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Evaluating emissions and air quality implications of residential waste incineration

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In Europe mainly at winter season the PM levels exceed air quality limits, which correlated with the operation of solid-fired boilers. More and more people are returning to using these devices due to energy shortage caused by the pandemic and regional conflicts. In addition, the phenomena of co-burning fuels and municipal waste in residential boilers in primarily fuel poverty households increases further the amount of pollutants in the atmosphere. This study aims to correlate the quantity and quality of air pollutants with the type of fuel (wood and wastes) burned. Combustion experiments were conducted using oak fuel mixed with three waste groups: (1) plastics (PP, HDPE, PET); (2) textiles (polyester—PES, cotton—COT); and (3) papers (cardboard—CARD, glossy coated paper—GCP, 84C/PAP). The addition of waste to wood fuel altered the morphology of emitted particles. While waste burning doesn't always increase particle quantity, it significantly raises PAH concentrations. A strong relationship exists between waste type, particle morphology, and PAH quality, where with lower molecular weight PAHs linked to tar agglomerates and higher ones to soot agglomerates with inorganic crystals.

Keywords Combustion, Morphology, PM, PAH, Residential, Waste

Abbreviations

BC	Black carbon
CARD	Cardboard
COT	Cotton
EC	Elemental carbon
GCP	Glossy coated paper
HDPE	High-density polyethylene
OC	Organic carbon
PAH	Polycyclic aromatic hydrocarbon
PET	Polyethylene-terephthalate
PES	Polyester
PM	Particulate matter
PP	Polypropylene
VOC	Volatile organic compound

At certain times in Europe (typically in winter), atmospheric levels of particulate matter (PM) exceed air quality limit values, posing a serious environmental and health risk[1]. The main contributor to health damage from air pollution is particulate matter in the air[2]. The smaller the diameter of the particulate matter, the easier it is to enter the human body, where it can have negative effects[2,3].

Although biomass is frequently cited as a clean energy source, urban air quality data indicates that the combustion of wood for residential heating and energy generation[4–6] is a significant contributor to carbon air pollutant emissions in the EU, creating new challenges for air quality, human health, and global and regional climate[7].

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Due to inflation and increasing fuel prices caused by the energy crisis, more households are turning to solid fuel burning devices. Multiple studies have reported that wood burning for residential heating during the winter season is a significant contributor to particulate matter emissions[8–10]. In Europe, emissions from biomass burning in residential boilers during the winter season can account for up to 70% of total organic particulate matter emissions[11]. According to data from the European Environment Agency, households are responsible for over 68% of PM2.5 emissions in Poland and the Czech Republic, and over 80% in Slovakia, Croatia, Romania, and Hungary[12]. Therefore, households are considered the primary source of emitted PM and PAH[13]. Additionally, an increase in PAH levels in the atmosphere is associated with an increase in PM2.5 emissions[14]. Despite these findings, the Hungarian government launched a wood-burning program due to skyrocketing gas prices, which has further encouraged the use of the solid-fired boilers.

Moreover, the practice of co-burning fuels and municipal waste in residential boilers, primarily in households experiencing fuel poverty, remains a persistent problem worldwide, despite being prohibited. This issue is prevalent in several European countries, including the Czech Republic, Poland, Lithuania, Finland, Estonia, Hungary, and Romania[15–21]. In Poland, at least 10% of the population burns various plastics and waste in their boilers, while in Hungary 2–10% of households do the same[17,22]. In the Czech Republic almost 20% of households (approximately 620,000) use solid fuels for heating, with 534,000 households still relying on outdated manually operated boilers. In suburban and rural areas with fuel-poverty households, the amount of waste collected from households decreases by 15–25% during the winter season, and in the landfilled waste increases significantly the percentage of ash and dust[20]. This further supports the fact that waste incineration occurs in these households.

Exposure to combustion products (PM, CH_4, CO, PAH, VOC) emitted from the burning of solid fuels contributes to chronic illnesses and acute health effects[23], as well as indirectly causing death in the form of stroke, heart disease, lung cancer, and chronic respiratory diseases[24]. Depending on their source, solid particles contain organic (VOC, PAH, BC) and inorganic components[25]. Yadav et al.[26] stated that during biomass burning, organic carbon (OC) and black carbon (BC) make up 50–70% of all PM emissions. Inorganic elements such as potassium, chlorine, and calcium account for about 10% of total PM emissions. In terms of BC and OC from biomass burning, Jayarathne et al.[27] obtained lower values compared to total PM2.5 emissions. However, burning plastic and foil packaging separately resulted in an increase of OC and BC emissions up to 60% of total PM2.5 emissions. Wu et al.[28] suggest that carbon-containing materials, especially PAHs, dominate PM2.5 emissions from residential burning, while metal emissions are mainly associated with coal-fired power plants. Therefore, PM2.5 from residential burning is more toxic due to the associated PAHs. As the main contributors to negative health effects among the PM emission fractions are the PAHs and metals[29,30]. Moreover, several studies have shown that 95% of emitted solid PAH containing particles have a diameter smaller than 3 μm[31–34].

Therefore, improving air quality can primarily be achieved through reducing residential waste burning and optimizing the operation of boilers. That is why this research focuses on the practice of residential solid fuel combustion, including the illegal burning of waste, and the mechanisms behind the generation of air pollutants that accompany it. The goal is to uncover the relationship between the type of fuel and the gas and solid air pollutants emitted during combustion.

