Emission factors for PM$_{10}$ and polycyclic aromatic hydrocarbons (PAHs) from illegal burning of different types of municipal waste in households

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Abstract. It is a common practice in developing countries and in some regions of Europe that solid wastes generated in 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 a 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 (BaP) 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$^{-1}$ (a factor of 5–40 times higher than that of dry firewood under the same conditions) were obtained for different types of plastic waste. The latter were also found to emit exceptionally high quantities 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 Lombardy 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 Company, 2020; 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 an 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; Estrelan and Lino 2010; Gullett et al., 2010; Wiedinmyer et al., 2014). Since burning any kind 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 in a few public reports of NGOs, some media coverage and a few documented court cases. More interestingly, there has been no single systematic study on 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. There are only a handful of studies that report EFs of PM$_{2.5}$ for the open burning of municipal mixed solid waste, on the order of 10 g kg$^{-1}$ (Christian et al., 2010; Park et al., 2013; Jayarathe et al., 2018).

Polycyclic aromatic hydrocarbons (PAHs) are among the most hazardous combustion products due to their carcinogenic and mutagenic nature (Bjorseth, 1983; Kim et al., 2013). Among them five-ring PAHs such as benzo(a)pyrene, benzofluoranthenes and dibenz(a,h)anthracene 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 are essential to assessing the impact of residential waste burning on air quality. To the best of our knowledge there has been only one study reporting PM and PAH emission factors for the co-combustion of polyethylene (PE) and polyethylene terephthalate (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. PET, polystyrene (PS), polypropylene (PP), PE, polyvinyl chloride (PVC) polyurethane (PU), acrylonitrile butadiene styrene (ABS), tyre material (TR), OSB, laminated melamine low-density fibreboard (LDF), rag (RAG), paper (PAP) and firewood (WOOD) were selected for waste combustion tests (Fig. 1).

The PET waste samples were prepared from beverage plastic bottles (volume 1.5 and 2 L) without caps and labels. The PS waste samples were hard cups or pots of dairy products (yoghurt and pudding), without aluminium foil caps, 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 a mixture of plastic cups and pots of dairy products (sour cream and pudding), without paper labels and aluminium foil, and quartered plastic trays of meat. The PE waste specimens were prepared from a 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 foil and plastic bags, respectively. The PVC waste samples consisted of soft packaging, small pieces of vinyl flooring and hard plastic water pipes. 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 borders. The TR sample specimens consisted of pieces of a new and old tyres of a van and a passenger car, respectively (average size 80 mm × 40 mm × 15 mm).
RAG samples consisted of a mixture of cotton, polyester and polyamide fabrics from different clothes. Ball-shaped specimens (average weight $\sim 70 \text{ g}$) 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 $\sim 130 \text{ g}$) of Turkey oak ($\text{Quercus cerris}$) and black locust ($\text{Robinia pseudoacacia}$). 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 $\sim 1 \text{ h}$ prior to the start of combustion tests which produced a 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 specimen was measured with an analytical balance and was recorded. The stove used was a commercially available model which allowed for setting the air supply to enter the combustion chamber of the stove through an adjustable slit. There were two endpoints of this slit; thus either a larger (high air supply ratio) or a smaller (low air supply ratio) volume of fresh air 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 this 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 $^\circ \text{C}$; testo 925) in the stack 11 cm above the exhaust opening of the stove. Between the different sample runs the stove and the stack were heated up to above 700 $^\circ \text{C}$ for a minimum of 10 min to minimise cross-contamination between combustion tests with different waste types. The temperature of the flue gas served as an indicator of the experimental conditions of each combustion test run. The temperature values of

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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 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 that 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 in 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 during combustions tests.

