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Effect of residential solid waste burning on ambient air quality in Central and Eastern Europe

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EFFECT OF RESIDENTIAL SOLID WASTE BURNING ON AMBIENT AIR QUALITY IN CENTRAL AND EASTERN EUROPE

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Abstract

The practice of burning waste in households, despite its illegal nature, is a non-negligible source of PM_{10} emissions. This study focuses on understanding the contribution of waste burning to PM_{10} concentrations by the tracer component approach.

Pyrolysis compounds, that are formed during the burning of different waste types such as 2-(benzoyloxy)ethyl vinyl terephthalate (2BEVT), styrene trimer (SSS) and melamine and additional compounds emitted during the burning of furniture panels were identified in samples collected during laboratory experiments and were suggested as indicators for the burning of specific types of waste. The relative, as well as the absolute emission factors of the tracer compounds for waste burning, were determined. The research reveals that certain plastic materials, particularly those containing aromatic rings in the polymeric structure, during burning emit higher levels of 1,3,5-TPB. Structural isomers of 1,3,5-TPB (124-TPB and quaterphenyls) were identified, and their ratios can be used as potential indicators of waste burning. Waste burning tracers were then analyzed in atmospheric PM_{10} samples collected in Hungary and Romania during consecutive heating seasons as well as in summer.

Based on the emission factors of the pyrolysis products characteristic for the burning of different waste types, the contribution of waste burning emission to the ambient PM_{10} mass concentration in Hungary and Romania during the winter and summer seasons was estimated. It was found that this contribution is a few percent, but it poses significant health risks taking into account the high emission factors and the toxicity of Polycyclic Aromatic Hydrocarbons (PAHs) emitted from these types of sources. It was estimated based on firewood consumption statistics and the absolute emission factor of levoglucosan (a tracer for wood burning) and those of the specific waste burning tracers that approximately 8-13% of household waste is illegally burned in household stoves during the heating season in Hungary and around 9-15% in Romania.

This research also provides insights into the identification and stability of organic compounds that can serve as tracers for waste burning. By examining the stability of the tracer compounds, a novel compound formed as a result of SSS oxidation was identified.

In conclusion, the use of waste burning tracer compounds facilitates the identification of pollution sources, helping to develop strategies for emission reduction and mitigate the adverse impacts of waste burning on human health and the environment.

Абстракт

Практика сжигания отходов в домашних хозяйствах, несмотря на ее незаконный характер, является немаловажным источником выбросов (ТЧ$_{10}$) PM$_{10}$. Данное исследование сосредоточено на понимании вклада сжигания отходов в концентрацию PM$_{10}$ с помощью метода индикаторных компонентов. Пиролизные соединения, которые образуются при сжигании различных типов отходов, таких как 2-(бензоилокси)этилвинилтерефталат (2BEVT), тример стирола (SSS) и меламин, а также дополнительные соединения, выделяющиеся при сжигании мебельных панелей, были идентифицированы в образцах, собранных в ходе лабораторных экспериментов, и были предложены в качестве индикаторов горения из конкретных видов отходов. Были определены как относительные, так и абсолютные коэффициенты выбросов индикаторных соединений при сжигании отходов. Исследование показало, что некоторые пластмассовые материалы, особенно те, которые содержат ароматические кольца в полимерной структуре, при горении выделяют более высокие уровни 1,3,5-TPB. Были идентифицированы структурные изомеры 1,3,5-ТРБ (124-ТРБ и кватерфенилы), и их соотношения могут быть использованы в качестве потенциальных индикаторов сжигания отходов. Затем были проанализированы индикаторы сжигания отходов в пробах атмосферных ТЧ10, собранных в Венгрии и Румынии в течение последовательных отопительных сезонов, а также летом.

На основе коэффициентов выбросов продуктов пиролиза, характерных для сжигания различных типов отходов, был оценен вклад выбросов при сжигании отходов в массовую концентрацию PM$_{10}$ в окружающей среде в Венгрии и

Румынии в зимний и летний сезоны. Было установлено, что этот вклад составляет несколько процентов, но он представляет значительный риск для здоровья, принимая во внимание высокие коэффициенты выбросов и токсичность полициклических ароматических углеводородов (ПАУ), выделяемых из источников такого типа. На основе статистики потребления дров и абсолютного коэффициента выбросов левоглюкозана (индикатора сжигания древесины), а также показателей конкретных индикаторов сжигания отходов было подсчитано, что примерно 8-13% бытовых отходов незаконно сжигается в бытовых печах во время отопительного сезона в Венгрии и около 9-15% в Румынии.

Это исследование также дает представление об идентификации и стабильности органических соединений, которые могут служить индикаторами для сжигания отходов. Изучая стабильность индикаторных соединений, было идентифицировано новое соединение, образующееся в результате окисления SSS.

В заключение следует отметить, что использование индикаторных составов для сжигания отходов облегчает идентификацию источников загрязнения, помогая разрабатывать стратегии сокращения выбросов и смягчения неблагоприятного воздействия сжигания отходов на здоровье человека и окружающую среду.

1. Introduction

Air pollution is one of the world's most significant issues. It refers to the contamination of the environment by hazardous chemicals or biological materials. It is well known that fossil fuel combustion and biomass burning are the two most important sources of fine particulate matter ($PM_{2.5}$, with an aerodynamic diameter 2.5 µm or less) in the atmosphere (Simoneit et al., 2002). Billions of people use solid fuels (wood, coal or agricultural waste) as the main source of household energy worldwide (Anenberg et al., 2013). 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).

Air pollution caused by waste combustion is harmful to human health and may contribute to climate change (Manisalidis et al., 2020). According to a recent report by Eurostat in Europe, municipal waste was produced at a rate of 513 kg capita^{-1} in 2022, 19 kg or 4% per person less than in 2021 (532 kg) and 46 kg more than in 1995 (467 kg). The most considerable fraction of municipal waste that is generated globally with an increasing rate is plastic (1-36%) (Hoornweg and Bhada-Tata, 2012) and according to Kantar Hoffman surveys, an unknown fraction of this waste is burned in household stoves. Since it is illegal to burn waste in households it is quite difficult to assess. According to sporadic media reports and available surveys conducted by non-governmental organizations, this practice may be widespread in impoverished regions of many European countries, particularly in Central and Eastern Europe. Emission studies for burning municipal waste in households are extremely scarce; the only report on PM_{10} and PAHs emission factors (EFs) for indoor combustion of 12 common types of municipal solid waste has been just recently published (Hoffer et al., 2020). According to this study, waste burning emits up to 40 times more PM_{10} (particulate matter with an aerodynamic diameter 10µm or less) and 800 times more PAHs than dry firewood combustion. These findings highlight the critical need for coordinated action to counter illegal waste combustion and the extreme health risks it poses. Because neither PM_{10} nor PAHs are specific for burning municipal waste in households, determining the potential contribution of these illegal activities to air pollution in various regions would necessitate the detection of tracers that are highly specific for the burning of various types of municipal waste (Hoffer et al., 2021).

2. Literature review

2.1 Carbonaceous aerosol particles

Understanding the complex processes that involve carbonaceous aerosols in the Earth's atmosphere is important, bearing profound implications for both climate dynamics and human health. This is emphasized by the increasing interest among scientists to study the characterization of these chemical species (Bond et al., 2013; Janssen et al., 2011). The carbonaceous fraction of the atmospheric particulate matter accounts for a considerable 20% to 50% of the overall aerosol mass (Kanakidou et al., 2005, Putaud et al., 2010). In the context of carbonaceous aerosols, like organic carbon (OC) along with the refractory light-absorbing component, commonly referred to as elemental carbon (EC when measured using thermal optical methods) or black carbon (BC when measured using optical methods). Furthermore, carbonate carbon might occasionally add to the overall carbon concentrations in aerosol samples (Giannoni et al., 2016). The combustion of solid fuels is a remarkable source of carbonaceous aerosol (soot and OC) (Streets et al., 2001; Zhang et al., 2016). The primary sources of BC lie mainly in anthropogenic combustion processes, including industrial emissions, transport and domestic heating (Querol et al., 2013, Sandrini et al., 2014). BC particles, known for their high ability to absorb light, play a key role in the positive radiation effects of aerosols. Nevertheless, there are significant uncertainties in our understanding of the spatial and temporal distribution of these sources, the effective mixing states of BC particles (internally mixed or core-shell), their size distribution and their nucleation potential for ice formation (IPCC, 2013). Moreover, it is important to recognize that BC can act as a carrier of various chemical components, many of which exhibit varying degrees of toxicity to sensitive human targets such as the respiratory system, protective cells, and even systemic circulation (Janssen et al., 2011). Therefore, black carbon plays an important role in addressing the impacts on health and climate caused by atmospheric aerosols.

On the other side, organic carbon (OC) takes on the role of a meaningful contributor to particulate matter mass concentrations. For example, Harrison et al. (2004) found that in London and Birmingham, organic compounds were 23% (equivalent to 18% organic carbon) and 21% (equivalent to 15% organic carbon) of the total PM_{10} at the kerbside and urban background sites, respectively. Its sources come from anthropogenic origins, usually linked to combustion processes and natural sources, such as sea spray and biogenic emissions. OC exhibits both primary and secondary origins, arising either as primary pollutants emitted directly or as compounds that form in the atmosphere through condensation processes. Subsequently, the OC/EC ratio in aerosol fractions exhibits vast variations, both spatially and seasonally, rendering it a valuable diagnostic ratio for investigating atmospheric sources and processes contributing to secondary organic compound formation. It's important to note that organic carbon's high solubility is a key factor in determining the oxidative potential of atmospheric particles, a feature that may carry adverse health consequences (Chirizzi et al., 2017, Velali et al., 2016). The study of the properties and dynamics of carbonaceous particles, including their sources and transformations, remains a formidable challenge that necessitates rigorous research efforts. Climate forcing is the change in the Earth's energy budget caused by the presence of a given compound in the atmosphere. Except for black carbon (BC) aerosol, aerosols primarily contribute to negative radiative forcing in the atmosphere and reduce the warming effect caused by greenhouse gases (Paasonen et al., 2013). The estimated radiative forcing by the direct effect (scattering and absorption) of aerosol in the fifth assessment report is -0.35 (-0.85 to $+0.15$)Wm^{-2}, which seems pretty small but associated with a large uncertainty range (IPCC, 2013). The total radiative forcing of biomass burning emissions for aerosol-to-radiation interactions is close to zero, with a strong positive forcing from BC and a strong negative forcing from OC. Because aerosols are highly variable in space and time, there is a high degree of certainty that they have offset a significant portion of the global mean forcing of greenhouse gases (Ramanathan, 2008).

Air pollution is a pressing global challenge with significant impacts not only on environmental effects but on human health too. It consists of a complex mixture of gases, vapors, and particulate matter that consistently degrades air quality (Goudarzi et al., 2018; Javanmardi et al., 2018). Cities all over the world are concerned about the declining quality of the air due to factors including unchecked traffic growth, urban sprawl, growing populations, disappearing green areas, and rising traffic emissions and waste burning (Kim et al., 2015; Kulshrestha et al., 2009; Raaschou-Nielsen et al., 2013; Rashki et al., 2013; Samet et al., 2000; Shen et al., 2010; Von Schneidemesser et al., 2015; Hoffer et al., 2020). These developments lead to increasing levels of air pollution in metropolitan areas.

Their concentrations and toxicity depend on factors such as composition, shape, size, the presence of other pollutants, and meteorological conditions (Arruti et al., 2012; Clements et al., 2014; Coronas et al., 2009; Jorquera, 2009; Kassomenos et al., 2012; Rashki et al., 2013; Wickramasinghe et al., 2011). Scientific studies establish health risks associated with inhaling these fine particulate matters, with both short-term and long-term exposure linked to respiratory and cardiovascular ailments (Stanek et al., 2011; Samoli et al., 2008). Short-term exposure worsens asthma symptoms and increases hospital admissions, especially for individuals with pre-existing respiratory conditions. Long-term exposure to PM_{10} is associated with higher mortality rates from cardiovascular and respiratory diseases, including lung cancer. The presence of recognized carcinogens among the PM components linked to black carbon raises questions regarding the possible toxicity of PM resulting from combustion. (Lelieveld, 2015). Studies using epidemiology demonstrate the link between health issues and PM_{10} exposure. In 2016, the World Health Organization estimated that outdoor air pollution, particularly exposure to PM_{10}, caused over 4.2 million premature deaths globally.

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).

2.2 Biomass burning as a source of carbonaceous aerosols

Biomass burning encompasses the combustion of living and deceased vegetation, comprising both deliberate human-initiated fires for land clearing and natural fires ignited by lightning. It is estimated that human activities are responsible for approximately 90% of all biomass burning, whereas natural fires contribute only a minor portion of the total vegetation burned (Keywood et al., 2011). The act of burning vegetation releases a substantial volume of particulate matter and various gases, including greenhouse gases that contribute to global warming (Levine, 2003). Emission of greenhouse gases has the potential to contribute to heightened Earth temperatures or anthropogenic alterations in global climate pattern. Studies have shown that biomass burning is a primary source of particulate matter, particularly during haze episodes (Abas et al., 2004). Karagulian et al. (2015) suggested that household fuel burning (involving wood, coal, and gas) could potentially account for up to 32% of $PM_{2.5}$ emissions and up to 45% of PM_{10} emissions in Central and Eastern Europe, compared to global averages of 20% and 15%, respectively. Solid fuels, predominantly wood and, in certain countries, coal, are widely employed for residential heating across Europe. During winter, residential wood combustion emerges as a primary source of fine particle emissions throughout Europe, as indicated by various studies (Tissari et al., 2008; Puxbaum et al., 2007; Gelencsér et al., 2007; Marmureanu et al., 2020). Wood burning emissions were estimated to contribute to approximately 30% of the $PM_{2.5}$ fraction in Portugal (Goncalves et al., 2012). In Lombardia, lower contributions (5−25% to PM_{10}) were estimated, even within the city of Milan (Pastorello et al., 2011; Piazzalunga et al., 2011).

In various European locations, the PM_{10} health limit is exceeded, particularly in winter when wood burning becomes more prevalent for residential heating (Chafe et al., 2015). Research findings indicate that wood burning contributes significantly to wintertime PM_{10} concentrations, constituting about 10% in Vienna and up to 20% in rural settlements (Molnar et al., 2005). Budapest experiences a substantial impact, with as much as 40% of carbonaceous aerosol emissions attributed to wood burning during winter (Salma et al., 2017). Moreover, in Bucharest, biomass burning, particularly wood burning, accounts for approximately 80% of the total carbon mass, as evidenced by a compact time-of-flight aerosol mass spectrometer (Marmureanu et al., 2020). By examining the relationship between tracer compounds produced during wood burning and meteorological parameters, it is evident that PM emitted from wood burning is not associated with long-range transport. Instead, all PM_{10} emissions from wood burning in London originate from local sources, particularly during evening hours and weekends (Fuller, 2014). In 2005, wood burning for residential heating contributed to about 40% of total $PM_{2.5}$ emissions in Europe, with 80% of these emissions being attributed to wood combustion, while the remainder resulted from coal combustion (Amann et al., 2018).

2.3 Waste burning

Besides biomass burning, the burning of different types of household wastes is also an important source of particulate matter worldwide (Christian et al., 2010, Wiedinmyer et al., 2014, Kumar et al., 2015). Open waste burning is quite common in many countries, especially where the organized waste collection system is missing, expensive or the collection frequency is low. Emissions from modern waste incinerator plants are incomparably lower than from open waste burning and from burning solid waste in household stoves (Lemieux et al. 2004, Jones and Harrison, 2016).

The World Bank estimates a significant increase in global waste production, projecting it to reach 2.2 billion tons per year by 2025 (Cogut, 2016). For instance, China with a population exceeding 1.4 billion is responsible for the largest share of the world's municipal solid waste (MSW), constituting approximately 15.5% according to the latest available data. In contrast, the United States contributed around 12% to the global MSW in 2018, despite having less than five percent of the world's population. Among OECD countries, Denmark takes the lead as the highest waste producer per capita, reaching 845 kg in 2022, nearly double the average per capita MSW generation within the EU. Denmark's substantial waste generation is linked to its predominantly urban population and elevated purchasing power (https://www.statista.com/topics/4983/

waste-generation-worldwide/#topicOverview). In the past years, waste production has improved in Hungary as mentioned in Fig. (1). It has been improved from 106 million tonnes a year^{-1} in 1990 (Ministry for Environment and Water, 2002) to around 15 million tonnes a year^{-1} in 2016 (Hungarian Central Statistical Office, 2017). This dramatic reduction can be attributed to the transition of industries to automotive and technology sector, as well as Hungary's accession to the European Union, which imposed new waste management regulations.