Materials and methods
Materials
During the experiments, the combusted wastes were classified into three groups: (1) Plastics: PP, HDPE, PET; (2) Textiles: Polyester (PES), Cotton (COT); (3) Papers: Cardboard (CARD), Glossy Coated Paper (GCP), 84C/PAP.

The selected wastes were fed into the boiler during the combustion experiments at a weight ratio of 1 waste/9 oak.

Methods
Raw material analysis
For the raw material analysis, the following procedures were used:

(1) Mettler Toledo HB43-S Halogen Moisture Analyzer is used for the determination of moisture content in a sample. The instrument uses the principle of loss on drying, where a sample is heated at a constant temperature while the weight loss due to the evaporation of moisture is recorded.
(2) Ash content determination is carried out by heating a sample at 850 ± 15 °C until a constant mass is achieved, following the MSZ EN ISO 18122:2016 standard.
(3) The analysis is focused on the determination of C-, H-, N-, S- components using a Carlo Erba EA1108 elemental analyzer, following the MSZ EN ISO 21644:2021 standard.
(4) The calorific value of a sample is determined using a Parr 6200 isoperibol bomb calorimeter, following the MSZ EN ISO 18125:2017 standard.

Residential-scale combustion experiments
Several examples of similar studies can be found in the literature, for example, Tomsej et al.[20] added plastic waste at a rate of 7% w/w to beech firewood, while Edo et al.[35] burned heterogeneous municipal solid waste with firewood, where the waste quantity was 20% w/w during combustion. The combustion experiments were carried out in a TOTYA S18 residential boiler, which is easily available and widely used in Hungary. A fan was placed in the flue pipe, which drew clean air with a pressure of 28.5 Pa. This method made the combustion more stable.

Three measurement lines were included in the measurement circuit (Fig. 1):
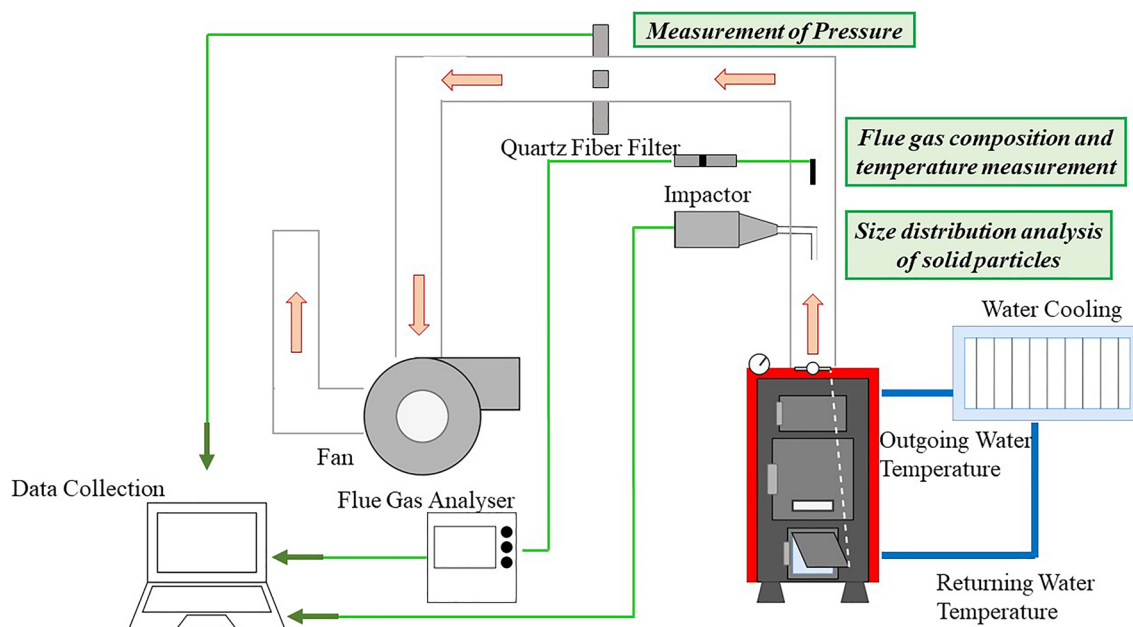
Fig. 1. Experimental setup employed during residential scale combustion experiments.

1) The first line is employed to measure the pressure in the flue pipe.
2) The second line is used to measure the composition and temperature of the flue gas, and to separate solid pollutants for PAH analysis.
3) The particle size distribution of solid particles was carried out by the third line.

The data from each measurement line was recorded and further analyzed.

The pressure of the flue pipe determination was carried out using a differential pressure probe for the TESTO 400 multifunctional base unit. The composition of the flue gas was determined using a Horiba PG 250 gas analyzer, with simultaneous measurement of the flue gas temperature during extraction. To examine the morphology of solid particles and to detect the solid-phase polycyclic aromatic compounds, a quartz fiber filter was placed in the extraction pipe. The particle size distribution of solid particles was determined using a Dekati PM10 impactor, which allowed for the classification of particles larger than 10 µm, between 2.5 and 10 µm, and less than 2.5 µm. The DEKATI PMP-500/450 pumping system assisted in the extraction, and the extraction rate was set using the MASS-VIEW sensor in the system.

The combustion experiments consisted of cycles, whose length was determined by the combustion time of the fuel mixture fed at once. Based on this, each combustion cycle lasted for 20–25 min. This method enabled the comparability of combustion experiments with different fuel mixtures.

