-1} (SD 6 mg g^{-1}) and 35 mg g^{-1} (SD 10 mg g^{-1}), respectively. Finally, the highest PM$_{10}$ EF values were established for the combustion of PS and ABS which may reach 53 mg g^{-1} (SD 15 mg g^{-1}) and 82 mg g^{-1} (SD 27 mg g^{-1}).

The total PAH emission factors varied in an even wider range. Burning of oak resulted in mean total PAH emission factor of 0.34 mg kg^{-1}, (SD 0.19 mg kg^{-1}), while wood based wastes (PAP, LDF and OSB) produced mean total PAH emission factors from 1.2 to 2.7 mg kg^{-1} (SD 0.83–1.4 mg kg^{-1}). Much higher total PAH emission factors were obtained for RAG, TR and plastic wastes ranging from 21 mg kg^{-1} (SD 19 mg kg^{-1}) to 257 mg kg^{-1} (SD 103 mg kg^{-1}). These high total PAH emission factors are the consequence of the higher PM$_{10}$ emission factors of plastic wastes as well as the higher total PAH content of unit mass of PM$_{10}$ released during combustion. To compare the hazard of residential waste combustion with wood burning the total PAH EFs were converted into benzo(a)pyrene equivalent emission factors. It was found that the toxicity of PAHs emitted during the combustion of wood-based wastes (PAP, LDF and OSB) was about 30 times higher than that of PAHs released during the burning of oak. For RAG, TR and plastic wastes this ratio ranged between 280 and 4050 as a consequence of the different composition of PAHs emitted during the combustion of different waste types.

These results clearly show that residential waste burning (especially combustion of tyres and plastics) poses a serious hazard on human health for numerous reasons: waste combustion may produce considerably more PM_{10} particles than wood burning, the particles contain more PAHs, and the PAHs formed during waste combustion are more carcinogenic than those released from wood burning. These findings underline the importance of concerted efforts of municipalities, authorities and NGOs to phase out the illegal burning of solid wastes in households. This would help significantly improve local air quality and reduce the number of limit exceedances of $PM_{10}/PM_{2.5}$. But more importantly, the health risks associated with air pollution would be reduced disproportionately more than would follow from the reduction in $PM_{10}/PM_{2.5}$ concentrations due to the extremely hazardous composition of particulate matter emitted during the combustion of wastes in households.

Data availability. Data used in this study are available from the first author upon request (hoffera@almos.uni-pannon.hu).

Author contributions. AH, BJT and ÁT collected the aerosol samples. GYK, AN, EAL and AM performed and/or coordinated the analytical measurements. BJT, AH, GYK, LM and AG were involved in the scientific interpretation and discussion of the results as well as in manuscript preparation. All co-authors commented on the paper.

Competing interests. The authors declare that they have no conflict of interest.

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Figure 1 Solid waste specimens prepared for combustion tests.