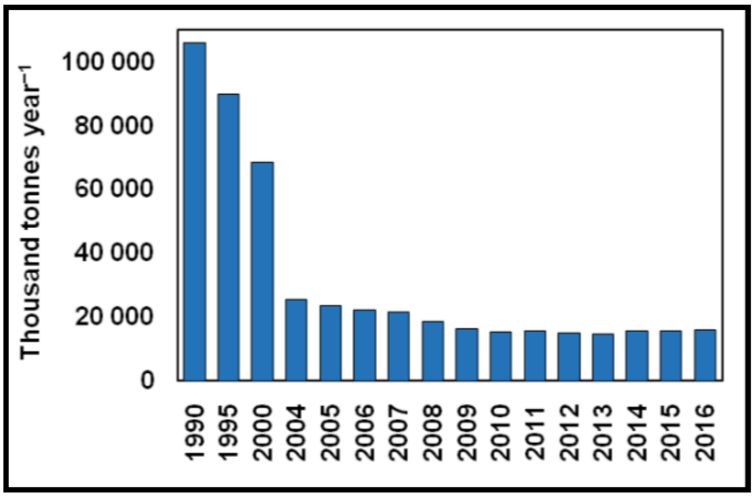

Figure 1. The total amount of the waste generated in Hungary between 1990 and 2016

According to Eurostat data, Romania produced 177,557,063 tonnes of wastes, of which 5,218,255 tonnes represent household wastes. Considering the number of inhabitants, in 2016, 9012 kg of wastes/capita were produced, while excluding mineral wastes 1084 kg of wastes/capita were produced. The total amount of wastes decreased from 369 million tonnes in 2004 to 178 million tonnes in 2016, while the household wastes increased from 1.3% (2014) to 2.9% (2016) of total wastes (Fig. 2). Generally, the main activities considered as sources of waste are: mining and quarrying, (over 80% of the total wastes generated in Romania), agriculture (approximately 10%), electricity, gas, steam and AC supply, forestry and fishing, households and services (Eurostat, 2019).

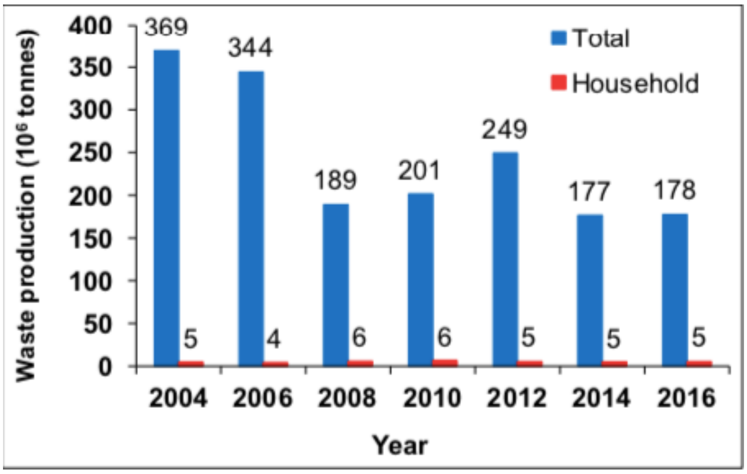

Figure 2. Trend of total and household waste production in Romania

Even though the amount of waste has been decreasing in both countries, waste can still end up in household stoves to provide domestic heating. Standard fuels (wood, coal) are typically burned alongside other household wastes such as plastic waste, textiles, furniture with resin and polishes, discarded oil, and garden waste (Czajczyńska et al., 2017).

Waste handling in Hungary involves material recovery, energy recovery, incineration, landfill disposal and alternative treatment methods (Hungarian Central Statistical Office, 2017). While in Romania the waste management primarily relies on landfilling for municipal wastes, co-incineration in cement factories and minimal composting, with incineration being the main option for industrial and medical wastes (ANMP, 2019).

2.3.1 Waste burning practice in Hungary

Assessing residential waste burning is a challenging task due to the lack of country-level records for domestic waste. To address this issue, recent studies in Hungary conducted two surveys on waste burning practices. One thousand people were polled aged between 18 and 65 years and the investigated group was deemed to be representative for age, gender, settlement type and region. Based on the answers approximately one quarter of the Hungarian population burn waste/litter in indoor appliances (furnace, stove, boiler). Similarly, about one-quarter of the population burns waste/litter in open fires outdoors. By combining the answers it turned out that 32% of the population burns waste/litter either indoors or outdoors with varying frequency (Kantar Hoffman LTD, 2017). Burning waste in general was observed to be more prevalent among individuals with primary education residing in small towns and villages. Meanwhile, indoor waste burning was more common in Eastern Hungary and the southern region of the Great Hungarian Plain, potentially linked to the lower incomes characteristic of this area.

People burn mainly garden and household waste (Fig. 3). Twenty-two percent of the population burns leaf litter and branches, 17% outdoor and 5% indoor for domestic heating. The second most common type of waste incinerated at home is household waste: 13% of people burnt such waste in the last few years. Both treated wood (furniture, OSB, hardboard, painted wood, plywood) and clothes were burnt by 6% of the total Hungarian adult population while 13% fired plastics, tires and used oil. In contrast to garden waste the majority of household waste, treated wood, clothes, plastics, neighborhood and used oil are burnt indoor for heat generation (Fig. 3). This implies again that there is a link between economic situation and waste combustion.

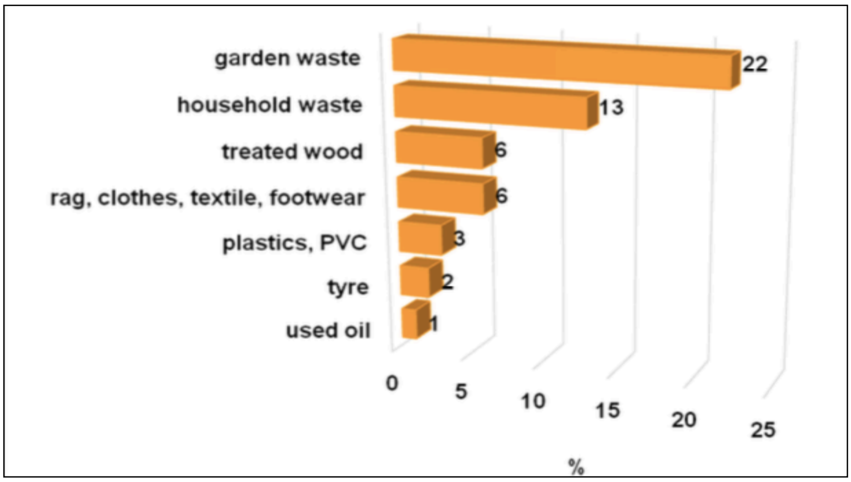

Figure 3. Abundance of waste types burnt in the households (n=1000)

It is essential to note that the frequency of burning differs considerably for different waste types: 90% of garden waste burners do this half-yearly or even more rarely and only 10% burns it monthly or weekly. Household waste is burnt more frequently: 28% of those people who burn household waste burn it monthly or weekly.

Besides that, the reasons for illegal waste burning were also investigated in the study. According to 54% of respondents, poverty, carelessness, and ignorance of the consequences for one's health and/or the environment as well as regulations all contribute to illegal waste burning, 30% reckoned lack of knowledge and irresponsibility as the main reasons and only 15% believed that poverty was the only cause. A total of 69% of respondents believed that poverty was a factor, whereas 84% blamed ignorance and carelessness.

According to the survey, since the cost of burning household or other waste is cheaper than other domestic heating fuels, last year around 48.5% of the Hungarian population experienced waste burning in their living environment (55.5% in Eastern Hungary, 37.3% in Central Hungary and 49.3% in Western Hungary), 38.7% outdoor and 22.5% assumed waste burning in indoor appliances in the neighborhood. According to the survey results, illegal waste burning is performed due to poverty, a lack of knowledge, and irresponsibility.

2.3.2 Waste burning practice in Romania

Romania has one of the lowest recycling rates in the EU and one of the highest rates of landfill disposal (96%). Waste incineration for electricity and thermal energy is not a common practice in Romania nowadays. Numerous projects are on hold because of high expenses and a deficiency of environmental approvals. The industry firmly believes that producing electricity from domestic waste through high-temperature combustion is a cost-effective substitute for disposing of waste. On the surface, it appears to be a highly appealing approach because incinerators generate energy and lessen the quantity of waste dumped in landfills. Currently, there are no energy recovery uses for feedstock made from municipal waste, such as incinerator plants that provide heat for central heating systems. In various Romanian villages, the daily burning of household waste is commonplace due to the absence of organized waste collection systems. The combustion of any type of waste in these areas poses severe health risks, emitting a plethora of toxic, carcinogenic, or mutagenic compounds in substantial quantities compared to authorized solid fuels like dry wood or high-quality coal (Lemieux et al., 2004; Estrellan and Lino, 2010; Gullett et al., 2010; Wiedinmyer et al., 2014). Industrial emissions Directive 2010/75/EU, which has been incorporated into national legislation by the Industrial Emissions Act 278/2013, covers all installations authorized for the co-incineration/incineration of waste on Romanian territory. Incineration plants for municipal solid waste are required to adhere to Directive 2008/98/EC on waste and repealing some directives, Annex 2, point R1. In this particular case, the process of incineration might be regarded as a recovery operation (EC, 2015). Biogas, storage gas, and wood waste (wood waste from demolition, wood processing, and wood waste from crops) are wastes from which energy can be recovered.

2.4 Tracer compounds

Tracer compounds, which are substances deliberately introduced or naturally occurring and serve as unique markers, have emerged as powerful tools in identifying and understanding biomass burning, fossil fuel, cigarette smoke, tire wear and illegal waste burning incidents. These molecular tracers are organic compounds that are chemically specific to particular emission sources. Moreover, they are non-reactive and do not undergo chemical transformations in the atmosphere.

For example, 135-TPB (1,3,5-triphenylbenzene) was proposed as a unique tracer for garbage burning emission and levoglucosan as a tracer for wood burning (Simoneit et al., 2005). Molecular tracer compounds are usually present at trace concentrations in atmospheric aerosol, so their contribution to the mass of aerosol particles is negligible. Levoglucosan is the only exception, its concentration can be important in the case of biomass burning. The presence (or absence) of these tracer compounds may provide crucial indications about the potential contribution of specific emission source types to air pollution. It should be mentioned, however, that the information obtained from the molecular tracer approach will never unveil the contribution of any individual point source in the vicinity of the sampling site due to the longer residence time of particulate matter and the extended time of sample collection for chemical analysis at a sampling location. Alternatively, it can be considered as a spatially and temporally weighted representative average of the region's pollution source contributions. When collecting a limited number of particulate samples at fixed sampling locations, this is a distinct advantage. The molecular tracer approach is widely used to assess the impact of a major source of air pollution.

2.4.1 Tracer compounds of biomass burning

Due to the unique signatures of organic molecular tracers in the atmosphere, they play an important role in identifying biomass fuel combustion (Simoneit et al., 2000; Simoneit et al, 2002). Cellulose, hemicellulose, lignin, are important units of biomass fuels, and their pyrolysis products are widely used as unique tracers of biomass burning in the atmosphere (Kawamura et al., 2012; She et al., 2019).

Wood is typically composed of lignins (20-30%), cellulose (40-50%), hemicellulose (20-30%), and external compounds (extractives and ash together (4-10%)). Cellulose serves as a structural mesh that is reinforced by lignin polymers. When heated (above 300°C), the wood's constituents begin to hydrolyze, and pyrolyze, forming combustible volatiles, tarry anhydrosugars, and carbonaceous char. This mechanism produces levoglucosan (1,6-anhydro-β-D-glucopyranose), the most abundant among anhydrosugars it resists photolysis and acid-catalyzed hydrolysis for at least 10 days (Simoneit et al., 1999); (Nolte et al., 2001); (Caseiro et al., 2009); (Locker, 1988); (Fraser and Lakshmanan, 2000); (Simoneit et al., 2004). Besides levoglucosan,; its stereoisomers mannosan (1,6-anhydro-β-D-mannopyranose); and galactosan (1,6-anhydro-β-D-galactopyranose)—are prevalent in the aerosol phase too. Furthermore, once wood is burned, this compound (LGS) is emitted at an extremely high rate (40-1200 mg kg^{-1} of wood) (Simoneit et al., 1999). In pine smoke, the concentration of levoglucosan in particulates can reach 150 mg g^{-1} PM (Sheesley et al., 2003). Levoglucosan is an ideal universal tracer for wood burning due to its exceptionally high concentrations and good atmospheric stability ((Locker, 1988; Meng et al., 2020; Li et al., 2020; Hennigan et al., 2010)). The concentration of levoglucosan is also used to estimate the quantity of particulate matter emitted from biomass burning (Abas et al., 2004).

As a biogenic source indication for conifer resin, dehydroabietic acid has been applied either directly to the aerosol or by the burning of wood (Standley and Simoneit 1994). In smoke samples from biomass combustion triterpenoids can be used as organic tracers for the contribution directly from vegetation (Elias et al. 1997). The triterpenoids that most frequently found in plants are α-amyrin, β-amyrin and their derivatives. These pentacyclic triterpenes are tracers for angiosperm waxes (Oros and Simoneit 2001a, b). Besides hardwood produces the guaiacol (2-methoxyphenol) derivatives in the smoke, as softwood, hardwood also originates high levels of syringeol (1,3-dimethoxyphenol) derivatives (Hawthorne et al. 1988, Simoneit 2002). Vanillin and vanillic acid are detected in pine wood smoke. Additionally present in trace levels are p-anisic acid, syringic acid, and syringaldehyde. Syringaldehyde and syringic acid are more prevalent in hardwood smoke (such as oak), with trace levels of additional compounds of the sinapyl and vanillyl types. P-anisaldehyde, p-anisic acid, and, to a lesser degree, p-coumaryl, vanillyl, and syringyl type lignin pyrolysis products are the components of grass smoke (Simoneit 2002). The leading lignans of softwood smoke are matairesinol and shonanin (tetrahydro-3,4- divanillyfuran). Minor quantities of methylated derivative are detectable. The hardwood smoke encompasses a variety of highly oxygenated isomers of dihydrovanil- lylsyringyl-2(3H)-furanone, dihydro-3 (2’, 3’, 4’-trimethoxybenzyl) - 4 - veratryl - 2(3H) - furanone and tetra- hydro-3-vanillyl-4-veratrylfuran, which are released unaltered from the combustion sources and may be considered tracers of biomass burning. In addition, softwood smoke includes derivatives from coniferyl-type precursors, such as divanillyl, divanillylmethane, bis(3,4-dimethoxyphenyl) methane and diveratryl. Hardwood smoke comprises components originated from sinapyl-type precursors, including bisguaiacylsyringyl, disyringyl and bis(3,4,5-trimethoxyphenyl)ethane. Dianisyl, a derivative from a p-coumaryl-type precursor, is the only tracer in the grass smoke (Simoneit 2002).