Morphological analysis of solid particles
The morphology of solid particles trapped on a quartz fibre filter from combustion experiments were investigated using a Zeiss EVO MA10 scanning electron microscope equipped with an EDAX probe.

Detection of polycyclic aromatic compounds
A gas chromatograph coupled with a mass selective detector (GC-MSD) was utilized to determine the PAHs. First, 10 µL deuterated PAH surrogate standard mix containing 20 µg/mL acenaphtene-d10, phenantrene-d10, fluoranthene-d10, benzo(a)anthracene-d12, benzo(a)pyrene-d12 and dibenzo(ah)antracene-d14 was added to the samples prior extraction. Ultrasonic agitation was used to extract each filter with 10 mL n-pentane at room temperature for 30 min. The solutions were then concentrated to 1 mL using a gentle stream of nitrogen, and 10 µL of benzophenone internal standard (20 µg/mL) was added to the resulting solutions before the gas chromatography measurement.

The GC–MSD system consisted of an Agilent 7890A GC and Agilent 5975C MSD. For the separation Restek Rxi-5MS (30 m × 0.25 mm × 0.25 µm) capillary column was used. The chromatographic conditions were as follows: injector temperature 300 °C, detector temperature 230 °C, quad temperature 150 °C. Helium was the carrier gas with 0.3 m/s linear velocity. 1 µL sample was injected with the use of pulsed splitless injection mode.

The recoveries of the surrogate dPAH ranged widely varied from 44% (acenaphtene-d10) to 88% (dibenzo(ah) antracene-d14). The recoveries were taken into account for each sample during the quantitative evaluation. It has to be noted, that the method is not necessarily suitable for analysing light PAHs such as naphtalene, acenaphthylene, acenaphtene, and fluorene.

The calculation of flue gas emission factors and PAH-emission factors were carried out via several calculation steps and using multiple equations. The corresponding details can be found in the Supplementary material.
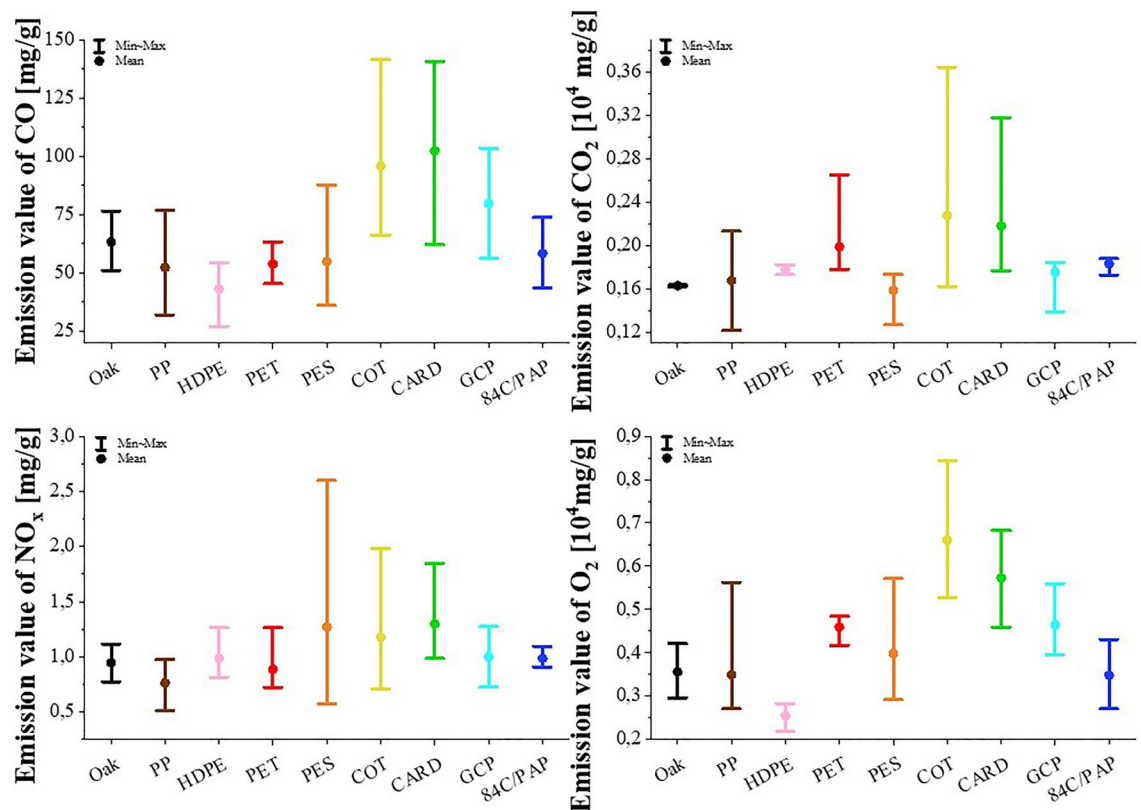
Fig. 2. Evolution of flue gas emission factors from the combustion of Oak and waste added in 10% w/w to the Oak. The dot represents the average, and the extreme values represent the minimum and maximum values.