![Figure 2. Example (PS-F9 sample) of the variations in 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.](image-url)

The quantity 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 of hexane in an ultrasonic bath for 15 min; then the extract was filtered through ashless quantitative (Grade 44, Whatman, UK) filter paper and cleaned on 0.5 g of Florisil adsorbent. The cleaned extract was gently evaporated to dryness and re-dissolved in 1.0 mL of acetonitrile. The concentration of 15 PAHs out of the 16 Environmental Protection Agency (EPA) priority pollutant PAHs was determined in the final aliquot by high-performance liquid chromatography (HPLC; Series 200, PerkinElmer, Shelton, CT, USA) on an Inertsil ODS-P 5 µm, 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 and 26.0 °C, on average it was 20.3 °C; which is higher than typical ambient temperatures during the heating season. This 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 lower-bound estimates for real-life conditions. It should be added that the standard procedure of the gravimetric PM$_{10}$ measurements includes the conditioning of the exposed filters for 48 h 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 two- and three-ring compounds might have been lost to the gas phase as compared to the case at ambient temperatures in winter, and they 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 two–three-ring compounds.

The extraction efficiency was tested by sequential extraction of the same filter spots in two consecutive steps. In the second extract the quantity 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 quantity of PAHs in the two extracts was considered to be 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 recovery of PAHs from the extract was also studied by performing the clean-up procedure on diluted PAH standard solutions. The 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 $\sim 2.1$ to $\sim 81.5$ mg g$^{-1}$), the lowest values being representative of the wood burning. In the literature, the PM$_{2.5}$ EF of wood burning in traditional woodstoves varies between $\sim 2.8$ and $\sim 13.3$ mg g$^{-1}$, whereas the PM$_{10}$ EF from a chimney-type (eco-labelled) woodstove is smaller ($\sim 1.1$–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 a 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$_{10}$ ($\sim 3.2$ 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).

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 5 times higher than that of 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 unlike in those reported by Tomsej et al., 2018). Our findings indicate that the burning of a given mass of PE and/or PU releases on
average 1 order of magnitude more PM$_{10}$ into the atmosphere than wood burning. The combustion of PP, tyre material 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 PM$_{2.5}$) 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.

### 3.2 Emission factors of PAHs from waste burning

The EFs of total PAHs (sum of 12 EPA priority pollutant PAHs from three-ring phenanthrene to six-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. EFs at least an order of magnitude higher 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 they were 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 Sect. 2.2.

The EFs of the individual PAHs (Table S2 in the Supplement) are different for wood and waste burning. While the combustion of WOOD resulted in the emission of primarily phenanthrene and four-ring PAHs (fluoranthene, pyrene, benzo(a)anthracene and chrysene) and the contribution of five- and six-ring compounds was only a few per cent, 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 four PAHs emitted from burning municipal solid waste mixed with wood in domestic heaters. They reported emission factors of 0.41, 0.18, 0.12 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, 1.86, 0.67 and 1.27 mg kg$^{-1}$ for the same compounds from the barrel burning of household waste. These emission factors are comparable to those found in our experiments and summarised in Table S2.

In Fig. 4 the EFs for total PAHs are shown 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 quantity 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) and 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 health effects are studied since the toxicity of PAHs varies from compound to compound. Among the 16 EPA priority pollutant PAHs five-ring compounds such as benzo(a)pyrene, dibenzo(a,h)anthracene and benzo(ghi)perylene; the four-ring benzo(a)anthracene; and the six-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. On 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). On this scale benzo(a)pyrene is taken into account with a toxicity equivalency factor of 1; benzo(a)anthracene, benzo(ghi)perylene, 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 3 orders of magnitude. The burning of 1 kg of wood (oak) produced PAHs with a total toxicity equivalent of 4.2 µg of BaP (SD 5.5 µg). The combustion of the same amount of wood-based waste led to the emission of PAHs equivalent to about 30 times more BaP. The combustion of PE, RAG, TR, PET and PU resulted in the emission of PAHs 280–800 times 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, and 2.2 mg kg$^{-1}$, SD 1.4 mg kg$^{-1}$, respectively) were similar to