2.4.2 Tracer compounds of waste burning

Key and macro-tracers

a) 1,3,5-triphenylbenzene

1,3,5-triphenylbenzene (TPB) was proposed as a unique tracer for garbage burning emission due to its high specificity and relatively high specific emission (Simoneit et al., 2005). Thus, it may be considered as a macro-tracer similarly to the case of levoglucosan in wood burning. The formation of 1,3,5-triphenylbenzene in smoke from the burning of plastics is linked to the self-condensation reaction of acetophenone which emerges from the styrene chain terminator added to formulations during ethylene polymerization (Simoneit et al., 2015). 1,3,5-triphenylbenzene has a melting point of 172-174°C, similar as benzo[a]pyrene (177-180°C). In the analysis, care has to be exercised since 2,9-dimethylpicene, a dominant isomer volatilized into smoke from the burning of semi-bituminous or bituminous coals, yields the very same molecular ion of m/z 306 (Simoneit et al., 2005). The emission factors of 1,3,5-triphenylbenzene are 0.38-1.87 mg kg^{-1} for mixed waste burning, 0.27 mg kg^{-1} for the burning of foil wrappers, and 0.55 mg kg^{-1} for the burning of plastic bags. However, 135-TPB was not detected in emission of other combustion sources (diesel generator, gasoline generator, gasoline powered motorcycles, burning of coal, sugarcane residue, hardwood), further highlighting its specificity to garbage burning (Jayarathne et al., 2018). Particulate-bound 1,3,5-TPB emission factor was found to be in the order of 0.025-0.075 mg kg^{-1} for the burning of PET in a recent study (Tomsej et al., 2018). It should be noted, however, that emission factors may span at least 2 orders of magnitude, thus caution must be exercised when calculating the contribution of waste burning to local atmospheric particulate (PM_{10}) pollution.

b) Organophosphate esters (OPEs)

Organophosphate esters (OPEs) are a diverse group of chemical compounds widely manufactured and extensively used in numerous household and industrial products. They are valued for their versatility and serve various purposes, with a primary focus on their role as plasticizers and anti-foaming agents. These compounds are integral components in a broad spectrum of consumer and industrial goods, e.g., OPEs are used in the manufacture of various furniture items, enhancing their flexibility and durability. They are also incorporated into textiles and fabrics, imparting flame resistance and enhancing their overall quality. OPEs contribute to the flexibility and insulation of electrical and data cables. These compounds find application in building materials, including insulation products and construction materials, in paint formulations to enhance their performance and durability (Liu and Mabury, 2018)). Unlike certain emissions commonly associated with combustion sources like coal, biomass, or vehicles, OPEs, including the specific compound tris (2,4-di-tert-butylphenyl) phosphite, are not typically identified in the smoke generated from these processes, thus considered as specific tracers for the combustion of thermoplastic materials. Among OPEs, one particular compound, tris (2,4-di-tert-butylphenyl) phosphite, has garnered attention due to its significant presence in the particulate matter resulting from the combustion of plastic materials. Notably, this compound is not directly released into the environment during the manufacturing or use of plastic products. Instead, it emerges as an oxidation product derived from a tris(2,4-di-tert-butylphenyl) phosphite, antioxidant known as Irgafos 168. This phosphite antioxidant is a prevalent additive in polymer products, including various thermoplastic materials. The formation of tris (2,4-di-tert-butylphenyl) phosphate during plastic combustion is an important indicator of the incineration of plastic waste. This chemical transformation underscores the complexity of emissions generated from plastic materials and provides valuable insights into the specific compounds released during their combustion (Medeiros et al., 2007). These components can be identified based on their molecular ions by GC-MS (Bi et al., 2010.) The research of Irgafos 168's leaching from plastics was prevented by the identification of the compound and its oxidation product, TBPP, in laboratory environments as well as in blanks, therefore the quantitative analysis is not a simple task (Hermabessiere et al., 2020).

c) Phthalate esters

Phthalate esters are a class of chemical compounds that find widespread application as additives in various plastic materials. These plastics include polyvinyl-chloride, polyethylene terephthalate, polyvinyl acetates, and polyurethanes. Phthalate esters are added to these plastics to impart characteristics such as softness, increased flexibility, and elasticity. However, it's essential to note that these additives are not chemically bonded to the polymers; instead, they are physically incorporated. This characteristic makes them susceptible to vaporization at elevated temperatures, particularly during combustion processes (Bi et al., 2008). Within the family of phthalate esters, di-(2-ethylhexyl) phthalate (DEHP) and n-dibutyl phthalate (DBP) emerge as the two most prevalent compounds found in the emissions generated by the combustion of plastic materials. These two compounds are distinctive tracers for plastic waste burning. While they are primarily detected in the gas phase of emissions, a notable fraction of these compounds can also be identified in the particulate phase, underscoring their significance as tracers for plastic combustion (Bi et al., 2008). Another noteworthy compound serving as a tracer for plastic waste combustion is diisobutyl phthalate (DiBP). In aerosol particles originating from the combustion of plastic, the typical concentration ratio of DiBP to DEHP is approximately 1:2 (Fu et al., 2010). While several other compounds, including diphenyl sulfone (CAS 127-63-9) and dioctyladipate, have been identified, their role as universal tracers has not been definitively confirmed (Giri et al., 2013). Additionally, research has shown that dimethyl phthalate is the predominant phthalate species in particles collected from environments such as electronic waste (e-waste) pyrolysis plants (Gu et al., 2010). Although phthalate esters are widely present in plastics, they are not specific, it is challenging to prevent cross-contamination during analysis. Additionally, because these chemicals are highly volatile, particulate-phase measurements may be impacted.

d) Phthalic acids

Terephthalic acid, also known as 1,4-benzenedicarboxylic acid or t-Ph, is a chemical compound used in the manufacturing of polyester fibre and thermoplastic polyethylene terephthalate (PET). It is essential to the synthesis of these materials, which are widely utilized in bags, fabrics, and beverage containers, among other applications. The burning of plastic bags and roadside trash results in the presence of t-Ph, which makes it a significant tracer chemical for these emissions (Kawamura et al., 2010). Experiments related to these activities have revealed t-Ph emission factors ranging from 176 to 5033 $\mu g\ g^{-1}$ PM (Simoneit et al., 2005). The significance of t-Ph as an atmospheric tracer is further highlighted by its prevalence in urban areas that are polluted. It has been noted that the mass concentration in these regions varies, ranging from 7.6 to 168 ng m^{-3}). Specifically, data from an open garbage burning location in India were very impressive and the most common diacid was found to be t-Ph. It demonstrated the frequency of waste combustion emissions by exceeding the concentration of oxalic acid at this site by a factor of 14 (Kumar et al., 2015). Materials like polyethylene, foam, paper, packing materials, and clothes made up the majority of the flammable waste composition at this location. It is especially important to remember that t-Ph's atmospheric stability is comparatively restricted. This restriction results from its vulnerability to photo-Fenton oxidation, which breaks it down into fumaric and maleic acids. This phenomenon has the ability to change the chemical makeup of t-Ph since it is fueled by light and reactive iron species. Because of this possible alteration under atmospheric circumstances, the utility of t-Ph as a long-range atmospheric tracer is questioned (Kumar et al., 2015).

An alternative compound for tracing aged aerosol particles is phthalic acid (Ph), which exhibits greater stability in the atmosphere. Phthalic acid is also found in fresh waste burning aerosol, and unlike t-Ph, it is preserved during atmospheric transport. Research has demonstrated that the transformation of diethylhexyl phthalate (DEHP) and terephthalic acid into phthalic acid occurs within plastic-waste burning smoke particles. This transformation is a result of hydrolysis, a chemical reaction that modifies the composition of these compounds. Consequently, over time and as the smoke plume disperses into the atmosphere, phthalic acid becomes increasingly prevalent compared to DEHP. This difference is due to the respective half-lives of these compounds, with DEHP having a half-life of about 6 hours, while phthalic acid boasts a significantly longer half-life of approximately 100 hours. This temporal evolution of phthalic acid concentration should be taken into consideration when interpreting measured phthalate concentrations as atmospheric tracers of plastic waste combustion, as it reflects the compound's transformation and stability during atmospheric transport (Kumar, S., 2015). However, phthalic acid was found in high abundance in the smoke from burning low-quality coal in China (He et al., 2018), and it was also found to be emitted from other (traffic-related or biomass burning) sources (Fraser et al., 2003; Fine et al., 2004; Al-Naiema and Stone, 2017), which challenges its use as a specific tracer.

Other tracers

a) Nitroarenes

Nitroarenes are chemical derivatives of pyrene, a polycyclic aromatic hydrocarbon (PAH) that is common in emissions from the combustion of these synthetic materials. These derivatives include 1-NP and 1,3-dinitropyrene, 1,6-dinitropyrene, and 1,8-

dinitropyrene. What makes them unique is the presence of NO_2 functional groups, which are introduced through chemical reactions during the high-temperature combustion processes. It has been found that these nitroarenes are strong mutagenesis agents that have the potential to alter the genetic material of an organism and endanger human health. The carcinogenic potential of 1-NP and DNPs emphasizes how crucial it is to comprehend their existence in emissions from burning synthetic materials. These substances' capacity to interact with cellular DNA and create genetic material alterations makes them very dangerous. They contribute to an increased risk of cancer and other harmful health effects in those who are exposed to them since they are mutagenic agents. These nitroarenes have been found in studies to be present in emissions from a variety of combustion sources, such as home burning of synthetic materials, industrial processes, and trash incineration. These results highlight the necessity of thorough evaluations of the health and environmental effects of emissions including nitroarenes as well as the creation of mitigation plans to lessen these effects on human health and air quality. Table 1 shows the 1-NP and DNP concentrations in particle extracts from the burning of different polymers. However, they were also found in diesel engine exhaust (Keyte et al., 2016).

Table 1. Measured concentrations (ng/g) of 1-NP and DNPs in the extracts of particulates from the combustion of different plastics (Lee et al., 1995).

Plastic type	1-NP	1,3-DNP	1,6-DNP	1,8-DNP	Total amount (ng/g)
PVC	25.1	17.0	73.1	31.6	146.8
PET	0.6	44.5	76.8	20.6	142.5
PS	23.6	2.0	3.7	12.0	41.3
PE	2.1	0.2	1.2	1.2	3.8

b) Bisphenol A

Bisphenol A is a chemical compound that serves as an intermediate in the production of epoxy resins and polycarbonate plastics. Notably, it is widely utilized in the lining of food cans and the manufacturing of beverage bottles, including those intended for baby bottles. What makes Bisphenol A particularly significant in the context of environmental research is its identification as a potential tracer in smoke particulates resulting from the combustion of plastic waste. Studies have revealed a robust correlation between Bisphenol A concentrations and those of other tracers such as 1,3,5-triphenylbenzene and tris(2,4-di-tert-butyl-phenyl) phosphate (TBPP). This correlation strengthens its role as a valuable marker for identifying the combustion of plastic materials, shedding light on the presence and fate of this compound during plastic waste incineration (Fu et al., 2010). P-quarterphenyl is another possible tracer for the burning of PET in co-combustion experiments (Tomsej et al., 2018).

Less specific elemental tracers for waste burning

a) Chlorine

Studies examining the chemical composition of particulate emissions from waste combustion processes have focused notably on chlorine, an elemental component included in many manufactured products. A significant amount of chlorine was found in the released particulates in a study on the composition of waste burning-related particles in central Mexico (Christian et al., 2010). This result emphasizes how important it is to comprehend how chlorine behaves and what effects it has on the ecosystem when garbage is burned. Researches of landfills within the European Union revealed that solid waste had a chlorine level of approximately 9 g kg^{-1} (Mersiowsky et al., 1999). Furthermore, nearly all of the chlorine was found in polyvinyl chloride (Costner, 2005), which makes up 57% of the trash's mass (Christian et al., 2010). It should be noted that a significant amount of the chlorine content is related to polyvinyl chloride (PVC), a typical synthetic substance with a mass percentage of roughly 57% chlorine. The majority of the chlorine is emitted as gaseous hydrochloric acid (HCl) when PVC-containing materials burn. This gaseous HCl is a well-known pollutant in waste combustion emissions.

b) Trace metals

Metals are used as stabilizers, pigments, and catalysts in plastic additives, among other functions. These metals, which are frequently included in synthetic materials and thus discharged into the environment during waste combustion processes, include antimony (Sb) and tin (Sn). It is crucial to comprehend both their presence and effects on the environment. Particularly when considering waste burning emissions, antimony (Sb) and tin (Sn) have been discovered to have the greatest concentrations among the metals detected in waste combustion emissions (Christian et al., 2010). Antimony is used in a number of instances, one of which is as a flame retardant in textiles. Brake wear emissions have been also cited as a potentially important source of antimony (Sb). Brake linings contain 1–5% Sb in the form of stibnite (Sb_2S_3), which is employed as a lubricant to reduce vibrations and improve friction stability (Roubicek et. al., 2008). Stibnite can be oxidized during the braking process to antimony trioxide (Sb_2O_3), which has been categorized as a potentially carcinogenic substance (Varica et. al., 2013). It is also an essential component of antimony trioxide, which is widely used in the synthesis of polyethylene terephthalate (PET), and lead alloys for batteries. PET is a multipurpose substance that is used to make polyester fibers for textiles and soft drink bottles. The concentration of antimony in $PM_{2.5}$ generated from waste incineration in Mexico was 0.011 g kg^{-1}. For an elemental tracer, this concentration is noteworthy, suggesting that antimony contributes significantly to emissions from the combustion of waste (Hodzic et al., 2012). Antimony's importance as a specific tracer for some synthetic materials is highlighted by its presence in waste combustion emissions, which is not surprising given its widespread use in the production of textiles, break wear, batteries and PET. In a different investigation, phthalates in open waste burning emissions showed a substantial association with tin (Sn), another metal identified in waste combustion emissions. According to this correlation, tin may be used as a useful tracer for the combustion of garbage connected to plastics and related materials (Kumar et al., 2015).

3. Objectives

Thousands of tons of various materials, including plastic, clothing, combustible construction waste, paints, and litter, may end up in residential stoves. This realization highlights the potential for substantial environmental and health impacts resulting from waste burning in residential areas. Waste burning, particularly in residential stoves, generates an exceptionally large amount of particulate matter (PM) emissions. These emissions pose a significant health risk to millions of people living in proximity to such sources. Moreover, it's important to note that this risk is associated with extremely hazardous particulate matter even before accounting for the additional health threats posed by the associated gaseous emissions.

The primary goal of this research is to identify specific tracer compounds associated with the burning of different types of waste in household stoves. These compounds are invaluable as they can serve as unique tracers, allowing us to distinguish waste burning emissions from other sources. This identification process is critical for accurately assessing the contribution of waste burning to air quality.

Gas-chromatography mass spectrometry will be employed to analyze the collected samples. This analytical approach is crucial for identifying new tracer compounds.

Emission factors for the identified tracer compounds will be determined. These factors are critical for quantifying the emissions associated with waste burning. They provide essential data for understanding the extent of waste burning's impact on air quality since some of the tracer compounds can be emitted from different types of waste.

Analyzing the concentration of tracer compounds in ambient samples, collected from various settlement types in Central and Eastern Europe, allows the research to estimate the contribution of waste burning to the PM_{10} concentration. This step provides concrete insights into the extent to which waste burning affects particulate matter levels.

Estimating the amount of material being incinerated in residential stoves is an exciting aspect of this research. This information can be compared to statistical data on firewood consumption to gain a clearer understanding of the scale of waste burning in relation to traditional wood combustion.

The chemical compounds identified as tracer compounds can undergo several complex processes in the atmosphere. Understanding how these compounds might evolve is critical. To this end, the research will involve the exposure of different samples to varying atmospheric conditions over time. This step will provide valuable insights into the behavior of these tracers in the environment.

In conclusion, by identifying specific tracers, quantifying emissions, and assessing their impact on air quality this research project aims to provide a solid foundation for informed decision-making and policy development related to waste management and environmental protection.

4. Experimental

4.1 Laboratory waste burning experiments

The solid waste specimens used for the combustion tests were selected based on their abundance in households (Fig. 3) and available information on illegal waste burning practices in Hungary (Hoffer et al., 2020) (see 2.3). The following are the selected for waste combustion tests: Polyethylene terephthalate (PET), Polystyrene (PS), Polypropylene (PP), Polyethylene (PE), Polyvinyl chloride (PVC) Polyurethane (PU), Acrylonitrile Butadiene Styrene (ABS), tire (TR), OSB, laminated Melamine Low-Density Fiberboard (LDF), rag (RAG), paper (PAP), and firewood (WOOD) (Fig. 4).

Figure 4. Solid-waste specimens for combustion tests

To investigate the PET waste samples for combustion testing, plastic bottles used for beverages with a volume of 1.5 or 2 L were selected, and the caps and labels were removed. The PS waste samples consisted of hard cups or pots used for dairy products such as yogurt and pudding, and pieces of expanded insulation board with a thickness of 80 mm. The hard and expanded PS waste samples were burned separately. For the PP waste sample specimens, a combination of plastic cups and pots used for dairy products like sour cream and pudding, without paper labels or aluminum foil, and plastic trays of meat were used. For the preparation of the PE waste specimens, a mixture of high-density polyethylene (HDPE) and low-density polyethylene (LDPE) was used. The HDPE and LDPE fractions consisted of plastic caps from beverage bottles and pieces of various foil and plastic bags, respectively. The PVC waste samples were made up of soft packaging, pieces of vinyl flooring, and water pipes. The PU waste samples consisted of pieces of packing sponge with an average size of 120 mm × 100 mm × 15 mm. The ABS samples used for combustion testing were made up of shredded pieces of computer monitor stands with an average size of 40 mm × 40 mm × 20 mm. The OSB samples consisted of slices of OSB material from various brands with an average size of 120 mm × 100 mm × 20 mm. The LDF samples were made up of pieces of different types of fiberboard, including colored laminated coating and plastic borders, with an average size of 130 mm × 100 mm × 20 mm. The TR samples were composed of pieces of both new and old van and passenger car tires with an average size of 80 mm × 40 mm × 15 mm. The RAG samples used in the combustion tests were made up of a combination of cotton, polyester, and polyamide fabrics from different types of clothing. Two types of PAP samples were used, one being colorful glossy-coated paper and the other uncoated paper from advertising flyers and newspapers, with ball-shaped specimens weighing approximately 70 g. The WOOD samples consisted of pieces of logs, weighing approximately 130 g, from Turkey oak (Quercus cerris) and black locust (Robinia pseudoacacia). All sample specimens were weighed using a precision balance (BSM-4200-2, Petra Mechatronics) before being tested for combustion.

Combustion experiments were conducted using a commercially available cast-iron stove (Servant S 114, with a heating capacity of 5 kW). The stove was preheated with charcoal for about 1 hour before the combustion tests commenced. This preheating process established a minimal baseline for particulate emissions during the entire measurement period. In each combustion trial, 1 to 10 portions of solid waste were incinerated, the number depending on the emission characteristics of the specific waste type. The stove employed was a readily available model that enabled to regulate the airflow into the stove's combustion chamber via an adjustable opening. This slit had two endpoints, enabling the introduction of either a larger amount of fresh air (high air supply ratio) or a smaller amount (low air supply ratio) into the combustion area. Each waste type (excluding PAP) was incinerated at high, mixed, and low air supply levels. This resulted in varying temperatures and conditions within the stove, but these factors were largely unaffected by the specific waste type burned at any given air supply ratio. A K-type thermocouple thermometer (Testo 925, maximum temperature 1000°C) was employed to measure the flue gas temperature both before and after each test. This measurement was taken at a point in the stack situated 11 cm above the stove's exhaust opening. In between different sample runs, the stove and stack were heated to temperatures exceeding 700°C for a minimum of 10 minutes to minimize any potential cross-contamination that might occur between combustion tests involving different waste types. The temperature of the flue gas was a crucial indicator of the experimental conditions for each combustion test run. Throughout the experiments, the temperature of the flue gas remained consistent, with mean temperature values of 299°C (SD 11°C) and 233°C (SD 10°C) recorded at high and low air supply, respectively.