Results and discussion
Evolution of emission factors from boiler combustion experiments

Flue gas emission factors were evaluated from the combustion of oak and different waste added in 10% w/w to the oak (Fig. 2). The co-combustion of PP and PE increases CO_2 emissions, while the emissions of certain oxygen-containing compounds[36], in this case, O_2 and CO, decrease. Furthermore, it was found that materials (COT, CARD, GCP, 84C/PAP) with higher ash content than oak had higher CO emissions. The CO and O_2 concentrations show a similar trend of variation. For example, with the addition of cotton and cardboard, not only CO but also O_2 concentrations in the flue gas increased significantly. In practice, when the air factor is higher and the fuel–air mixing is better, the CO emissions will be lower[37]. This is counteracted by the introduction of too much air, which cools the firebox temperature significantly, thus favouring CO formation[38]. In poorer combustion conditions, the CO_2 concentration should also decrease, but the addition of cotton and cardboard gave the highest CO_2 concentrations.

This can be explained by the fact that materials with a high ash content, once burnt, spread across the woodpile, blocking the primary combustion air path. This leads to a deterioration in combustion quality, as indicated by increased CO. As the primary combustion air path is blocked, the flue gas fan draws in combustion air through the secondary vent, reducing the firebox temperature and increasing both the combustion air and the flue gas flow rate (Table S1). The CO_2 value detected by the gas analyzer must also take into account the CO_2 content of the air, which increases with the amount of air introduced.

The amount of CO_2 generated is essentially determined by the temperature and design of the firebox and the C content of the fuel. Since plastics have a higher C content and at the same time less CO_2 is produced during wood co-firing, it can be assumed that this parameter is not the determining factor in this investigation.

Emissions of solid-phase PAHs and PM during combustion

The co-incineration of wastes affected the amount of emitted PMs, although the percentage distribution of PMs by mass did not change for different sizes (Fig. 3). There is a close relationship between the emission factors of particulate matter, CO, and O_2, as their trends are similar in change with waste materials. Indicators of incomplete combustion include not only CO but also PMs and PAHs[39]. However, the relationship between them is not clarified. Although some studies have found a strong correlation between PM and CO[40], Carter et al.[41] suggest that this close relationship cannot be described by a ratio or other measures.

Furthermore, it is clearly demonstrated that co-burning increased the amount of PAHs (polycyclic aromatic hydrocarbons) since burning waste generates more solid-phase PAHs than burning only wood (Fig. 3). In some cases, such as in the textile group, the volume of waste fed into the boiler was too high, blocking the flow of combustion air through the grate, which choked the combustion. In the case of the paper group, a large amount
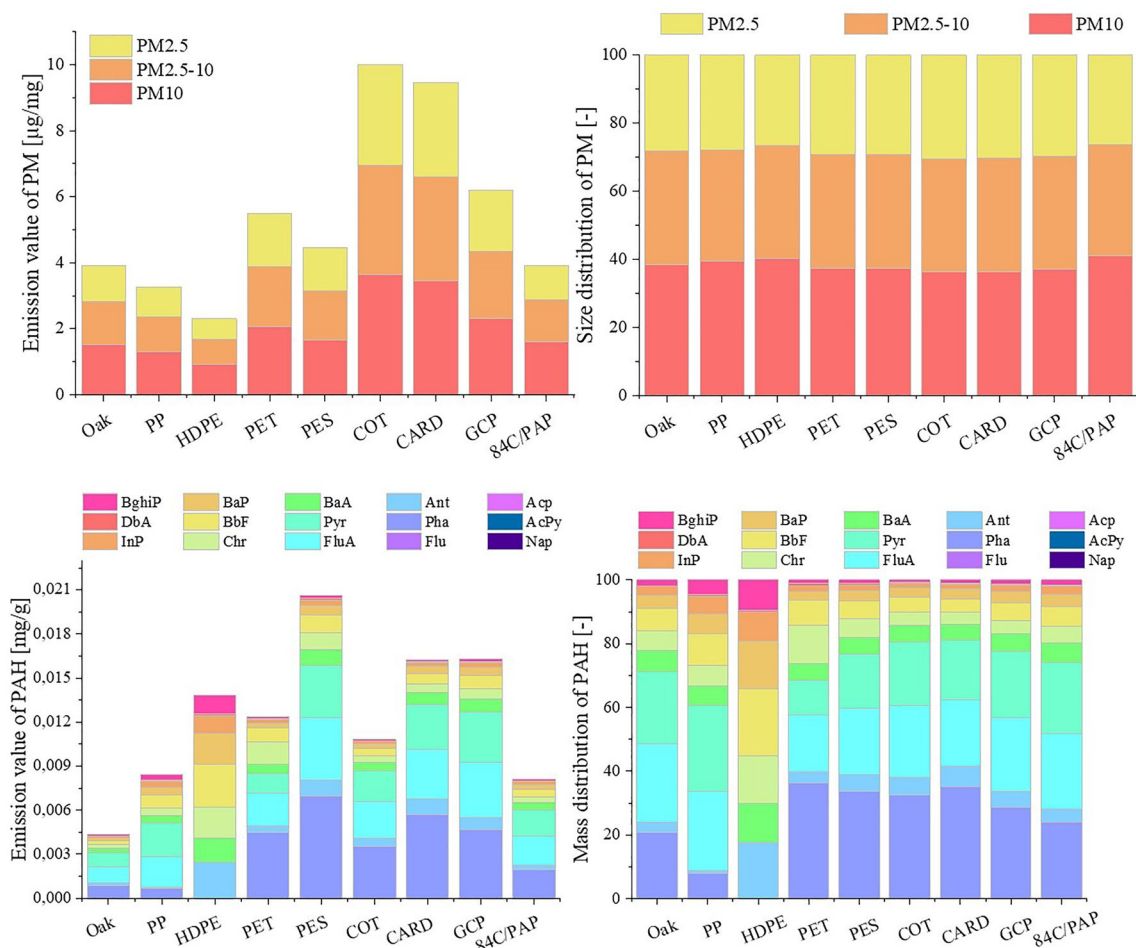
Fig. 3. Particulate matter emissions per mass of fuel and mass percentage distribution of PM of different sizes in boiler experiments (up); emission value and percentage distribution of PAHs on a quartz fiber filter (down).

of residual ash left on the unburned wood caused the aforementioned fire suppression. These unfavourable conditions further increased the amount of PAHs produced.