Table 1. The absolute emission factors of PM$_{10}$ (mg g$^{-1}$), total PAHs (mg PAHs kg$^{-1}$ of fuel) and total PAHs expressed in BaP toxicity equivalent (mg kg$^{-1}$ of 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 | 21.9  | 19.1 | 1.63  | 2.0  | 390                |
| PET         | 11    | 1.6  | 32.14 | 14   | 2.2   | 1.4  | 520                |
| PE          | 18    | 7.2  | 31.23 | 23   | 1.2   | 1.1  | 280                |
| PU          | 22    | 8.4  | 63.27 | 27   | 3.4   | 0.12 | 800                |
| PP          | 33    | 18   | 98.83 | 8.3  | 16.0  | 4.7  | 3700               |
| TR          | 35    | 6.0  | 25.91 | 9     | 2.0   | 1.8  | 480                |
| PVC         | 35    | 10   | 52.51 | 5.1  | 5.3   | 1.9  | 1300               |
| PS          | 53    | 15   | 135.60| 60   | 11.0  | 4.8  | 2500               |
| ABS         | 82    | 27   | 257.103| 103   | 19.0  | 12  | 4500               |
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 and 1.8 ± 0.3 mg kg\(^{-1}\), respectively. Toxic PAHs in the highest quantity 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 Fig. 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 those produced in wood burning. The combustion of plastic wastes results in the formation of the significantly more carcinogenic four- to six-ring compounds.

It should be added, however, that the burning conditions affect the profile and total quantity of PAHs considerably. The low quantity of five-ring and six-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 the combustion of wood, paper, LDF, OSB, rag, PET, PE, PU, PP, tyre material, PVC, PS and ABS, which are all abundant waste types frequently burned in households and open fires in 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 5 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 a nearly 10-fold increase in 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 a 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 of the higher total PAH content of the 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 to 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\(_{10}\) and PM\(_{2.5}\). But more importantly, the health risks associated with air pollution would be reduced disproportionately more than would normally follow from such reductions in PM\(_{10}\) and 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 (hoffer@almos.uni-pamonn.hu).

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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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References

Anenberg, S. C., Balakrishnan, K., Jetter, J., Masera, O., Mehta, S., Moss, J., and Ramanathan, V.: Cleaner cooking solutions to achieve health, climate, and economic co-benefits, Environ. Sci. Technol., 47, 3944–3952, https://doi.org/10.1021/acs.est.3b04923, 2013.

Bai, Y., Brugha, R. E., Jacobs, L., Grigg, J., Nawrot, T. S., and Nemery, B.: Carbon loading in airway macrophages as a biomarker for individual exposure to particulate matter air pollution – A critical review, Environ. Int., 74, 32–41, https://doi.org/10.1016/j.envint.2014.09.010, 2015.

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

Christian, T. J., Yokelson, R. J., Cárdenas, B., Molina, L. T., Engling, G., and Hsu, S.-C.: Trace gas and particle emissions from domestic and industrial biofuel use and garbage burning in central Mexico, Atmos. Chem. Phys., 10, 565–584, https://doi.org/10.5194/acp-10-565-2010, 2010.

EPA: Evaluating the Toxicity and Assessing the Carcinogenic Risk of Environmental, Mixtures Using Toxicity Equivalency Factor, Supporting material for Cleanup Levels and Risk Calculation (CLARC), Washington State Department of Ecology – Toxics Cleanup Program, 2007.

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

Gelencsér, A., May, B., Simpson, D., Sanchez-Ochoa, A., Kasper-Giebl, A., Puxbaum, H., Pio, C. A., and Legrand, M.: Source apportionment of PM2.5 organic aerosol over Europe: Primary/secondary, natural/anthropogenic, and fossil/biogenic origin, J. Geophys. Res.-Atmos., 112, D23S04, https://doi.org/10.1029/2006JD008094, 2007.

Gonçalves, C., Alves, C., and Pio, C.: Inventory of fine particulate organic compound emissions from residential wood combustion in Portugal, Atmos. Environ., 50, 297–306, https://doi.org/10.1016/j.atmosenv.2011.12.013, 2012.

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, https://doi.org/10.1016/j.chemosphere.2010.04.078, 2010.

Gullette, 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, Environ. Sci. Technol., 44, 394–399, https://doi.org/10.1021/es902676w, 2010.

Jayaratne, T., Stockwell, C. E., Bhave, P. V., Praveen, P. S., Rathnayake, C. M., Islam, M. R., Panday, A. K., Adhikari, S., Maharjan, R., Goetz, J. D., DeCarlo, P. F., Saikawa, E., Yokelson, R. J., and Stone, E. A.: Nepal Ambient Monitoring and Source Testing Experiment (NAMaSTE): emissions of particulate matter from wood- and dung-fueled cooking fires, garbage and crop residue burning, brick kilns, and other sources, Atmos. Chem. Phys., 18, 2259–2286, https://doi.org/10.5194/acp-18-2259-2018, 2018.