The concentrations of CO_2 and O_2 in the flue gas were determined using a CO_2-O_2 analyzer (Servomex). A fraction of the flue gas was extracted from the end of the stack through 5 mm i.d. brass tubing, introduced into an 80 L dilution unit, and mixed with ambient air to achieve dilution. The CO_2 concentration in the diluted flue gas was continuously monitored using a CO_2 analyzer (SensAir) placed at the inlet of the filter sampling head. The dilution ratio was calculated by comparing the CO_2 concentrations at the stack's exit with those in the dilution unit, considering the ambient CO_2concentration. On average, the dilution ratio was approximately 81.2 (with a standard deviation of 12). This ratio remained consistent across various air supply settings. The sampling procedure closely resembled the natural cooling and dilution of smoke released from a chimney into the open air, as both involve rapid cooling and dilution of the smoke. Sampling for each PM_{10} aerosol sample commenced once stable baseline values for CO_2 and O_2 concentrations were achieved. Sampling concluded when the combustion of all waste samples was completed, and the CO_2 and O_2 concentrations returned to stable baselines.

PM_{10} aerosol samples were obtained using 150 mm diameter quartz filters (Advantec QR100 quartz fiber, binder-free) and a high-volume aerosol sampler (flow rate: 32 m^3h^{-1}; Kalman System Co., Hungary). Blank samples, which served as background measurements, were collected alongside the waste samples. These blanks were generated by burning only charcoal and had comparable sampling durations to the actual waste samples. For each waste type, a single blank sample was collected under combined air supply settings.

The quartz filters were conditioned in a controlled environment with a temperature of 20±1°C and a relative humidity (RH) of 45%–50% for a period of 3 days. Before and after aerosol sampling, the filters were weighed following the European standard (MSZ EN 12341:2014). The environmental parameters (RH and temperature) were continuously monitored and recorded using a data acquisition system.

To prepare for sampling, the weighed filters were stored in glass petri dishes preheated at 450°C. After exposure to aerosols, the filters were preserved in the freezer, placed in glass petri dishes, and wrapped in aluminum foil until they underwent conditioning and subsequent measurements.

Preliminary tests were conducted to optimize the mass of waste sample specimens burned, aiming to achieve PM_{10} concentrations of approximately the same magnitude in each combustion test. This step was essential due to the significant variation in particulate emissions from different waste types (e.g., PS, PP, PE, PVC, PU, ABS, and TR emitted significantly more particulates compared to PET, OSB, LDF, RAG, PAP, and WOOD). The goal was to prevent excessive loading of the filters and the measurement equipment, ensuring that the measurements remained comparable. As a result, the measured PM_{10} mass on the filters fell within the range of 5.4 to 37.2 mg for all combustion tests.

It's worth noting that the mass of the specimens may influence the combustion mechanism and, consequently, the calculated emission factors (EFs). However, the consistency observed with previous EFs suggests that the approach and the results are reliable. The EFs were determined based on the weight of the waste specimens placed into the stove. It's pertinent to mention that the ash content of the plastics, as reported by Zevenhoven et al. (1997) for LDPE, HDPE, PP, PVC, and PS, is less than 3%. It is reasonable to assume that other combustible plastics have similarly low ash content.

4.2 Ambient samples

4.3.1 Sampling sites

PM_{10} samples were collected in 9 locations (5 from Hungary (Veszprém (VES), Putnok (PUT), Miskolc (MSK), K-puszta (KPS), Budapest (BUD) and 4 from Romania (Cluj-Napoca (CLJ), Deva (DEV), Focsani (FOC), Magurele/Rado (MAG/R)). Furthermore, in winter 2020 PM_{10} samples were collected also in Magurele/Mars (MAG/M) site as shown in Figure 5.

The Miskolc measurement station, situated in Búza Square, is classified as a traffic-impacted site due to its proximity to two main roads in the city center. Putnok is located in the Sajó valley, which is considered the most polluted area in Hungary, and the sampling site was classified as suburban, located in the courtyard of the High School for Agriculture. Samples collected in Budapest represented suburban conditions, taken from the garden of the Hungarian Meteorological Service György Marczell Observatory, which is situated 14 km southeast of the city center. K-puszta is a regional background air quality monitoring station located in a forest close to Kecskemét. In Veszprém, urban background samples were collected at Patak Square. The urban sampling station in Deva was located in Dragos Voda Street in the yard of Devagold Company, in Deva city center, approximately 10 km from the Mintia-Deva Power Station. The sampling station in Cluj-Napoca was classified as a suburban station, situated in the western part of the city (Donath Street), in the yard of the Research Institute for Analytical Instrumentation, near two main roads and the suburban air quality monitoring station of the Environmental Protection Agency. In Focsani, the sampling was performed in the yard of a private house on Cuza Voda Street in a suburban area. PM_{10} samples were collected near Bucharest in Măgurele, about 12 km southwest of the city. Two sampling stations were used in Măgurele for PM_{10} sampling: one located in the yard of the National Institute for Research and Development for Optoelectronics - INOE 2000 (BUC-R), and the other (BUC-M) situated 2 km away from the first station on Atmosferei Street, which was only utilized during the winter of 2020.

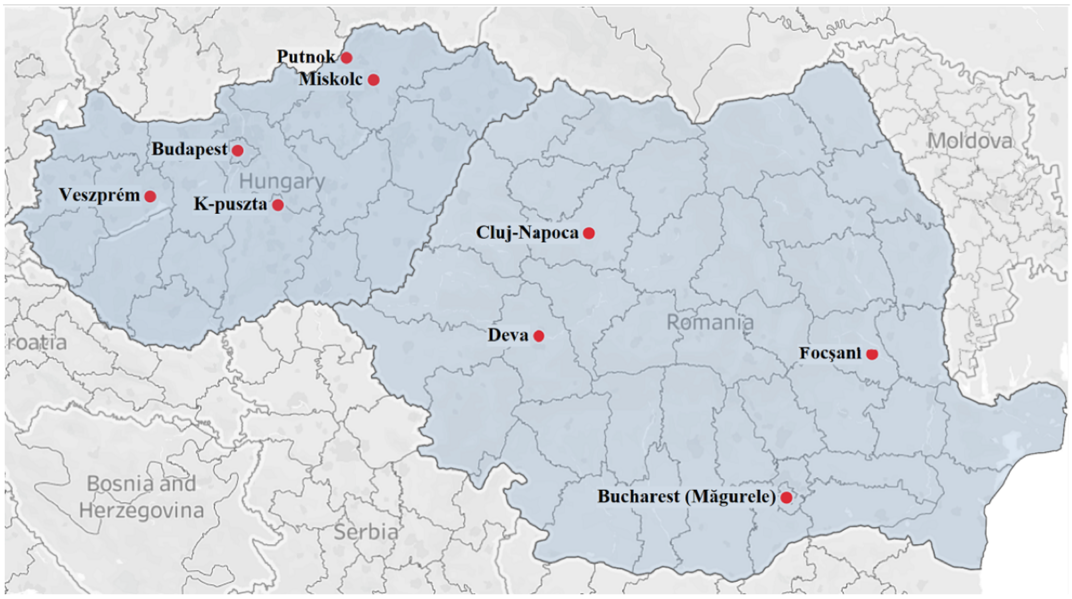

Figure 5. Geographic locations of the sampling sites.

The Romanian samples were collected by cooperation with the Institute for Environment and Energy, Technology and Analytics (IUTA) in Romania. The sampling and the weighting of the filters were done by partners involved in the project "Analyzing the effect of residential solid waste burning on ambient air quality in central and eastern Europe and potential mitigation measures" (no. 07.027737/2018/788206/SER/ENV.C.3) and the project RRF-2.3.1-21-2022-00014 of the National Multidisciplinary Laboratory for Climate Change.

4.3.2 PM_{10} filer sampling and meteorological conditions

Samples of PM_{10} were collected at all stations using high-volume samplers (Digitel DHA-80) and quartz filters (Advantec QR-100) for 24 hours, starting at midnight. The conditioning of the filters before and after the sampling took 3 days in a weighing room. The temperature was set to 20 ± 1°C and the relative humidity to 45−50%. The parameters were measured and collected by a data acquisition system. The PM_{10} mass concentration was measured using the gravimetric method specified in EN 12341:2014.

In the heating season of 2018/2019 and 2019/2020, as well as in the summer of 2019, PM_{10} filter samples were collected and analyzed (Table 2). During the winter of 2019, the 10 most polluted samples for GC-MS analysis were selected. In contrast, almost all samples collected on consecutive days during 2020 were analyzed. This is evident in Table 2, which shows lower average PM_{10} concentrations for the winter of 2020 compared to 2019. Throughout the sampling periods, blank samples were collected at the sampling sites and were also subject to analysis. Table 2 summarizes the sampling dates, the number of samples analyzed, and the average PM_{10} concentrations obtained from the gravimetric analysis.

Meteorological conditions during the sampling and measurement campaigns were examined across different seasons: In the heating season of 2018/2019, high-pressure regimes prevailed in December and February, while January experienced a cyclonic regime. Warmer temperatures were observed in December, while January was cooler than average. Summer 2019 was notably warmer than average, with anticyclonic surface pressure regimes dominating the region. The heating season of 2019/2020 also witnessed warmer temperatures than average, with high-pressure systems prevailing throughout the entire winter season. January 2020 stood out with particularly high-pressure values and negative temperatures over Pannonian and Transylvanian basin, due to a prolonged overcast period with stratus cloud and fog.

Table 2. The sampling dates, the number of samples analyzed and the average PM_{10} concentrations with standard deviations (SD).

Location	Sampling period	Number of analysed samples	PM10 (µg m^{-3}) average (SD)
BUD	21.01–10.02.2019	10	58.6 (10.8)
	01.07–07.07.2019	7	22.9 (3.8)
	07.01–29.01.2020	21	47.4 (16.7)
KPS	21.01–10.02.2019	10	41.1 (4.7)
	08.07–14.07.2019	7	10.0 (2.3)
	07.01–27.01.2020	21	39.0 (14.1)
MSK	14.01–03.02.2019	10	64.6 (12.6)
	24.06–30.06.2019	7	23.5 (5.5)
	07.01–27.01.2020	21	53.6 (16.3)
PUT	14.01–03.02.2019	10	81.0 (14.8)
	24.06–30.06.2019	7	17.9 (4.7)
	07.01–27.01.2020	21	55.8 (24.0)
VES	28.01–17.02.2019	10	34.8 (8.1)
	08.07–14.07.2019	7	10.5 (1.8)
	07.01–27.01.2020	21	32.8 (10.6)
BUC-R	22.01-11.02.2019	10	52.1 (8.7)
	19.06-25.06.2019	7	32.4 (3.6)
	06.02-26.02.2020	19	36.5 (16.9)
BUC-M	06.02-26.02.2020	21	34.4 (18.1)
CLJ	26.01-15.02.2019	10	52.9 (11.9)
	19.06-25.06.2019	7	17.2 (4.5)
	10.01-03.02.2020	21	39.9 (24.8)
DEV	30.01-19.02.2019	10	67.8 (11.0)
	02.07-08.07.2019	6	28.1 (16.0)
	08.01-28.01.2020	21	71.3 (24.3)
FOC	19.02-11.03.2019	10	61.2 (15.3)
	19.06-25.06.2019	7	28.0 (3.2)
	09.01-29.01.2020	20	49.5 (22.6)
Total: 359			

4.4 Tracer compound stability assessment in the atmosphere for waste burning

Selected waste types, including PET, PS, and LDF (these types of waste contain all the important tracers), were collected and subsequently incinerated on charcoal. The resulting smoke was captured by High-Volume sampler with a flow rate of 32 m^{3}h^{-1}. To capture PM_{10} particles on quartz filters, Whatman 150 mm filters were employed. In the aging experiment, filter spots with a diameter of 2.4 cm from each waste type were prepared, as well as their corresponding blanks. Then they were placed into spot holders created from the filters (Fig. 6). These were then subjected to aging for different durations: 1 hour, 3 hours, 9 hours, 24 hours, 3 days, and 7 days. This aging experiment was conducted in winter, starting from the 16th of February and finishing on the 4th of March. Throughout this process, the sampler's flow rate was maintained at 16.5 lpm. The samples were categorized into three groups: blanks (the blank sample is a filter with the ambient PM_{10} during the different aging/sampling times), aged samples, and original samples (which were promptly refrigerated).

Figure 6. Sample holder

4.5 GC-MS analysis

To determine the concentration of organic tracer compounds in the aerosol samples, GC-MS was used. In the laboratory experiment, the process involved spiking a known area of the exposed filter with a recovery standard solution (p-terphenyl-d14, from Sigma-Aldrich) of 150 µL, which was then dried using a nitrogen stream. The samples were then extracted in three steps with 2 × 4 mL and 1 × 3 mL dichloromethane – methanol mixture (2 : 1 v/v) using a Vortex agitator (3 × 15 min, 750 min^{-1}). The extracts were filtered using syringe filters (Millex-LCR, hydrophilic PTFE membrane, 0.45 µm pore size, 13 mm diameter, Millipore) and then dried gently using nitrogen in 2 mL vials. To measure non-polar tracer components using GC-MS, the extracts were redissolved by adding a 100 µL internal standard solution (chrysene-d12, 98 atom% D, from Sigma-Aldrich) and 150 μL of a dichloromethane-methanol mixture (2 : 1 v/v). Then, approximately 1 μL of the samples was manually injected into the injector (operating at a temperature of 300°C in splitless mode) of an Agilent 6890N gas chromatograph that was coupled to an Agilent 5973N mass spectrometer. Separation of the components was carried out using a capillary column (DB-5MS UI, with dimensions of 30 m × 0.25 mm × 0.25 μm, from Agilent). The temperature program used in the experiment was adopted from Simoneit et al. (2005). First, the samples were measured in SCAN mode to determine retention times and characteristic ions, then in SIM mode to determine their concentrations. After the measurements, a recovery standard (sedoheptulose anhydride monohydrate, from Sigma-Aldrich) was added to selected samples, and after drying, they were derivatized by adding 100 μL of BSTFA-TMCS (N,O-bis(trimethylsilyl) trifluoroacetamide-trimethylchlorosilane, with a ratio of 99:1, from Sigma-Aldrich) and 100 μL of anhydrous pyridine (from Merck) at 80°C for 1 hour. Approximately 1 μL of the derivatized samples was manually injected into the injector (which was set to a temperature of 280°C in splitless mode) of GC-MS. The same capillary column that was used for analyzing non-polar components was also used for separation during the measurements. The column was initially heated to 60°C for 1 minute, and then the temperature was increased to 300°C with a temperature rate of 10°C per minute, and maintained at this temperature for 5 more minutes. SIM mode was utilized for quantitative analysis, and the instrument was calibrated using available standards such as p-terphenyl, 135-TPB, melamine, 2,4,6-triphenyl-1-hexen (SSS), and terephthalic acid. Based on the recoveries and repeated measurements, the analytical measurements were estimated to have an uncertainty of approximately 20%. The quantities of o-terphenyls and m-terphenyls were expressed in the amount of p-terphenyl. The amount of 1,2,4-triphenyl benzene, quaterphenyls, and 2-(benzoyloxy)ethyl vinyl terephthalate (2-BEVT) were expressed in the amount of 135-TPB, while the quantities of the ABS pyrolysis compounds (2-methylene-4-phenylheptanedinitrile (ASA), 2-methylene-4,6-diphenylhexanenitrile (ASS), 4,6-diphenylhept-6-enenitrile (SSA), and 2-phenethyl-4-phenylpent-4-enenitrile (SAS)) were expressed in SSS.

All types of samples in this thesis underwent a similar GC-MS analysis, however, the extraction methods for the cases where the concentration was variable were different. From the ambient samples 18.1 cm^2 of the filters were extracted 3 times in dichloromethane-methanol 2:1 mixture. A recovery standard (p-terphenyl-d14) was added to the samples before the extraction. During the first extraction step, 7.5 ml of dichloromethane: methanol (2:1) mixture was applied to the filter portion and the sample was shaken in a vortex agitator for 1 h. The extracts with the second and third volumes were 6 and 5 ml, respectively. The combined extracts were filtered with a syringe filter (0.45 μm) and dried under N_2 stream. The re-dissolved (in dichloromethane: methanol 2:1 mixture) sample extracts were then directly analyzed for the less polar tracer compounds, while the more polar compounds were measured after the derivatization by BSTFA-TMCS as in the case of laboratory samples. Blank samples were analyzed as well and the LOQ values were calculated as the average of the blanks + 10 times the standard deviation of the blanks. The LOQ values were obtained separately for the 2019 winter and 2020 winter campaigns as well as for the summer campaign and treated separately for the Hungarian and Romanian samples.