In the case of PP and HDPE, although the amount added to wood does not appear significant (10% w/w), it still significantly affects the composition of the resulting PAHs. This is because the PAHs from plastics can be up to 4 orders of magnitude greater than the amount of PAHs from natural materials such as cardboard, glossy coated paper[42], and presumably from wood. Additionally, the dominant compounds are those with 4–5 rings, which shifts the mechanism towards the formation of higher molecular weight compounds during co-burning with PP and HDPE.

Morphology of solid particles from combustion tests in boiler

Particulate matter emitted from residential combustion can take different morphologies, which consist of organic and/or inorganic. Atiku et al.[43] investigated how the properties of fuels (different elemental composition, and physical properties) affect the composition of the resulting soot. During the flaming phase of combustion, black carbon emissions dominated. The black carbon consisted of spherical particles that were compressed into agglomerate chains and had a high C content. Inorganic compounds such as Na and K in the fuel contributed strongly to the formation of soot aggregates[44]. Scanning electron microscopy of wood-burning smoke particles from wood combustion by Hunter et al.[45] observed soot agglomerates with similar chain structures. In this study, soot agglomerates with high C content are only detected during the co-combustion of PP and HDPE. These soot agglomerates were linked with large inorganic minerals (Fig. 4, Fig. S1).

Inorganic crystals are formed from inorganic compounds absorbed as nutrients by the plant and defined as ash. The inorganic content of wood is strongly influenced by its type, age and environment[46,47]. According to Torvela et al.[48], the amount of component of inorganic ash emitted with solid particles from wood combustion increases with increasing burning temperature. This observation is strongly related to the fact that the highest flue gas temperatures were achieved by co-combustion of PP and HDPE waste, which can be attributed to their high C content (Supplementary Data). Torvela et al.[48] further observed that primary ash particles (also known as fly ash agglomerate) were increased by sintering, deposition on larger particles, or condensation of gaseous hydrocarbons.

a) 90% m/m Oak and 10% m/m PP – 2 500x   b) 90% m/m Oak and 10% m/m HDPE – 1 500x
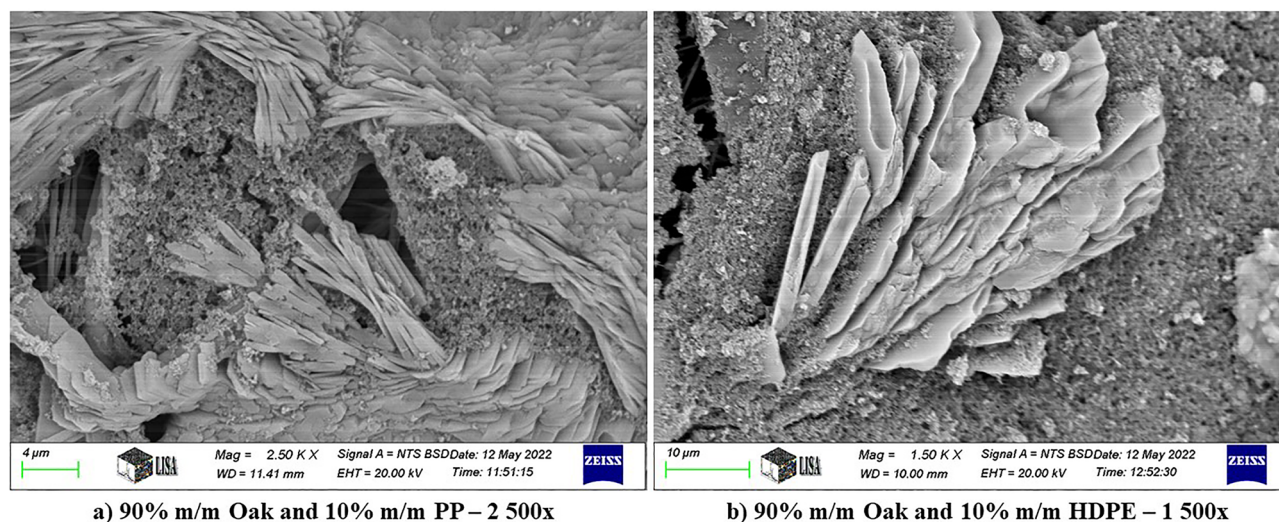
Fig. 4. Soot agglomerates attached to inorganic crystalline particles.

According to Bhandari[49] and Luo et al.[50], soot aggregates can mix with other chemical components, such as inorganic and organic materials, which can lead to more complex morphologies. These morphologies can take one of two forms: (1) the soot particles may be completely or partially coated by condensing inorganic materials; (2) or the soot particles may be attached to the coagulated particles which is also shown in Fig. 4. Freshly released soot particles are generally hydrophobic in nature, but the organic and inorganic substances coating their surface make them hydrophilic[51,52]. The chain fractal-like morphology of soot particles provides active sites for attachment to the surface, such as adsorption and deposition of other chemical species[53,54].