Kantar Hoffmann Company: Lakossági hulladékégetési szokások Magyarországon, https://www.legevo.hu/sites/default/files/Kantar_Hoffmann_Levego_MCS_Hulladekegetes_2017dec.pdf, last access: 2 July 2020.

Karagulian, F., Belis, C. A., Francisco, C., Dora, C., Prüss-Ustün, A. M., Bonjour, S., Rohani, H. A., and Amann, M.: Contributions to cities’ ambient particulate matter (PM): A systematic review of local source contributions at global level, Atmos. Environ., 120, 475–483, https://doi.org/10.1016/j.atmosenv.2015.08.087, 2015.

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, Environ. Int., 60, 71–80, https://doi.org/10.1016/j.envint.2013.07.019, 2013.

Kistler, M., Schmidl, C., Padouvas, E., Giebl, H., Lohninger, J., Ellinger, R., Bauer, H., and Puxbaum, H.: Odor, gaseous and PM10 emissions from small scale combustion of wood types indigenous to Central Europe, Atmos. Environ., 51, 86–93, https://doi.org/10.1016/j.atmosenv.2012.01.044, 2012.

Kumar, S., Aggarwal, S. G., Gupta, P. K., and Kamwura, K.: Investigation of the tracers for plastic-enriched waste burning aerosols, Atmos. Environ., 108, 49–58, https://doi.org/10.1016/j.atmosenv.2015.02.066, 2015.

Lemieux, P. M., Lutes, C. C., and SANTOIANI, D. A.: Emissions of organic air toxics from open burning: a comprehensive review, Prog. Energ. Combust., 30, 1–32, https://doi.org/10.1016/j.proengcomb.2003.08.001, 2004.

Lemieux P. M.: Evaluation of emissions from the open burning of household waste in barrels, EPA-600/R-97-134a (vol. 1), NTIS PB98-127343 and EPA-600/R-97-134b (vol. 2), PB98-127350, 1997.

Maassikmens, M., Kupri, H. L., Teinemaa, E., Vainumae, K., Arumae, T., Roots, O., and Kimmel, V.: Emissions from burning municipal solid waste and wood in domestic heaters, Atmos. Pol-
Nisbet, I. C. and LaGoy, P. K.: Toxic Equivalency Factors (TEFs) for Polycyclic Aromatic Hydrocarbons (PAHs), Regul. Toxicol. Pharm., 16(3), 290–300, https://doi.org/10.1016/0273-2309(92)90009-X, 1992.

Park, Y. K., Kim, W., and Jo, Y. M.: Release of harmful air pollutants from open burning of domestic municipal solid wastes in a metropolitan area of Korea, Aerosol Air Qual. Res., 13, 1365–1372, https://doi.org/10.4209/aaq.2012.10.0272, 2013.

Pastorello, C., Caserini, S., Galante, S., Dilara, P., and Galletti, F.: Importance of activity data for improving the residential wood combustion emission inventory at regional level, Atmos. Environ., 45, 2869–2876, https://doi.org/10.1016/j.atmosenv.2011.02.070, 2011.

Piazzalunga, A., Belis, C., Bernardoni, V., Cazzuli, O., Fermo, P., Valli, G., and Vecchi, R.: Estimates of wood burning contribution to PM by the macro-tracer method using tailored emission factors, Atmos. Environ., 45, 6642–6649, 2011.

Pipal, A. S. and Gursumeeran Satsangi, P.: Study of carbonaceous species, morphology and sources of fine (PM_{2.5}) and coarse (PM_{10}) particles along with their climatic nature in India, Atmos. Res., 154, 103–115, https://doi.org/10.1016/j.atmosres.2014.11.007, 2015.

Puxbaum, H., Caseiro, A., Sánchez-Ochoa, A., Kasper-Giebl, A., Claeyts, M., Gencencér, A., Legrand, M., Preunkert, S., and Pio, C.: Levoglucosan levels at background sites in Europe for assessing the impact of biomass combustion on the European aerosol background, J. Geophys. Res., 112, D008114, https://doi.org/10.1029/2006JD008114, 2007.