Regarding the aging samples, a recovery standard of 100 μl was added to each sample. In the first extraction step 5 ml of DCM: MeOH was applied, then subjected to the Vortex for 15 min. The mixture in the second and third steps were 5 and 3 ml, respectively, followed by the Vortex of 15 min. After these steps, the extracts were filtered and dried under the N_2 flow. Then the samples were measured directly and after derivatization the same way as in the cases above.

5. Results and discussion

5.1 Emission factors of previously identified waste burning tracer components and their isomers

Triphenylbenzenes and quaterphenyls are characterized by four aromatic rings connected via C–C bonds, forming non-condensed structures. Their presence in chromatograms can be distinguished by the prominent molecular ion at $m/z = 306$. The identification of quaterphenyls was achieved by comparing their mass spectra and calculated retention indices with data from the literature reported by Tsuge et al. (2011). For example, the calculated retention index of m,p-QTPH was 476.5 (Lee RI), aligning with the one obtained from the literature, 476.3. The retention indices of other compounds, like o-TPH, m-TPH and p-TPH were also calculated, 316.5, 357.6 and 363.2 accordingly with the results yielded from Marynowski et al. (2004), 317.42, 356.74 and 362.4, respectively. The presence of 135-TPB, 124-TPB, and p-QTPH was confirmed using authentic standards. The mass fractions of 1,3,5-triphenylbenzene were determined for the combustion of various waste types (as shown in Figure 7). The highest emission factors were observed for materials containing aromatic rings within their polymeric chains, with values of 800 and 100 $\mu g\ g^{-1}$ PM for PS and PET, respectively. However, for the burning of PAP, LDF, and ABS, its emission remains notable, averaging between 23 and 75 $\mu g\ g^{-1}$ PM. This aligns with findings by Jayarathne et al. (2018) who reported similar results (12–51 $\mu g\ g^{-1}$ PM) for open burning of mixed waste comprising cardboard, chip bags, food waste, paper, plastic bags, clothes, diapers, and rubber shoes. Tomsej et al. (2018) investigated the emission factor of 135-TPB during the co-combustion of PE plastics (shopping bags) and PET bottles with wood (beech logs) in a 20 kW boiler. The mass fraction of 135-TPB for mixed PET burning was 13 $\mu g\ g^{-1}$ PM, while for PE burning, it was 3 $\mu g\ g^{-1}$ PM.

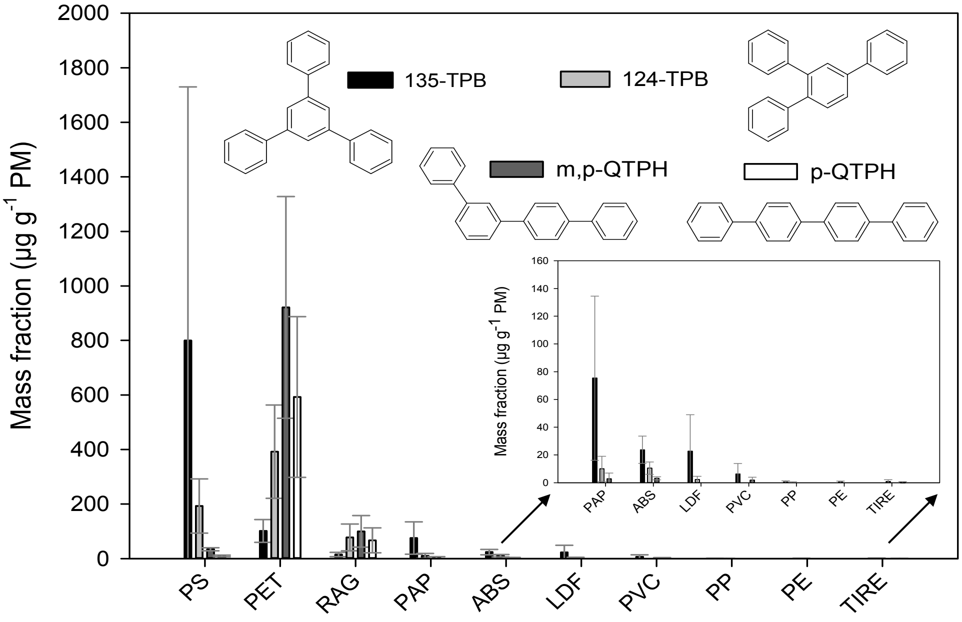

Figure 7. The mass fraction (in units of µg g^{-1} PM) of triphenylbenzenes and quaterphenyls from the burning of different waste types. The error bars represent the standard deviation of the EF data obtained for a given type of waste under different burning conditions.

Quaterphenyls were predominantly emitted during the combustion of waste materials containing PET. Their emission factor ranged from 33 to 1200 µg g^{-1} PM in individual samples, with an average relative EF of 920 and 100 µg g^{-1} PM for m,p-quaterphenyl, and 590 and 67 µg g^{-1} PM for p-quaterphenyl in PET and RAG, respectively (Fig. 7).

Similar to terphenyls (containing three non-condensed aromatic rings), the ratio of quaterphenyls to triphenylbenzenes (containing four non-condensed aromatic rings in linear and branched structures, respectively) can be employed as an indicator for different types of plastic burning. The average ratio of m, p-quaterphenyl (m, p-QTPH) to 124-TPB was below 1 for ABS, PS, and PAP (0.3, 0.2, and 0.2, respectively). In the case of samples collected during LDF burning, which also indicated the presence of styrene-containing materials, only 124-TPB was measured in higher quantities than the limit of quantification in three out of five samples. In samples collected during PET and

RAG burning, the average ratios of these two components were 2.3 and 1.3, respectively.

5.2 Identification of potential new tracers for waste burning and their mass fractions in the emitted PM_{10}

5.2.1 Terphenyls

The three isomeric forms of terphenyls (ortho-, meta-, and para-) can be readily distinguished due to their distinct molecular ions at m/z=230 and the presence of a doubly charged ion at m/z=115. Terphenyls have been identified in the particulate matter resulting from the incineration of various plastic waste types (Hoffer et al., 2021). Notably, they are more prevalent in plastics that inherently contain aromatic structures, such as PET, PS, and ABS, as illustrated in Figure 8. Additionally, samples from textiles (RAG) contained polyester (PET) alongside cotton and polyamide, contributing to significantly higher emission factors for terphenyls during their combustion (with values of 14, 610, and 920 $\mu g\ g^{-1}$ PM for ortho-, meta-, and para-terphenyl, respectively).

The emission factors for terphenyls were observed to be most pronounced during the combustion of PET bottles (190, 5400, 7700 $\mu g\ g^{-1}$ PM for ortho-, meta-, and para-isomers, respectively). In contrast, the combustion of PS and ABS yielded lower emission factors (ranging from 18 to 56, 250 to 1100, and 100 to 690 $\mu g\ g^{-1}$ PM for the ortho-, meta-, and para-isomers, respectively). It is noteworthy that terphenyls have not been commonly reported in particulates resulting from wood combustion, except for instances of liquid wood smoke (as noted by Guillén et al., 2000). However, terphenyls have been positively identified in particulates from one sample of lignite combustion (as reported by Fabbri et al., 2009).

It is important to note that the mass of PM_{10} measured gravimetrically and the amount of the compound obtained by GC-MS were used to calculate the relative EF ($\mu g\ g^{-1}$ PM) of the given chemical, while the absolute EF ($mg\ kg^{-1}$) for a given compound was calculated by taking into account the dilution factor in the sampling line and the mass of the waste burned (Hoffer et al., 2020). The absolute emissions factors for o, m, p-TPH were observed to be higher in the case of PET burning as well (2.1, 61, 87 mg kg^{-1}, respectively) and lower absolute EF in the case of PS and ABS burning (3-1.5, 57-21, 35-8.7 mg kg^{-1} for o-TPH, m-TPH, p-TPH, respectively).

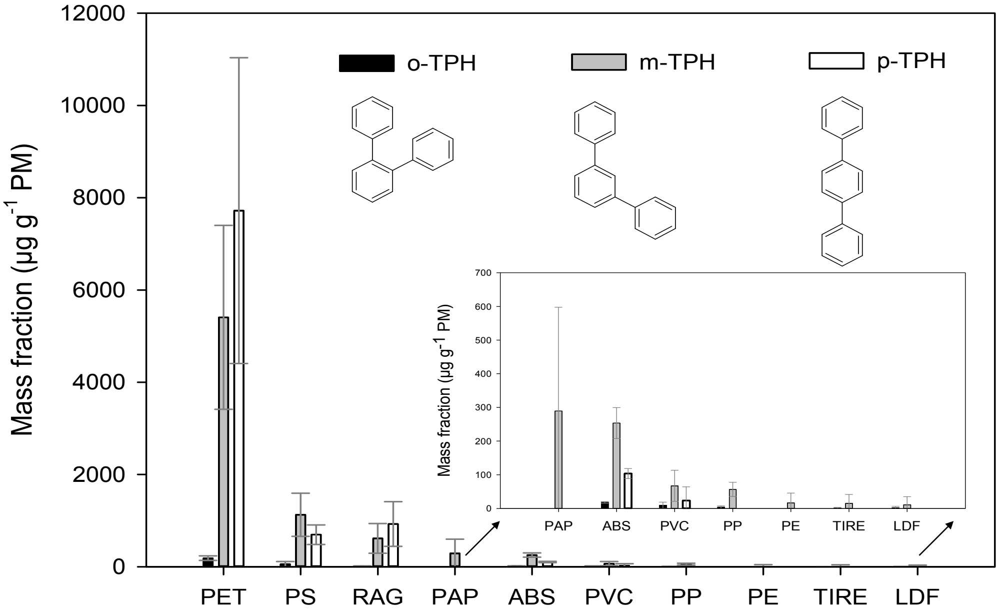

Figure 8. The mass fraction of terphenyls (in units of μg g^{-1} PM) in particulates from the burning of different waste types. The error bars represent the standard deviation of the mass fractions data obtained for a given type of waste under different burning conditions.

Furthermore, I should highlight that the position of the aromatic ring in the polymer structure may influence the specific terphenyls formed during combustion. This effect is also observed in the case of quaterphenyls (as described below). Our findings clearly indicate that p-terphenyl is emitted in the highest quantity during the burning of PET. This distinction arises from the para-position bonding of the aromatic ring in the PET polymer chain (positions 1-4), while in other polymers like ABS and PS, it is bonded through a single C-C bond to the polymer chain. Given the low emission of the ortho-isomer during combustion, the ratio of meta- to para-isomers can be used as an indicator of the predominant type of plastics being incinerated. In the context of PET and RAG combustion, the concentration ratio of emitted p-terphenyl to m-terphenyl varied between 1.1 and 1.7 (with an average of 1.5), whereas this ratio was

0.5–0.8 (averaging at 0.6) and 0.4 for PS and ABS, respectively. For other types of waste, only the meta-isomer was identified, and the concentration of p-terphenyl fell below the quantification limit, resulting in lower p-terphenyl to m-terphenyl ratios in ambient samples affected by the combustion of PS and/or ABS.

Given that the contribution of other sources to the concentrations of terphenyls in atmospheric particulates is minimal, terphenyls may serve as valuable atmospheric tracers for identifying plastic waste combustion, particularly involving PET, PS, and ABS. Additionally, it's important to note that the relative emission factors for terphenyls during wood and wood-based waste combustion (specifically OSB and LDF) were below the quantification limit.

5.2.2 2-(Benzoyloxy)ethyl vinyl terephthalate (2-BEVT)

The pyrolysis of PET polymer produces various compounds, among which 2-(benzoyloxy)ethyl vinyl terephthalate was consistently detected in most PET containing samples (PET and RAG). The identification of this particular compound relied on comparing the mass spectra and retention index. The calculated retention index was 2615.7 which was consistent with the literature's result 2636, reported by Tsuge et al. (2011). Notably, the emission factor for this compound was exceptionally elevated, reaching 1000 μg g^{-1} PM in one PET burning experiment characterized by the lowest average flue gas temperature (measured at approximately 185°C, 50 cm above the stove outlet) during the initial burning of waste pieces.

Conversely, this compound was not present in PET samples collected under conditions involving a high air supply ratio and elevated flue gas temperatures (around 280°C). This clear distinction highlights that the formation of this compound occurs during pyrolysis, and consequently, its emission factor is strongly influenced by the specific burning conditions. In another PET sample, where the air supply was reduced but the temperature remained higher, the emission factor of 2-(benzoyloxy)ethyl vinyl terephthalate was two orders of magnitude lower, measuring at 4.2 μg g^{-1} PM. On average, the relative emission factor of 2-(benzoyloxy)ethyl vinyl terephthalate for PET bottles was determined to be 250 µg g^{-1} PM. The absolute emission factor of 2BEVT, obtained during the PET burning, was 2.8 mg kg^{-1}.

Interestingly, this compound was identified in all three RAG samples, with its emission factor in the RAG samples showing variability linked to the adjustments in air supply and/or the temperature during combustion, the smaller the air intake the higher the EFs. The emission factor of this compound in the RAG samples ranged from 0.36 to 16 µg g^{-1} PM, with an average of 6.9 µg g^{-1} PM.

5.2.3 2,4,6-Triphenyl-1-hexen (styrene trimer, SSS)

The compound known as 2,4,6-triphenyl-1-hexen, also referred to as styrene trimer (SSS), has been successfully identified and quantified in filter samples through GC-MS analysis employing an authentic standard from Wako Chemicals. The calculated retention index was 2444, which was compared with the value of 2484 from the literature (Tsuge et al., 2011). The presence of this compound was verified by the co-eluting fragment ions of 91, 117, 194, and 207. Tsuge et al. (2011) previously detected SSS using direct pyrolysis gas chromatography mass spectrometry (Py-GC-MS) in materials containing styrene, including polystyrene (PS) and acrylonitrile butadiene styrene (ABS), along with other styrene-containing substances such as styrene–methyl acrylate copolymer, styrene–maleic anhydride copolymer, and styrene–divinylbenzene copolymer.

In the experiments, I primarily identified the styrene trimer in particulate matter resulting from the combustion of polystyrene, with its mass fraction varying between 56 and 1400 µg g^{-1} PM depending on combustion conditions, averaging at 2900 µg g^{-1} PM. SSS was also detected in particulate matter from the combustion of low-density fiberboard (LDF) with relative emission factors (EFs) ranging from below the limit of quantification to 1400 µg g^{-1} PM (with an average of 380 µg g^{-1} PM) (Fig. 9). In the case of paper burning, the EFs were measured at 97 and 670 µg g^{-1} PM, with waxy, coated papers yielding higher relative EFs for this compound. The Py-GC-MS investigation conducted by Tsuge et al. (2011) on ABS revealed minimal quantities of

SSS, similarly to our findings where the combustion of ABS samples resulted in a relative EF of SSS below the limit of quantification. The absolute EF of this compound was the highest (150 mg kg^{-1}) during the combustion of the PS containing waste.

It's important to note that traces of SSS were detected in nearly all of the filter samples, including blank samples, suggesting that it might be a widespread contaminant. Thus, precautions should be taken when determining and quantifying its presence.

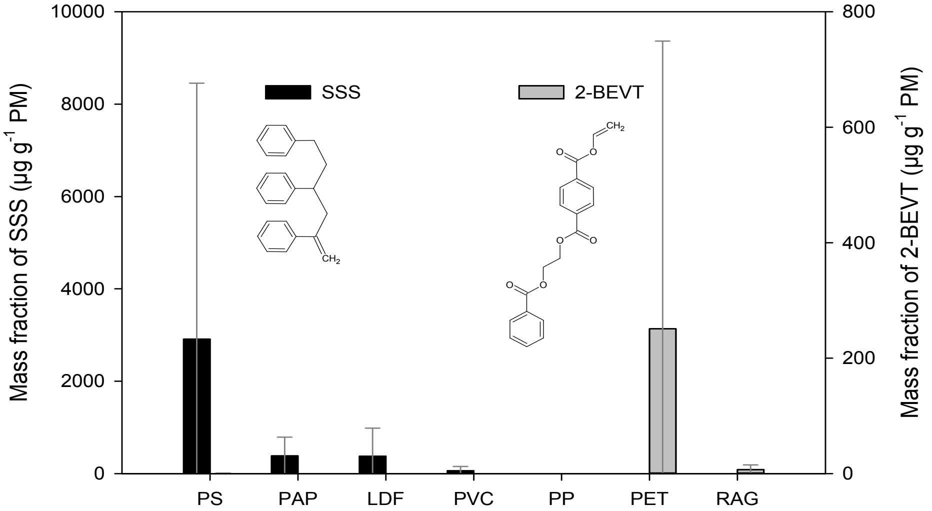

Figure 9. The mass fraction (in units of µg g^{-1} PM) of styrene trimer (SSS) and that of the 2-(benzoyloxy)ethyl vinyl terephthalate (2-BEVT) from the burning of different waste types. The error bars represent the standard deviation of the EF data obtained for a given type of waste under different burning conditions.