Considering additional morphologies, irregular particles detached from the structure of burning material may also appear on the filter in individual cases (Fig. 5).

Lyubov et al.[55], in experiments with emitted particles from a residential boiler, observed morphologies with sharp structures that were considered primary structures. Franus et al.[56] discovered unburned carbonaceous matter in coal combustion particulate matter. According to Rabacal et al.[57], irregular agglomerates are typically composed of unburnt carbon and non-volatile compounds. Some volatile compounds indicate that the ash volatiles in the solid product residue from biomass pyrolysis agglomerate in the fuel bed during combustion and then form alkali chlorides and sulphates on the surface by heterogeneous condensation, which are subsequently emitted with the flue gas. Yao et al.[58] found that irregular fragments from coal combustion are mainly composed of unburnt coal, anhydrite, and calcite. At the relatively low temperatures (750–800 °C) available in a residential boiler, most of the minerals in coal do not melt but only soften.
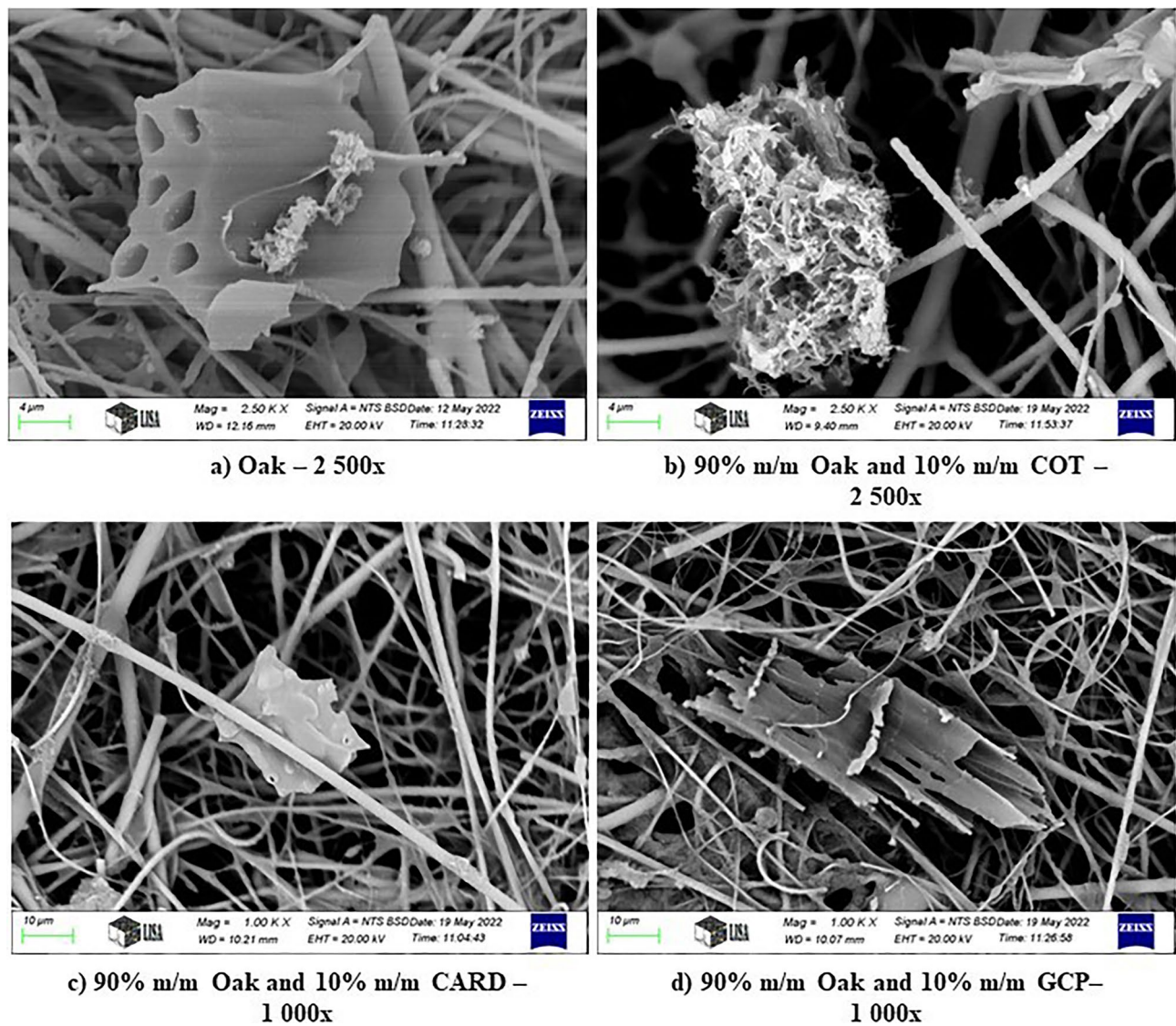
Large spherical morphologies detected among the solid particles (Fig. 6). The first column shows Ca–Si mineral spheres, while the second column illustrates high-carbon tar spheres. The tar spheres are likely to contain an inorganic coating, as detectable amounts of Si, Na, K, and Ca are present. During the combustion of waste fuels with the same composition, both tar and mineral spheres can be found on the filters simultaneously (cotton, 84C/PAP).

By adding waste materials (except for PP and HDPE), larger-sized spherical structures were detected on the filters. Due to their relatively large size, these morphologies can primarily be identified as tarball agglomerates (Fig. 7, Fig. S2). According to Girotto et al.[59], a particle can be considered an agglomerate if it consists of 8 or more spheres. Zangmeister et al.[60] suggest that particles containing 2–7 spheres are in an intermediate growth phase before forming agglomerates.