Querol, X., Karanasiou, A., Amato, F., Vasconcelos, C., Alastuey, A., Viana, M., Moreno, T., Planas, F., Perez, N., Cabanas, M., Bartoli, R., Martinez, S., Sosa, M., Montfort, E., Celades, I., Escrig, A., Sanfelix, V., Gomar, S., Harrison, R., Holman, C., Beddows, D., Harding, M., Eleftheriadis, K., Diapoulis, L., Vratolis, S., Gini, M., Bairaktari, E., Dalaina, S., Galifianakis, V., Lucarelli, F., Nava, S., Calzolai, G., Udisti, R., Becagli, S., Traversi, R., Severi, M., Borselli, S., Giannoni, M., Alves, C., Pio, C., Nunes, T., Tarelho, L., Duarte, M., Cerqueira, M., Vicente, E., Custódio, D., Pinto, H., Gianelle, V. L., and Colombi, C.: Airuse Life 11 ENV/ES/584, Testing and development of air quality mitigation measures in Southern Europe, Emission factors for biomass burning, Report 9, 2016.

Rafael, S., Tarelho, L., Monteiro, A., Sá, E., Miranda, A. I., Borrego, C., and Lopes, M.: Impact of forest biomass residues to the energy supply chain on regional air quality, Sci. Total Environ., 505, 640–648, https://doi.org/10.1016/j.scitotenv.2014.10.049, 2015.

Safe, S. H.: Hazard and risk assessment of chemical mixtures using the toxic equivalency factor approach, Environ. Health Persp., 106, 1051–1058, https://doi.org/10.2307/3434151, 1998.

Schmidl, C., Luiisser, M., Padovas, E., Lasselsberger, L., Rzaca, M., Ramirez-Santa Cruz, C., Handler, M., Peng, G., Bauer, H., and Puxbaum, H.: Particulate and gaseous emissions from manually and automatically fired small scale combustion systems, Atmos. Environ., 4, 7443–7454, https://doi.org/10.1016/j.atmosenv.2011.05.006, 2011.

Simoneit, B.: Biomass burning – A review of organic tracers for smoke from incomplete combustion, Appl. Geochem., 17, 129–162, https://doi.org/10.1016/S0883-2927(01)00061-0, 2002.

Simoneit, B. R. T., Medeiros, P. M., and Didyk, B. M.: Combustion products of plastics as indicators for refuse burning in the atmosphere, Environ. Sci. Technol., 39, 6961–6970, https://doi.org/10.1021/es050767x, 2005.

Századvég Foundation: Survey on domestic heating and opinion on air pollution of the Hungarian population conducted by the Századvég Foundation on behalf of the Ministry of Agriculture, 2018.

Tissari, J., Lyyrinen, J., Hytönen, K., Sippula, O., Tapper, U., Frey, A., Saarnio, K., Pennanen, A. S., Hillamo, R., Salonen, R. O., Hirvonen, M.-R., and Jokiniemi, J.: Fine particle and gaseous emissions from normal and smouldering wood combustion in a conventional masonry heater, Atmos. Environ., 42, 7862–7873, https://doi.org/10.1016/j.atmosenv.2008.07.019, 2008.

Tiwari, S., Pandithurai, G., Attrl, S. D., Srivastava, A. K., Soni, V. K., Bisht, D. S., Anil Kumar, V., and Srivastava, M. K.: Aerosol optical properties and their relationship with meteorological parameters during wintertime in Delhi, India, Atmos. Res., 153, 465–479, https://doi.org/10.1016/j.atmosres.2014.10.003, 2015.

Tomsje, T., Horak, J., Tomsjeova, S., Krpec, K., Klanova, J., Dej, M., and Hopan, F.: The impact of co-combustion of polyethylene plastics and wood in a small residential boiler on emissions of gaseous pollutants, particulate matter, PAHs and 1,3,5-triphenylbenzene, Chemosphere, 196, 18–24, https://doi.org/10.1016/j.chemosphere.2017.12.127, 2018.

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, Environ. Sci. Technol., 48, 9523–9530, https://doi.org/10.1021/es502250z, 2014.