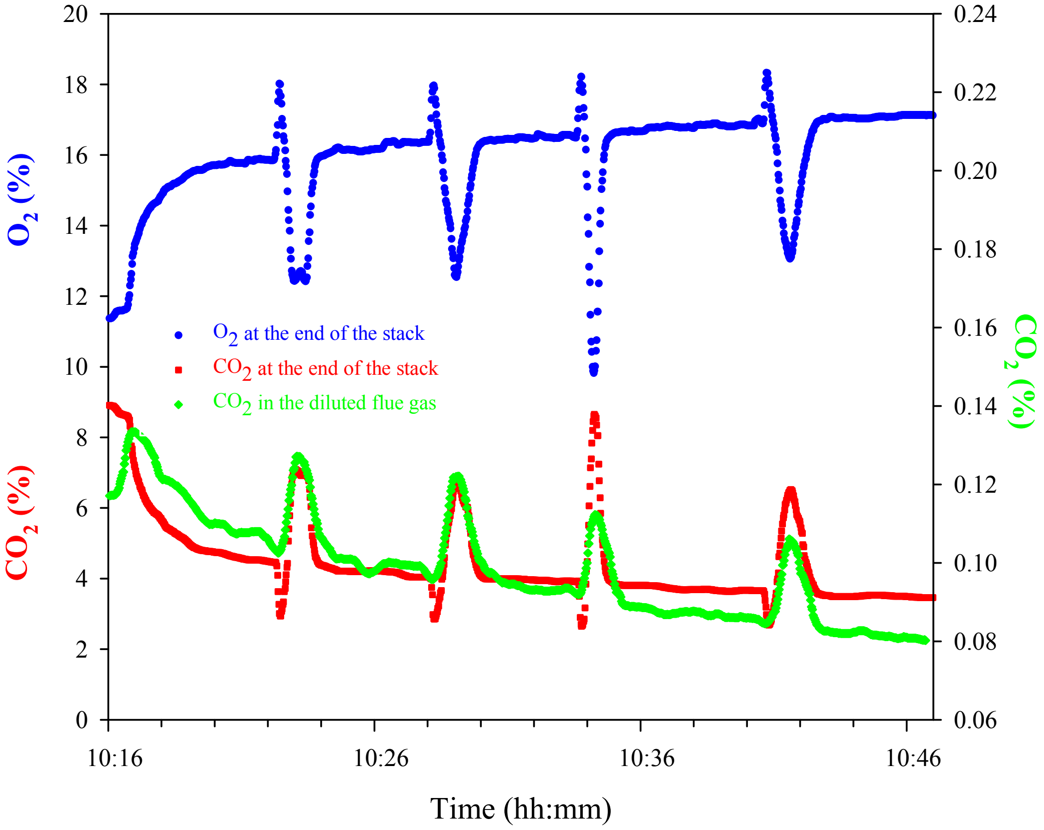

Figure 2 Example (PS-F9 sample) for the variations of the measured CO_2 and O_2 mixing ratios at the end of the stack and the CO_2 mixing ratio in the diluted flue gas during combustions tests.

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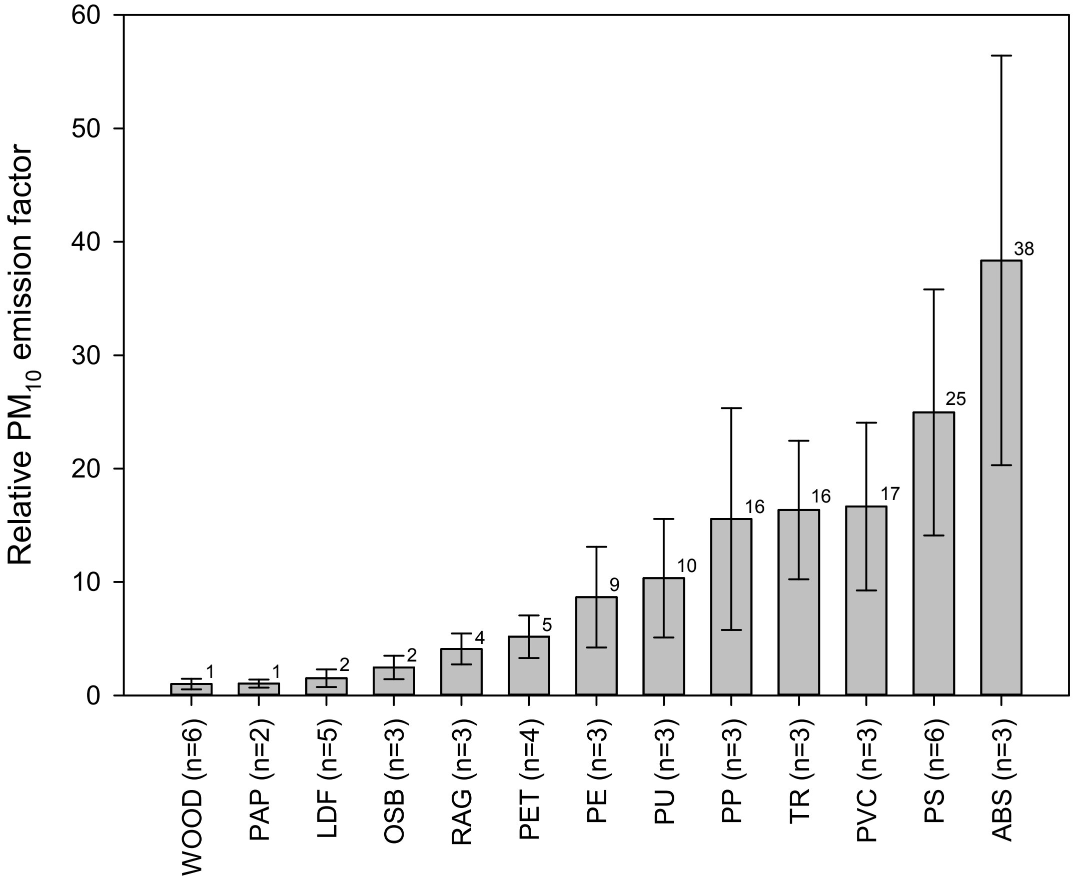
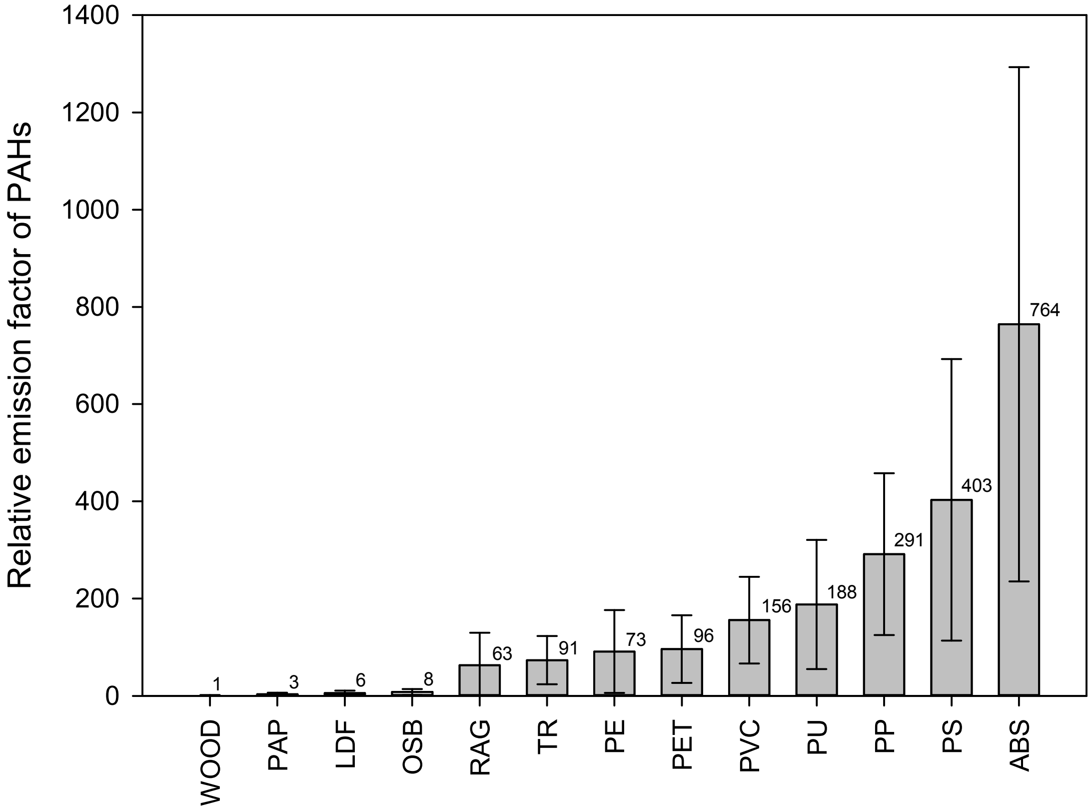