Although many people believe that tarballs cannot form block structures[61–63], Yuan et al.[64] and Corbin and Gysel-Beer[65] have provided examples to the contrary, where tarballs appeared as agglomerates with other materials.

Due to the fact that tarballs are mainly composed of carbon and oxygen, with only traces of inorganic components such as nitrogen, potassium, silicon, and sulphur[59,66], the tarballs are likely coated with an inorganic layer whose composition varies as waste fuels are burned. Except for glossy paper, where calcium strongly dominates, the main components of the coating are silicon and potassium, with small amounts of sodium and sulphur also present. As explained by Leskinen et al.[67], particles generated from the efficient combustion of biomass are predominantly composed of different alkali salts that correspond to the ash properties of the fuel. As a result, the emitted solid particles are compacted into a highly sintered regular chain-like structure. In their analysis of solid particles generated from burning cardboard, Liu et al.[68] found that 91% of the primary particles were irregularly shaped organic-containing particles containing carbon and oxygen, as well as small amounts of silicon. The degree of sintering depends on the ratio of sulphate, chloride, and carbonate compounds in the alkali salt mixture.

These tarball agglomerates may likely contain significant amounts of PAHs, as the lowest PAH emissions were obtained during wood burning (where this morphology type was not detectable).

a) Oak – 2 500x

b) 90% m/m Oak and 10% m/m COT – 2 500x

c) 90% m/m Oak and 10% m/m CARD – 1 000x

d) 90% m/m Oak and 10% m/m GCP– 1 000x

Fig. 5. Irregular grain structures detached from the fuel structure.

Conclusion

The study identified several morphologies of solid particles emitted during combustion and deposited on filters, including

(1) soot agglomerates associated with inorganic crystal particles,
(2) unique spherical particles that may be organic or high-carbon content,
(3) irregular particle shapes detached from the fuel structure, and
(4) tarball conglomerates.