5.2.4 Melamine

Melamine ($C_3H_6N_6$, heterocyclic aromatic compound) is the component in melamine-formaldehyde resin, primarily utilized in the furniture industry for surface coating on fiberboard panels. Additionally, this resin can be found in laminate flooring and various objects, for instance, kitchen tools. A direct Py-GC-MS study conducted by Tsuge et al. (2011) confirmed the production of melamine during the pyrolysis of melamine-formaldehyde resins.

Interestingly, melamine was predominantly identified in particulates resulting from the combustion of low-density fiberboard (LDF) samples, displaying relative emission factors (EFs) ranging from approximately 50 to around 41,000 μg g^{-1} PM, with an average of approximately 19,000 μg g^{-1} PM. In contrast, melamine levels in all other samples examined were comparable to those found in blank samples and, in some cases, fell below the detection limit. Moreover, the burning of LDF samples yielded 51 mg kg^{-1} as an absolute EF. Nevertheless, if detected, melamine could still serve as a useful particulate tracer for identifying localized waste burning events, particularly during the winter months. However, its quantitative application may be significantly limited by the substantial variation in the melamine-formaldehyde resin content present in commercially available LDF products.

5.2.5 Specific tracers for burning of styrene copolymers

One of the noteworthy styrene copolymers is known as ABS (acrylonitrile butadiene styrene), produced through the copolymerization of acrylonitrile, butadiene, and styrene. ABS finds extensive application in the automotive industry, and it's also commonly used for housing electronic equipment and the edge foil of furniture panels, particularly low-density fiberboard (LDF). In the direct pyrolysis of samples containing ABS, I identified several hybrid trimer compounds composed of styrene and acrylonitrile units, relying on mass spectra and retention index data as reported by Tsuge et al. (2011). These compounds include ASA, ASS, SSA, and SAS, all of which have been detected in samples of ABS, LDF, and PAP (see Figure 10). The calculated retention indexes of which were 1822.6 (ASA), 2101.5 (ASS), 2144.7 (SSA), 2171 (SAS) in accordance with the results from Tsuge et al. (2011) 1843, 2129, 2175 and 2200, respectively.

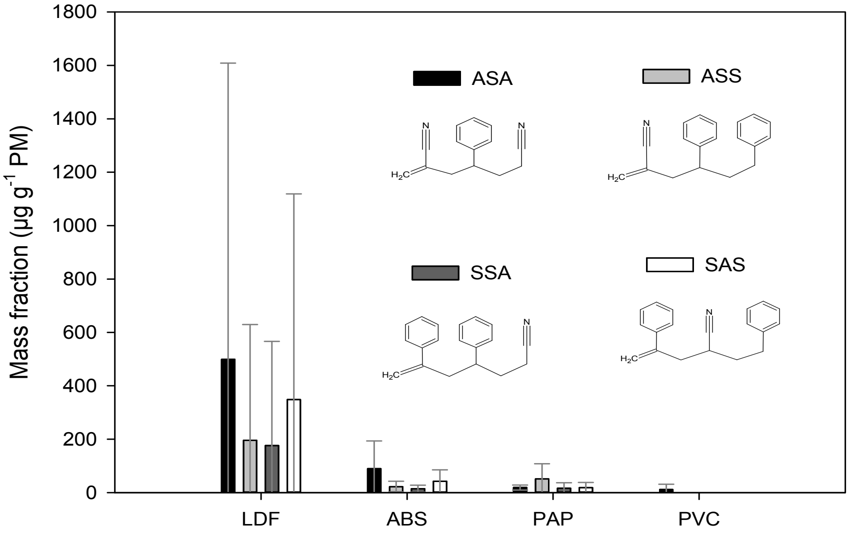

Figure 10. The mass fraction of the compounds (in units of µg g^{-1} PM) emitted during the burning of styrene-containing copolymers. The error bars represent the standard deviation of the EF data obtained for a given type of waste under different burning conditions.

Within the samples collected during ABS burning, the EF of ASA was smallest in the sample collected with high air intake (its amount was below the limit of quantification). However, on average, the EF of ASA was the highest in the case of ABS burning (as depicted in Figure 10). The absolute emission factors of SSA, ASS, SAS and ASA were 1.4, 2.2, 4.2, and 9.2 mg kg^{-1}, respectively. In the LDF samples, the concentration of ABS pyrolysis products displayed considerable variability. In one sample (collected with a low air intake setting), they were below the limit of quantification, whereas in another LDF sample, their relative EFs ranked among the highest, ranging from about 870 to 2500 µg g^{-1} PM. This significant variation in the EF of ABS pyrolysis products in LDF samples is attributed to the heterogeneous composition and ABS content of the combusted materials. Similar to ABS in the case of LDF combustion, the relative EF of ASA was the highest, followed by SAS (Fig. 10). The absolute emission factors were lower during the LDF burning, ranging from 0.6-1.7 mg kg^{-1}.

In samples from paper burning, the EF of ABS pyrolysis products was relatively low, with individual component levels varying between about 3 and 91 µg g^{-1} PM. The absolute EFs of ABS products during the paper burning were in the range of 0.036-0.11

mg kg^{-1}, with the highest value obtained for ASS. In the paper industry, sizing plays a crucial role in altering the surface properties of papers. Sizing agents often contain styrene, such as the common styrene–butyl acrylate copolymer used as a surface sizing agent (Hagiopol and Johnston, 2012). Additionally, binders used in the paper industry to bind coating pigments together and to the base paper surface consist of emulsions of polymers that are copolymers of various monomers, including styrene, butadiene, acrylic esters, vinyl acetate, and acrylonitrile (Bajpai, 2015). This could explain the presence of these compounds in the paper samples.

5.3 Estimated vapor pressures of the newly suggested waste burning tracers

The vapor pressures of the compounds were estimated using US EPA (2012). The estimated vapor pressures of the identified compounds are in the range between 5.16×10^{-9} Pa at 25°C and 2.187×10^{-3} Pa at 25°C; the latter is lower than that for the syringic aldehyde, for example (8.65×10^{-3} Pa at 25°C), suggested as a biomass burning tracer by Simoneit et al. (2002), and is comparable with that of terephthalic acid (1.5878×10^{-3} Pa at 25°C). It also should be noted here that the estimated vapor pressures of SSS and 2-BEVT (4.453×10^{-5} Pa at 25°C and 1.0013×10^{-4} Pa at 25°C, respectively) are very close to that of levoglucosan (4.6235×10^{-5} Pa at 25°C), the most widely used biomass burning tracer compound (Fig. 11).

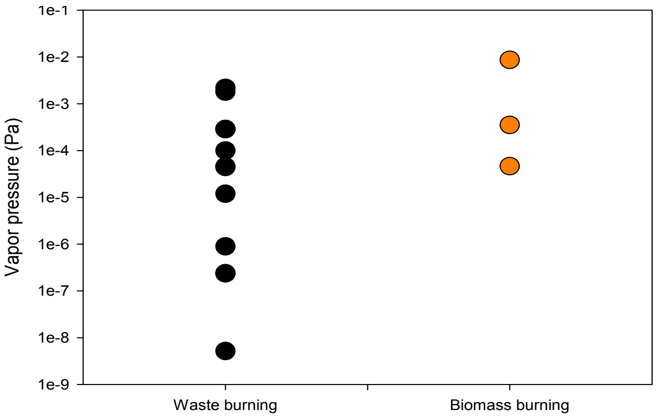

Figure 11. The estimated vapor pressure of compounds from waste and biomass burning (Pa).

5.4 Waste burning practices based on tracer compounds in Hungary and Romania

5.4.1 Burning of waste containing PET

The combustion of PET-containing waste, mainly originating from PET bottles and polyester textiles, can be distinguished by several compounds. However, the most distinctive among these compounds, occurring in larger quantities, is 2-BEVT. In Figure 12, I present the average ratio of 2-BEVT to levoglucosan (LGS) for winter samples gathered from various sampling locations in Hungary and Romania. This ratio serves as a means to assess the prevalence of PET burning relative to firewood burning in different settlements. In some cases, certain samples had 2-BEVT concentrations that fell below the quantification limit (LOQ), and these were treated as zero during the calculation of averages. The numbers contained within the bars indicate the percentage of 2-BEVT values exceeding the LOQ (with LGS concentrations having 100% data coverage), while the error bars represent the standard deviation of the data.

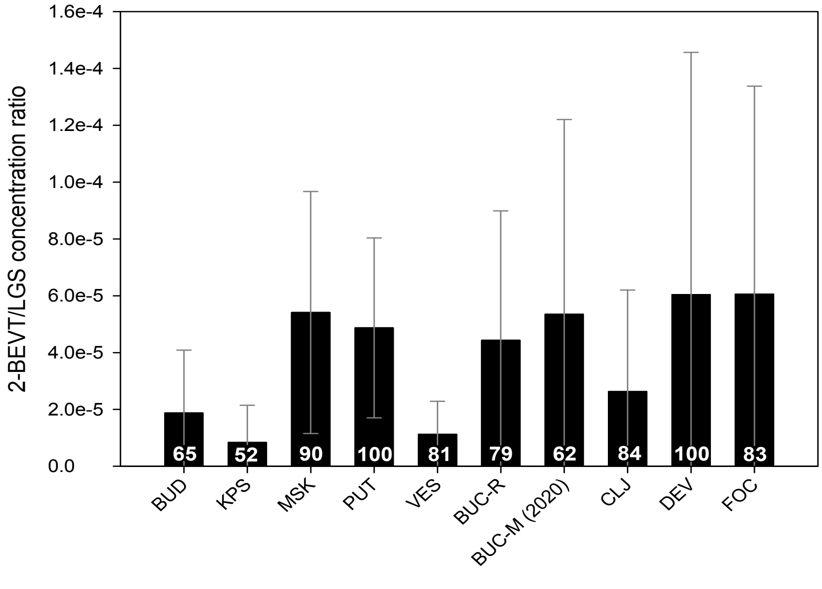

Figure 12. The average mass concentration ratios of 2-BEVT to LGS in the PM_{10} samples collected at the different 160 locations in the winters of 2019 and 2020. The numbers shown inside the bars represent the percentage of cases in which the concentrations of 2-BEVT were above the limit of quantification. The error bars represent the standard deviation of all data.

In Hungary, the predominant occurrence of PET burning is most pronounced at sampling sites situated in the northeastern part of the country, particularly in MSK and

PUT. Within Putnok, 2-BEVT was identified in all analyzed samples, whereas in K-puszta, it was detected in only half of the collected samples. This observation is further corroborated by the fact that levoglucosan concentrations were also highest in Putnok (Fig. 13). Consequently, this outcome confirms that the absolute concentrations of 2-BEVT were notably highest in Putnok. On average, the measured levels of 2-BEVT in Putnok were 4.6 times greater than those in Budapest and a remarkable 14.7 times higher than those in Veszprém. This significant difference exceeds the expected impact of generally elevated PM_{10} concentrations in Putnok (see Table 4).

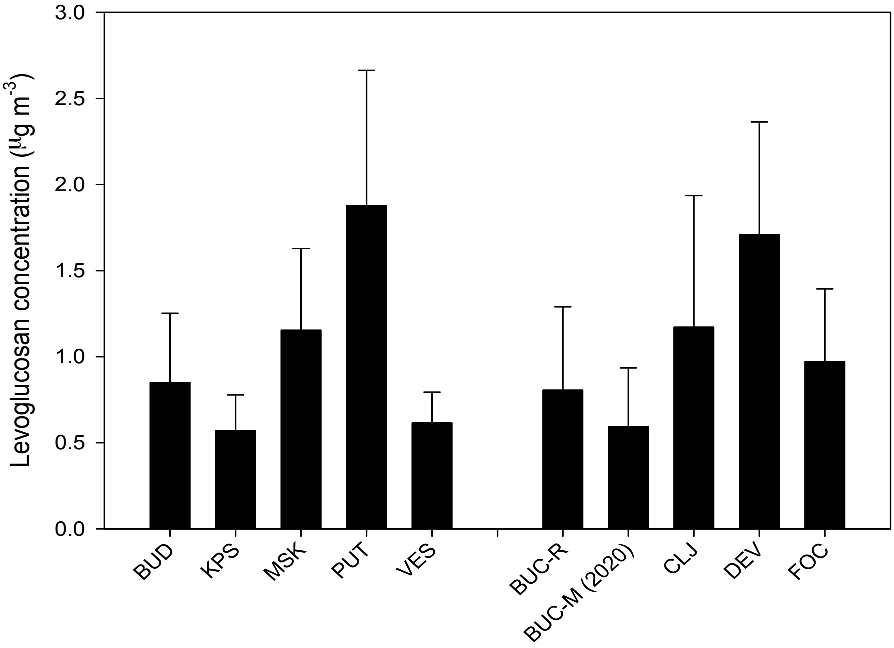

Figure 13. Average concentration of levoglucosan measured at the sampling locations in the heating seasons. The error bars represent the standard deviation of all data.

It's worth noting that a portion of the levoglucosan in Putnok is likely attributed to lignite burning, as this solid fuel is extensively used in the region. As demonstrated by Fabbri et al. (2009), the emission factor of levoglucosan from lignite burning can even surpass that from wood burning. Consequently, the ratio of 2-BEVT (and other compounds discussed later) to levoglucosan in Putnok is somewhat lower than it would have been if wood burning were the sole source of levoglucosan.

In Veszprém, the significance of PET burning is relatively low, despite a high rate of confirmed tracer detection (approximately 80%). Conversely, in Budapest, the relative prevalence of PET burning appears to be higher than in Veszprém, despite the fact that 2-BEVT was identified in only about two-thirds of the samples collected.

Regarding the Romanian sampling sites, the prevalence of PET burning generally aligns with that observed in the polluted Hungarian sites, with the exception of Cluj-Napoca. In Bucharest, 2-BEVT was detected in approximately 70% of the samples, and the concentration ratio to LGS was among the highest, suggesting an intensive incineration of PET-containing waste in the vicinity. Deva and Focsany were the sites most affected by PET burning during the study period, with 2-BEVT being identified in all samples collected in Deva and in 84% of samples from Focsany. In Cluj-Napoca, the proportion of PET burning is relatively low but still surpasses that observed in Budapest, with data coverage similar to Focsany. The ratio of 2-BEVT to LGS in most sampling locations remained consistent, with only a slight decrease of about 20% between the two heating seasons. However, in Cluj-Napoca, Focsany, and K-Puszta, higher ratios were observed in 2020 than in the heating season of 2019, indicating potential variations in local meteorological conditions and sources. During the summer sampling, 2-BEVT concentrations were primarily below the quantification limit, except for one sample from Putnok.

I observed that the combustion of PET and textiles containing PET leads to the release of significant amounts of m-terphenyl and p-terphenyl. Biomass burning does not contribute substantially to these compounds, although they have been detected in lignite smoke. Fossil fuel combustion does not seem to be a source of these compounds, as their concentration remained below the quantification limit in the summer samples. It was also demonstrated that the incineration of PS, ABS, and paper (specifically, the meta isomer) serves as notable sources of terphenyls. Figure 14 displays the average concentration ratio of p-terphenyl to m-terphenyl, excluding a few outliers, across different sampling sites.

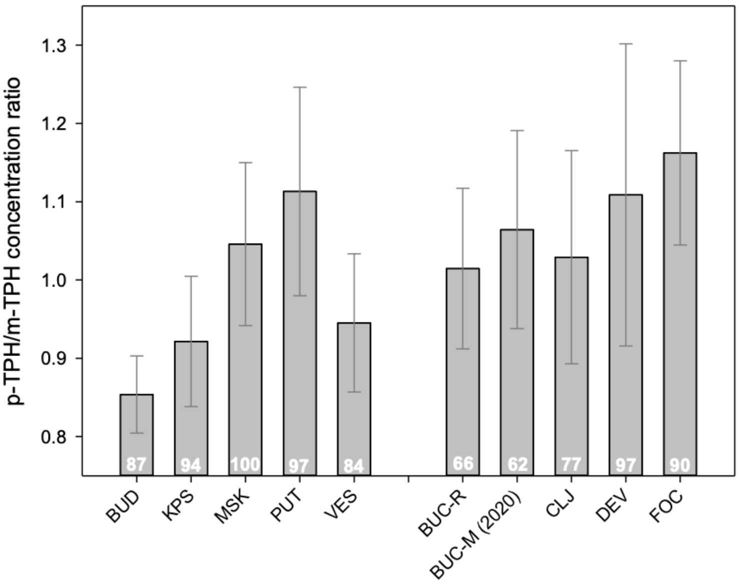

Figure 14. The average mass concentration ratio of p-TPH and m-TPH in the PM_{10} samples collected at the different sampling sites. The numbers shown inside the bars represent the percentage of cases in which the concentrations of both isomers were above the limit of quantification. The error bars represent the standard deviation of all data.