Figure 3 PM_{10} emission factors for burning of different waste types relative to the burning of dry fuel wood with a fixed experimental setup.

Table 1 The absolute emission factors of PM$_{10}$ (mg g^{-1}), total PAHs (mg PAHs kg^{-1} fuel) and total PAHs expressed in BaP toxicity equivalent (mg kg^{-1} fuel) from wood burning and residential waste burning.

Waste types	EFs of PM$_{10}$		EFs of total PAHs		EF of total PAHs expressed in BaP toxicity equivalent		
	Mean	SD	Mean	SD	Mean	SD	Relative to wood
WOOD	2.1	0.70	0.34	0.19	0.0042	0.0055	1
PAP	2.2	0.11	1.2	0.83	0.16	0.12	37
LDF	3.2	1.3	2.0	1.3	0.14	0.12	32
OSB	5.2	1.4	2.7	1.4	0.14	0.094	34
RAG	8.7	0.22	21	19	1.63	2.0	390
PET	11	1.6	32	14	2.2	1.4	520
PE	18	7.2	31	23	1.2	1.1	280
PU	22	8.4	63	27	3.4	0.12	800
PP	33	18	98	8.3	16	4.7	3700
TR	35	6.0	25	9.1	2.0	1.8	480
PVC	35	10	52	5.1	5.3	1.9	1300
PS	53	15	135	60	11	4.8	2500
ABS	82	27	257	103	19	12	4500



515 **Figure 4 Emission factors of total PAHs for different waste types relative to the burning of dry fuel wood with a fixed experimental setup.**