The addition of municipal waste to the wood fuel at 10% w/w was found to clearly alter the morphology of emitted solid particles increasing PAH emissions. A close relationship was observed between the appearance of tarball conglomerates during waste-wood combustion and the increased concentration of polycyclic aromatic hydrocarbons (PAH) observed on the filter. These detected morphologies are closely related not only to polycyclic aromatic compounds but also to the fact of waste combustion. Based on the results, recommendations can be developed for fuel-poverty households to mitigate the effect of waste incineration.
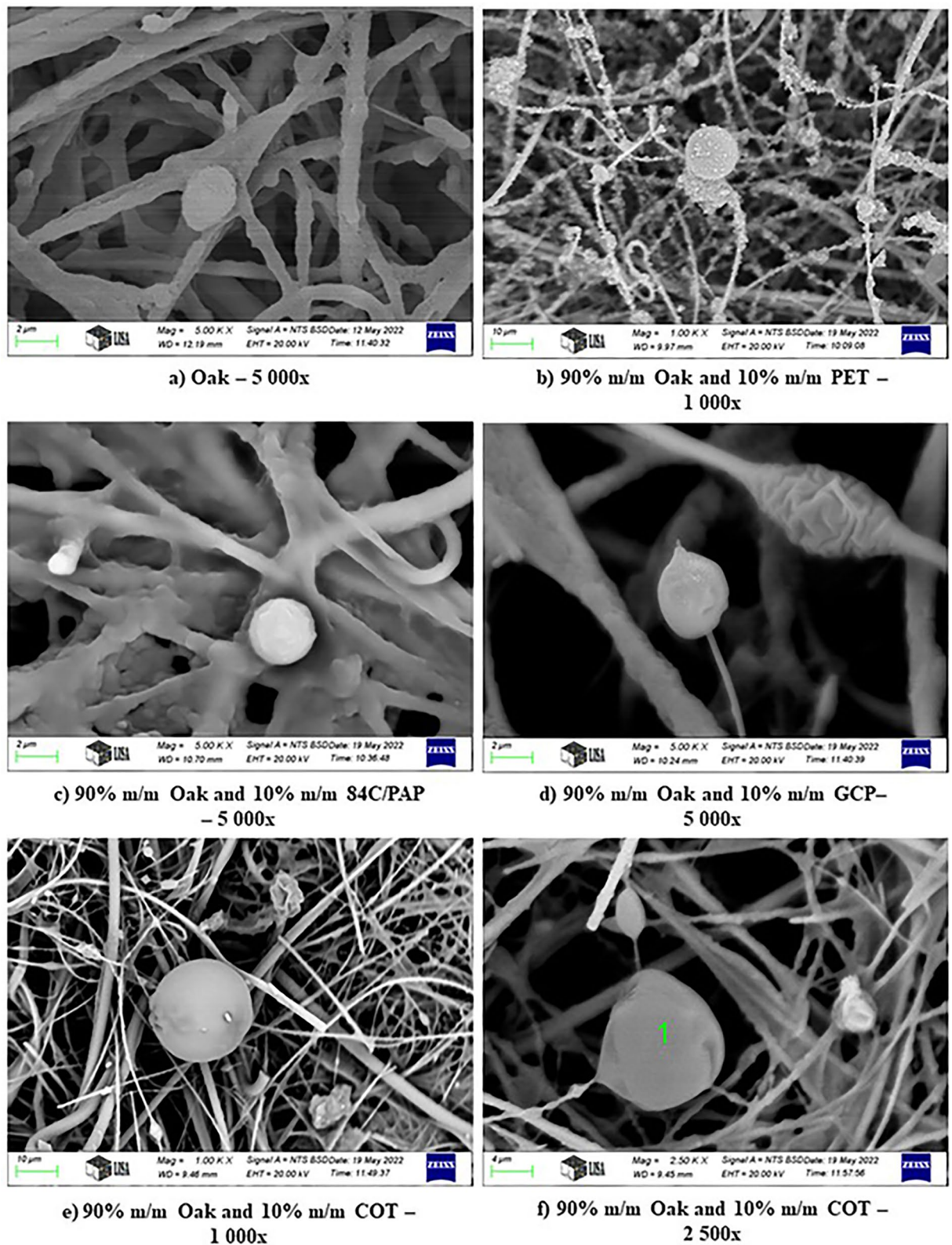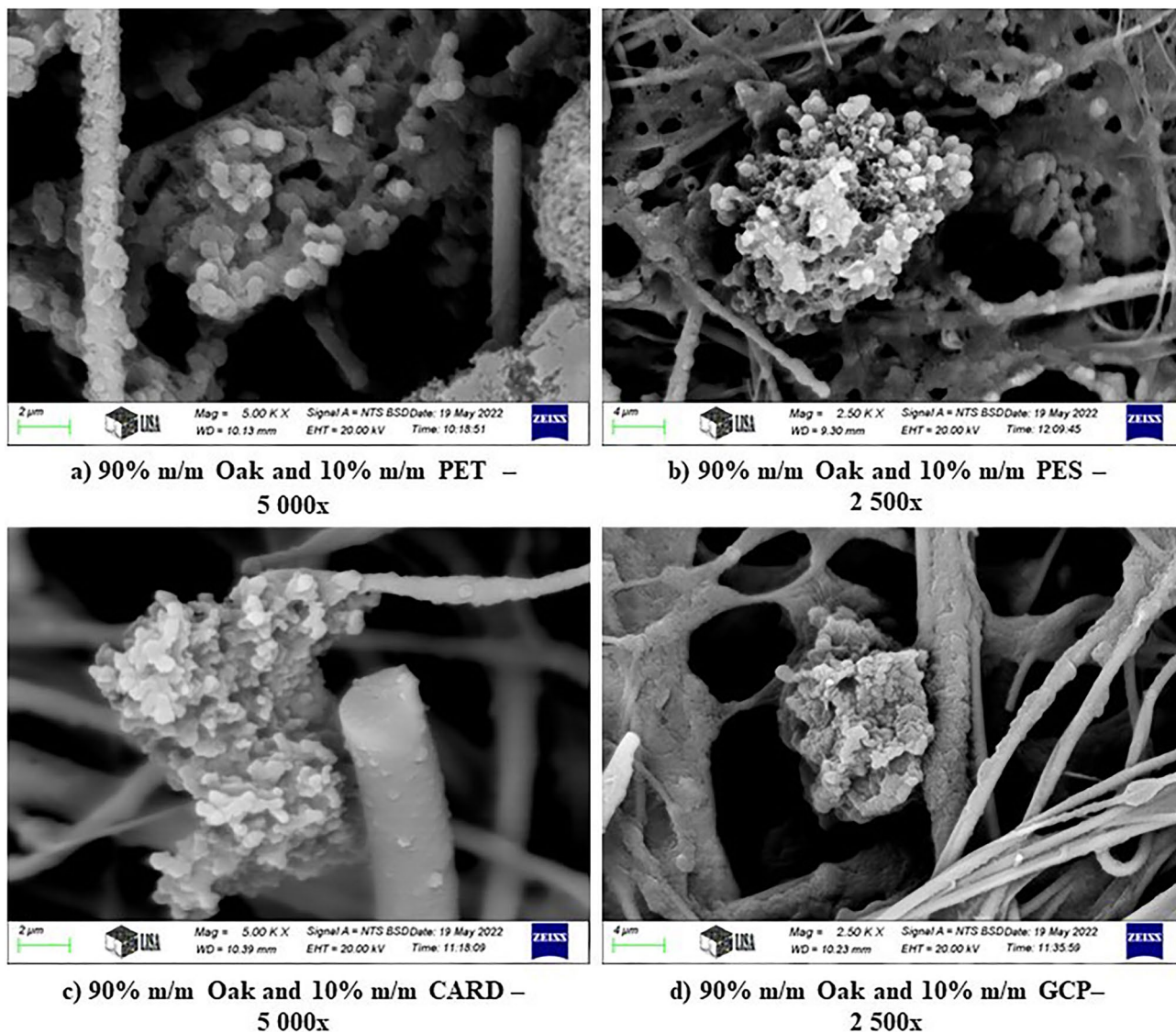
Fig. 6. Large spherical morphologies (**a,c,e**) mineral spheres; (**b,d,f**) tar spheres.

Fig. 7. Tar agglomerates with inorganic coating.

Data availability
Data is provided within the manuscript and supplementary information files.

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Author contributions

D.M. and C.P. conceived the scientific idea. D.M. carried out the experiments, analysed data and wrote the manuscript. B.F., B.V. carried out analysis and contributed to the manuscript text. A.J., L.F., and G.M. were carried out characterisation of the samples. All authors reviewed the manuscript.

Competing interests

The authors declare no competing interests.

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