Although there isn't a strong correlation between the 2-BEVT/LGS ratio and the p-TPH/m-TPH ratio within samples collected at a specific location, Figure 12 clearly shows that in areas with a higher prevalence of PET burning (such as MSK, PUT, BUC-M, DEV, FOC), the ratios of the two terphenyl isomers exceeds 1. This suggests that PET burning significantly contributes to these compounds in the ambient aerosol, supporting the notion that these communities burn PET-containing waste in larger quantities. However, it's essential to acknowledge that other sources may also contribute to terphenyl concentrations, given that the p-terphenyl to m-terphenyl ratio is lower than what is typically observed for PET burning (1.1-1.7).

5.4.2 Burning of fiberboard, PS and paper

The primary source of styrene trimer (SSS) is the incomplete combustion of PS, but its emission factor is also noteworthy during the incineration of coated LDF, printed and coated waxy paper (Hoffer et al., 2021). Despite polystyrene (PS) emitting approximately two orders of magnitude more SSS compared to LDF or paper (PAP), the burning of these waste materials, likely common in households, significantly influences the atmospheric concentration of this constituent. Figure 15 provides an illustration of the concentration ratio of styrene trimer to levoglucosan (LGS) in the ambient samples collected in Hungary and Romania.

The concentration of SSS was observed to be the lowest at the KPS station in Hungary, most likely because it was only identified in half of the samples. In contrast, SSS was detected in all PM_{10} samples from Budapest and Putnok and in the majority of the samples of Miskolc and Veszprém.

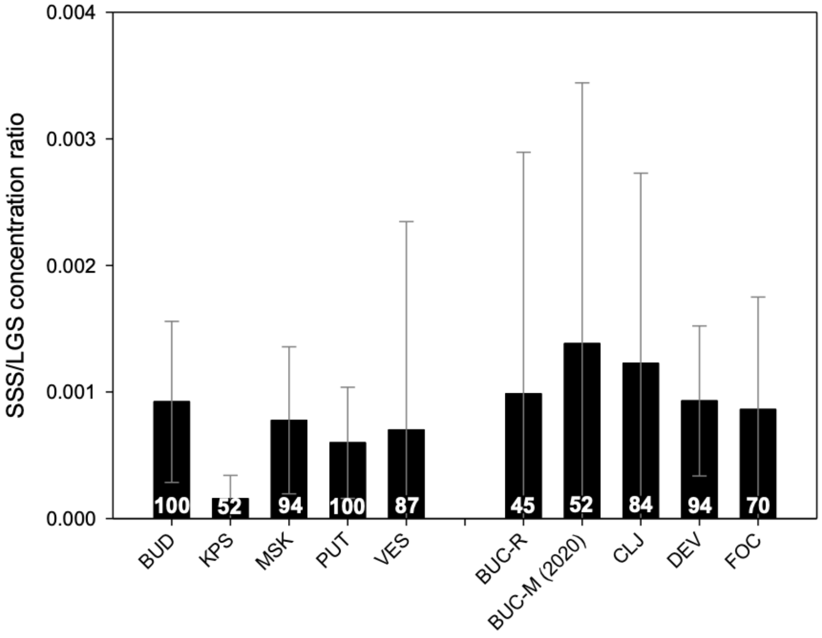

Figure 15. The average relative mass concentration ratio of styrene trimer (SSS) to levoglucosan in the PM_{10} samples at the different sampling sites. The numbers shown inside the bars represent the percentage of cases in which the concentrations of styrene trimer (SSS) were above the limit of quantification. The error bars represent the standard deviation of all data.

The SSS/LGS concentration ratios exhibited relatively consistent patterns at all of the sampling sites in Hungary, except for KPS. In Romania, the average SSS/LGS ratio was slightly elevated in most of the PM_{10} samples, except for FOC. Notably, the concentration of SSS in FOC displayed significant year-to-year variability, with concentrations above the LOQ detected in only 20% of the samples in 2019 but in 95% of the samples in 2020. SSS was identified in roughly half of the samples collected near

Bucharest (60% in BUC-R and 37% in 2019 and 2020, respectively), but the average SSS/LGS ratio was high. This suggests that waste burning in these areas may be intermittent or influenced by specific wind directions. Interestingly, larger cities exhibited higher SSS/LGS ratios compared to smaller settlements in both countries, which might indicate that waste types leading to SSS emissions during combustion are more readily available in larger cities per unit number of wood-burning households. During the summer season, SSS concentrations were consistently below the LOQ in both Romanian and Hungarian samples.

In the research, I identified 135-TPB in most of the PM_{10} samples collected during controlled burning experiments involving various waste types. This compound is recognized as a universal indicator of waste burning according to Simoneit et al. (2005). 1,3,5-triphenylbenzene was detected in nearly all of the samples, with data coverage averaging over 93% for the winter samples.

The highest average atmospheric concentrations of 135-TPB during the study period were observed in Bucharest and Cluj-Napoca, reaching 1.3 ng m^{-3} and 1.6 ng m^{-3}, respectively. These concentrations fall within the lower range of levels recorded for Beijing during the summer season of 2008 (1.58–4.58 ng m^{-3}, as reported by Li and Fang 2009) but are moderately higher than those measured in South Poland during the heating season of 2017 (0.8 ng m^{-3}, according to Furman et al., 2020). In Hungary, the average atmospheric concentrations of 135-TPB ranged from 0.25 to 0.42 ng m^{-3} across different sampling stations.

At the background station (KPS), the levels of compounds containing four aromatic rings (Fig. 16), such as quaterphenyls and triphenylbenzene isomers, are notably higher when compared to 2-BEVT and SSS. This observation suggests that these compounds have a longer atmospheric lifetime due to the absence of double bonds.

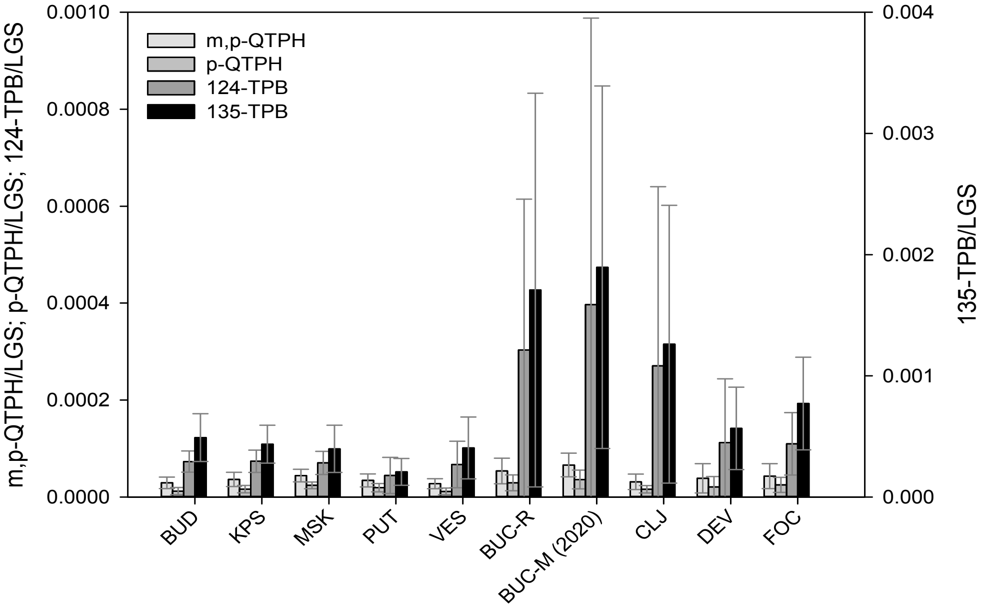

Figure 16. The average relative mass concentration ratio of quaterphenyls and triphenylbenzenes to levoglucosan in the PM_{10} samples at the different sampling sites. The error bars represent the standard deviation of all data. The percentage of cases in which the concentrations of the presented compounds were above the limit of quantification was larger than 87%.

In Hungary, the highest relative concentrations of quaterphenyls compared to levoglucosan (LGS) were observed in MSK, where PET burning is a common practice. In Romania, the highest quaterphenyl-to-LGS ratio was found in Bucharest, where substantial amounts of both SSS and 2-BEVT were also detected. Although the ratio of quaterphenyls to LGS is only slightly higher in Romanian samples, the relative amount of triphenylbenzenes to LGS is 2-3 times higher in Romania compared to Hungary. Triphenylbenzene concentrations were exceptionally elevated near Bucharest and in Cluj-Napoca, showing significant variability between individual samples, which implies the presence of strong and intermittent emission sources near these sampling stations. Li and Fang (2009) believed that 135-TPB emissions in Beijing could potentially originate from waste incineration plants and fossil fuel combustion. If this is the case, then 135-TPB may not serve as a reliable tracer for burning household waste. However, it might still be a general indicator of waste burning if the wide variation in the atmospheric concentration of triphenylbenzenes can be linked to significant variations in the kinds of solid waste that are burned (for example, the emission factor of triphenylbenzenes for PS burning is very large).

During the summer season, both quaterphenyls and triphenylbenzenes were detected in the samples, although their relative concentrations were lower compared to the winter samples (Fig. 17, 18). However, the ratio of 135-TPB to PM_{10} was comparable to, or in some cases even higher than, what was observed in the winter samples. This suggests that the sources of these compounds, such as polymer burning and industrial activities, continue to be active during the summertime. Previous research conducted in southern Poland during the non-heating season reported that the relative amount of 135-TPB to PM_{10} was approximately 60% of the value observed during the heating season. Additionally, it's worth noting that p-quaterphenyl has been identified as an emission from aircraft engines and has been proposed as a tracer for polycyclic aromatic hydrocarbon (PAH) pollution originating from airplanes (Krahl et al., 1998).

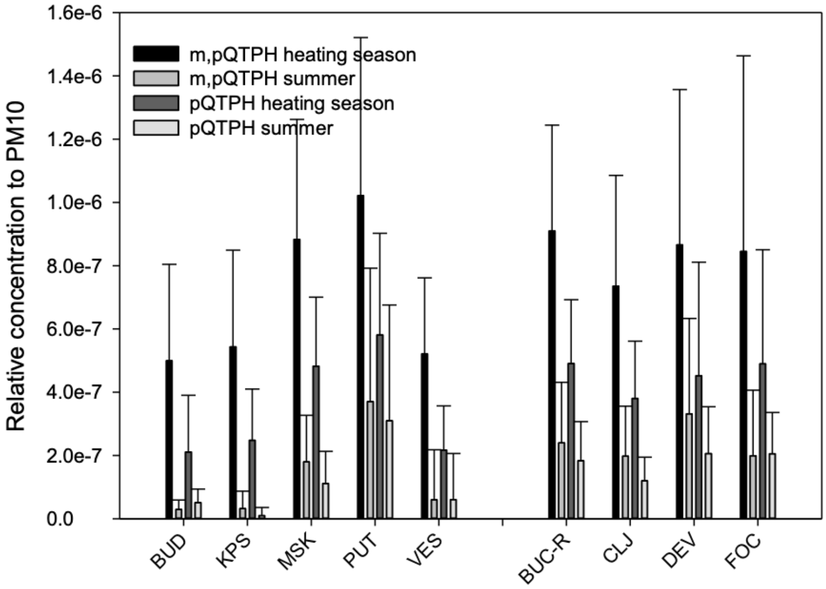

Figure 17. The relative concentration (concentration ratio) of quaterphenyls to PM_{10} in the samples collected in the heating season as well as in the summer samples. The error bars represent the standard deviation of all data.

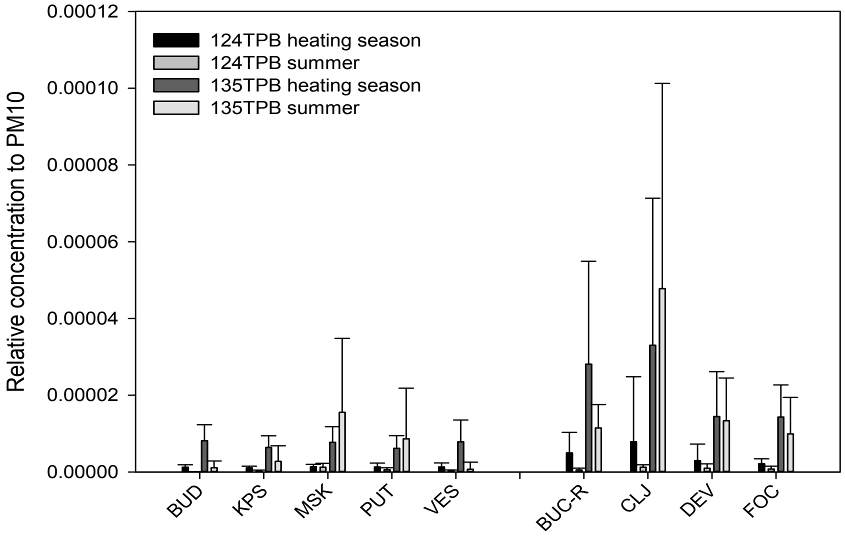

Figure 18. The relative concentration (concentration ratio) of triphenylbenzenes to PM_{10} in the samples collected in both seasons.

It was noted that the ratio of triphenylbenzenes to quaterphenyls showed significant variation depending on the type of plastic being burned. The burning of waste specimens containing styrene, such as PS, LDF, PAP, and ABS, had much higher concentrations of triphenylbenzenes compared to PET and RAG samples, where the concentration of quaterphenyls was somewhat higher.

Figure 19 demonstrates that the largest m,p-QTPH/124-TPB ratios were observed in MSK and PUT, implying that a larger quantity of PET-containing waste may be undergoing incineration near these stations. Conversely, lower m,p-QTPH/124-TPB ratios were found at all Romanian stations, coupled with elevated concentrations of SSS, indicating that a greater quantity of styrene-containing waste (such as PS/LDF/PAP) might be burned in the vicinity of the sampling sites. The higher m,p-QTPH/124-TPB concentration ratios found at DEV and FOC support an increased rate of PET burning (as indicated by the 2-BEVT/LGS ratio) at these locations. It is noteworthy that the m,p-QTPH/124-TPB concentration ratio remains below 1 in both countries, suggesting that the concentration of these components might be influenced by the incineration of LDF/PS/PAP waste in both regions.

The presence of SSS can originate from various sources, while the emission of melamine is primarily associated with the combustion of materials containing melamine-formaldehyde resins, such as coated fiberboards commonly used in the furniture industry. To assess the extent of furniture panel burning at different sampling sites, the melamine/LGS ratio is analyzed, as depicted in Figure 20. It's worth noting that during the winter of 2019, the concentration of melamine was below the limit of quantification (LOQ) in the PM_{10} samples from the winter 2019 in Hungary, except for one sample from Putnok. However, the compound was qualified in only half (43-57%) of the samples from 2020 in Hungary. In contrast, the data coverage was higher in the Romanian samples in both years (50-100%), indicating that the burning of furniture panels is more common in Romania compared to Hungary. Moreover, based on the data, it appears that the incineration of furniture panels is more frequent in capital cities than in other settlements, possibly due to the easy availability of discarded furniture.

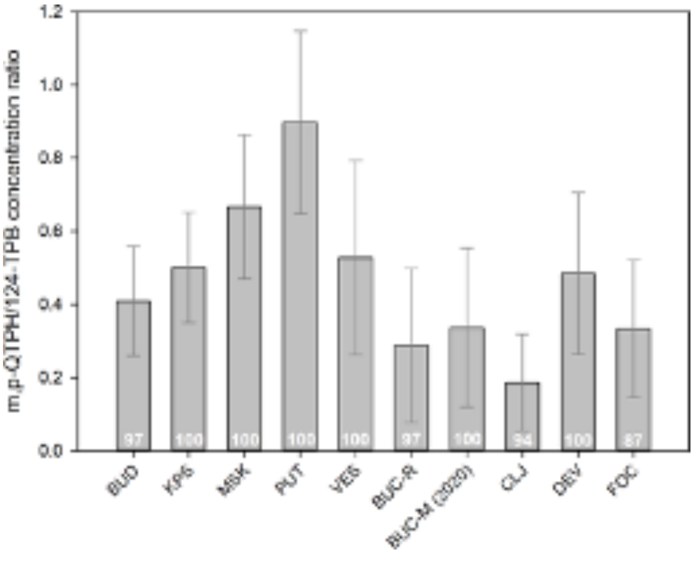

Figure 19. The average relative concentration ratio of m,p-QTPH to 124-TPH at the different sampling locations. The numbers shown inside the bars represent the percentage of cases in which the concentrations of both isomers were above the limit of quantification. The error bars represent the standard deviation of all data

During the summer samples, melamine was identified in certain samples from Hungary and Romania, yet its occurrence in PM_{10} concentrations was notably lower (on average 10% of that found in the winter samples). This discrepancy might be attributed to sporadic instances of furniture panel combustion or emissions originating from nearby furniture manufacturing and processing facilities.

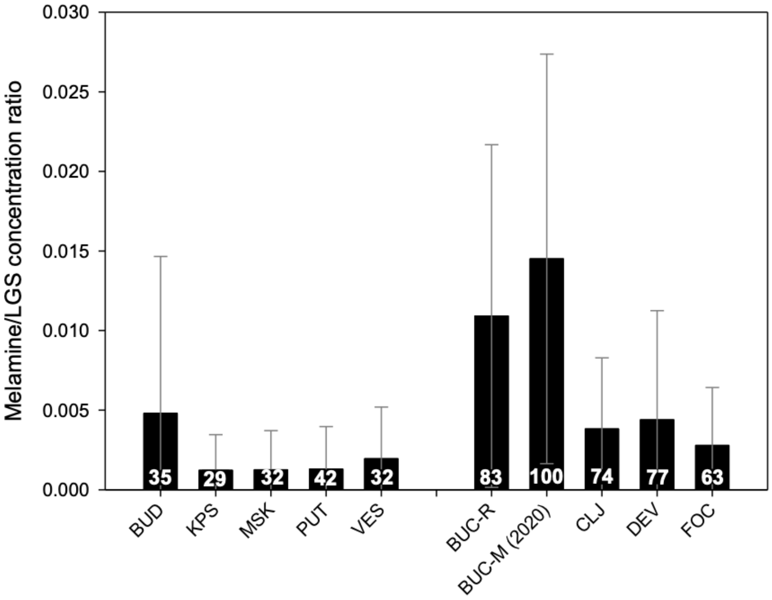

Figure 20. The relative concentration of melamine to levoglucosan in PM_{10} samples from the heating periods. The numbers shown inside the bars represent the percentage of cases in which the concentrations of melamine were above the limit of quantification. The error bars represent the standard deviation of all data.

In the winter months, the samples contained detectable levels of ABS pyrolysis products, specifically ASS, SAS and SSA (illustrated in Figure 21). These compounds are emitted not only during the incineration of electronic devices and household waste but also from the combustion of furniture panels, where they are employed as edge banding tapes for chipboards, medium-density fiberboard (MDF), or high-density fiberboard/hardboard (HDF). Similar to the case of melamine, the concentrations of these ABS pyrolysis products in relation to LGS were most elevated in samples collected from Budapest and Bucharest, suggesting that larger quantities of furniture panels are likely being incinerated in these areas.

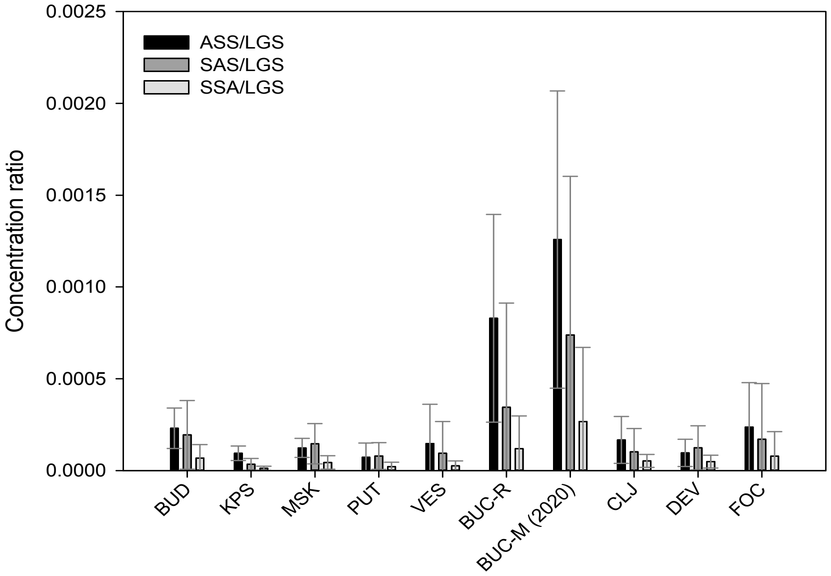

Figure 21. The concentration ratio of ABS pyrolysis products to LGS in PM_{10} samples from the heating seasons. The error bars represent the standard deviation of all data, the numbers above the error bars indicate the frequency of occurrence expressed in percentages.

5.5 Contribution of illegal waste burning to the atmospheric PM_{10} concentration.

The impact of burning waste from households on ambient PM_{10} mass concentrations was conducted at the sample locations in Romania and Hungary for the first time, based on measured atmospheric concentrations of quaterphenyls, 2-BEVT, SSS, ASS, SAS, SSA, and melamine. Both the absolute EFs (mg kg^{-1}) and the relative EFs (µg g^{-1} PM_{10}) of the tracer compounds, and the PM_{10} emission factor (on a relative scale) were used to calculate the average emission factor that was used in the estimation of the contribution of waste burning. As the emission factors of the tracer compounds depend on the type of burned material and a given marker compound may also be emitted from the burning of different waste types, a sensitivity analysis was assessed in order to determine the average emission factors for each tracer compound for various waste mixtures. (For the relative EFs, the quantity of PM_{10} released was determined and the amount of the tracer compound in the PM_{10} released for various waste mixtures; whilst for the absolute EFs, presented in Table 5, the amount of the released tracer compound was correlated with the weight of the various waste mixtures). At first, the average emission factor for each tracer component was calculated by assuming that on a mass basis, the burned household waste was composed of 52.6% furniture panels, 15.8% paper, 15.8% rag, and 15.8% plastics (the mass ratios of the waste types are 10:3:3:3, respectively). Plastic-type household waste was also assumed to consist of 42% PE, 28% PET, 14% PP and PS, 0.7% PVC, and ABS, based on the plastic composition of municipal waste (Bodzay and Bánhegyi, 2016). The relative amount of the various waste types and/or groups in the waste mixture was increased by ten when performing the calculations. For each of the 15 possible combinations, the weighted averages of the emission factors of the tracers were determined. EFs were also calculated for waste mixtures with varying mass percentages of furniture boards (10% to 91.7%) and rag, paper, and mixed plastics from household waste (1.8% to 65.2%).

Table 3. The estimated emission factors of different tracer compounds for waste mixtures.

		Relative EF (µg g^{-1}PM10)											
	m/m%	PM10 EF	135-TPB	124-TPB	m-TPH	p-TPH	m,p-QTPH	p-QTPH	2-BEVT	SSS	ASS	SAS	Mela-mine
ABS	0.013 - 0.46	38	24	10	250	100	3.2	0	0	0	22	43	0
LDF	10 - 92	2	23	2.2	11	0	0	0	0	380	200	350	19000
PAP	1.8 - 65	1	75	10	290	0	2.9	0	0	380	51	18	0
PE	0.78 - 28	9	0	0.43	17	0	0	0	0	0	0	0	0
PET	0.52 - 18	5	100	390	5400	7700	920	590	250	0	0	0	0
PP	0.26 - 9.2	16	0.49	0.21	57	0	0	0	0	0	0	0	0
PS	0.26 - 9.2	25	800	190	1100	690	34	9.7	0	2900	0	0	0
PVC	0.013 - 0.46	17	6.2	0	67	24	1.9	0	0	0	0	0	0
RAG	1.8 - 65	4	15	78	610	920	100	67	6.9	0	0	0	0
Mixture minimum			45	19	183	1106	96	79	15	491	98	136	19000
Mixture maximum			343	101	959	2392	250	167	196	2449	197	344	19000
Mixture average			159	64	593	1783	165	125	89	1248	157	260	19000

The derived minimum and maximum relative EF values, as well as the average relative EFs of the 15 potential mixture combinations, are displayed in Table 3, which summarizes the findings of these calculations. It should be noted that as the effects of the various waste burning emission sources are addressed separately, the obtained EF values of the various tracers are characteristic only for those waste types the combustion of which produces the given tracer compound. It is not an average EF for the entire waste mixture. Therefore, the estimated EF values of melamine that is emitted when burning furniture panels remain constant despite changes in the chemical composition of the burned waste.

It's important to note that the estimation of the contribution of a residential waste burning to ambient PM_{10} concentration carries a significant level of uncertainty. This uncertainty arises from the variability of emission factors (EFs), which depend on factors such as burning conditions and the type of waste being incinerated. In addition, many tracer compounds are emitted during the combustion of various waste types, like SSS from PS but also from paper and LDF, with highly variable emission factors, complicating precise attribution. Furthermore, in the atmosphere, all waste burning tracers are regarded as refractory, meaning that they do not break down during atmospheric transport. Therefore, it's crucial to view this assessment as a first order approximation. The estimations were evaluated using the obtained average mass ratios for the various tracer components and their concentration in the ambient samples. The PM_{10} concentration emitted from waste burning was determined and compared to the ambient PM_{10} concentrations, the obtained results with the EFs are summarized in Table 4.

Table 4 demonstrates that, based on the observed concentrations and emission factors of tracer compounds associated with waste burning, the estimated contribution of PM_{10} originating from the combustion of various waste materials to the overall atmospheric PM_{10} concentration is relatively low, typically only accounting for a few percent. However, it's important to note that, due to potential additional sources of 135-TPB and the associated uncertainty in its emission factor, the table only provides an indication of its contribution to waste burning.

When considering the estimated impact of waste incineration calculated from terphenyls, quaterphenyls, 2-BEVT, SSS, ABS pyrolysis products, and melamine, the smallest contribution of waste burning to PM_{10} levels is observed at the regional background station KPS, as well as in Veszprém. In Budapest, there is evidence of substantial burning of scrap furniture panels in households and shops, while in MSK and PUT, household waste combustion (including PET and/or RAG) is more typical. In Bucharest and Cluj, the contribution of burning furniture panels to PM_{10} concentrations appears to be significant, with a higher proportion of burning PET and other waste types. In Deva and Focsany, the predominant contribution comes from PET and/or RAG burning, similar to what was observed in PUT and MSK, with some additional input from the combustion of other waste materials such as PS and LDF.

Table 4. The contribution of waste burning to the PM_{10} concentration (expressed in percentages) based on the average ambient concentration and the estimated relative emission factor of various tracer compounds.

Tracer component	135-TPB	124-TPB	m-TPH	p-TPH	m,p-QTPH	p-QTPH	2-BEVT	SSS	ASS	SAS	SSA	Melamine
Most characteristic waste type (s):	PS/ LDF/ PET/ PAP	PET/ PS	PET/ PS/ PAP	PET/ PS	PET/ PS	PET/ PS	PET	PS/ LDF/ PAP	LDF/ PAP/ ABS	LDF/ PAP/ ABS	LDF/ PAP/ ABS	LDF
EF (μg^{-1} PM_{10})	200	100	600	1800	200	100	100	1200	200	300	100	19000
KPS	+	1.1	0.3	0.1	0.3	0.3	0.1	0.2	0.7	0.2	0.2	0.1
VES	+	1.3	0.3	0.1	0.3	0.2	0.2	1.1	1.6	0.7	0.5	0.2
PUT	+	1.2	0.4	0.2	0.3	0.2	0.4	1.4	2.0	1.2	1.3	0.5
MSK	+	1.4	0.5	0.2	0.4	0.5	1.2	1.4	1.2	1.0	0.9	0.1
CLJ	+++	1.3	0.6	0.2	0.5	0.6	1.5	1.5	1.1	0.8	0.7	0.2
DEV	++	7.9	0.2	0.1	0.4	0.4	0.7	2.4	1.8	0.8	1.2	0.5
BUD	++	2.9	0.3	0.1	0.4	0.5	1.4	2.0	1.2	1.0	1.2	0.7
FOC	++	2.1	0.3	0.1	0.4	0.5	1.3	1.5	2.3	1.2	1.6	0.3
BUC-R	+++	5.0	0.3	0.1	0.5	0.5	0.8	1.4	7.1	2.3	2.3	1.0
BUC-M (2020)	+++	7.1	0.3	0.1	0.6	0.6	1.0	2.1	10.0	4.3	4.6	1.2

5.5.1 Estimated amount of the waste burned.

The amount of solid waste burned in households can be estimated in relation to the quantity of firewood for which statistical data are available, using the absolute emission factors of the various tracer compounds found during controlled waste burning in the laboratory and reported emission factors for levoglucosan from wood burning. These estimations are loaded with significant uncertainties stemming from emission measurements, varying combustion conditions, co-firing levels, tracer stability in the atmosphere, and other underlying assumptions. Table 5 summarizes the obtained results along with the applied EFs. In our computations, the assumption was made that levoglucosan is exclusively released from wood burning at a rate of 200 mg kg^{-1} based on Jimenez et al., 2017.

The information presented in Table 5 indicates that the mass of household waste burned in comparison to firewood is relatively low, typically amounting to less than 5%. However, in certain regions of Romania (specifically, BUC, DEV, FOC), this proportion is slightly higher due to an increased practice of burning materials like LDF/PAP. It is clear from the concentrations of melamine and the pyrolysis byproducts of SSS and ABS that furniture panels constitute a substantial portion of the waste that is being burned.

Table 5. The estimated mass of burned waste relative to firewood (in percentage) at the different sampling sites based on the ambient concentration of different tracer compounds.

Burned waste relative to firewood (%)	PS/ LDF/ PET/ PAP	PS/ PET	PET/ PS	PET/ PS	PET	PET/ other	PS/ LDF/ PAP	LDF/ PAP/ ABS	LDF/ PAP/ ABS	LDF
	1,3,5-TPB	1,2,4-TPB	m,p-QTP	p-QTPH	2BEVT	TPHA	SSS	SSA	SAS	Melamine
EF (mg kg^{-1})	1.5	0.5	1.6	2.1	1	90.7	6.9	0.5	0.9	51.4
KPS	6	3	0.5	0.2	0.2	4	0.5	1	1	0
VES	5	3	0.3	0.1	0.2	4	2	1	2	1
PUT	3	2	0.4	0.2	1	5	2	1	2	1
MSK	5	3	1	0.2	1	7	2	2	3	0
CLJ	17	10	0.4	0.1	1	5	4	2	2	1
DEV	8	4	0.5	0.2	1	6	3	2	3	2
BUD	7	3	0.4	0.1	0.4	5	3	3	4	2
FOC	10	4	1	0.2	1	9	2	3	4	1
BUC-R	23	12	1	0.3	1	10	3	5	8	4
BUC-M (2020)	25	15	1	0.3	1	9	4	12	16	6

Assuming that the ratio of waste burned relative to firewood is 3–5% of that of the firewood in both countries and taking into account that the total mass of firewood annually consumed in Hungary and Romania in 2019 were 7.4 and 12.2 million tonnes, respectively (Clean Air Action Group, Romanian Statistical Office), the calculations reveal that approximately 8-13% of solid waste is incinerated in households in Hungary. Similarly, in Romania, this estimate would be around 9-15%. These calculations are based on the assumption that the annual production of household waste in Hungary and Romania stands at 2.7 million tonnes and 4.1 million tonnes, respectively, as reported by EUROSTAT (https://ec.europa.eu/eurostat/databrowser/view/ten00106/default/table?lang=en).

5.6 Assessment of the atmospheric stability of novel tracers for residential waste burning

It is known that the aging process of tracer compounds can be affected by a range of factors, including temperature, light, moisture, and the presence of other chemicals. For instance, in air environments, tracer compounds can be oxidized by atmospheric pollutants or UV radiation, which can change their chemical structure and make them less effective as tracers (Simoneit, B. R. (2016)). The data in this chapter should be interpreted on a relative scale; that is, since the precise half-life or residence time cannot be determined due to the difference of the kinetics and processes of the reactions in the aerosol phase compared to the filter material, the findings will allow us to speculate about potential changes and/or the formation of new components.

In the results, I observed the variation in the stability of the compounds, with some declining rapidly while others were relatively stable. According to Table 6, such an aromatic compound as pTPH was quite stable as expected, on average 80% of the original amount remained in the filter after 7 days of aging time. In the case of 135TPB on average 78% of the original compound was identified during the period, and p-QTPH showed about 81% stability.

Besides aromatic compounds, the results of pyrolysis products such as melamine and ABS compounds from LDF samples were quite stable as well (Table 6). The ABS pyrolysis products (ASS, SAS, SSA) decreased by about 16-24% during 7 days of aging time, in spite of the fact that they contain a double bond. However, due to the wood content of the LDF, humic-like substances (HULIS) were present in the samples, therefore they could act as antioxidants by inhibiting the oxidation of these components. In the case of melamine, around 94% of the concentration was left on the filter.

For the 2-BEVT in the PET sample, I found that only 7% remained on the filter after 7 days of aging time, the larger loss of this component can be explained by the absence of HULIS in the filter samples, as to prepare PET (and also PS) samples, the waste was burned on glowing charcoal. It is important to note that in the ambient samples the HULIS presence in the samples might extend the lifetime of this component.

Table 6. The relative amount of the compounds expressed as % of the amount measured on the original filter. (The values larger than 100% is the result of the possible inhomogeneity of the original filter sample (which was sampled directly from the smoke of the burned waste) and the measurement uncertainty).

		Aging time (H)						
Tracer compound	Waste type	0	1	3	9	24	72	168
p-terphenyl	PET	100	93	90	90	91	91	81
	PS	100	109	99	100	87	80	65
	LDF	100	89		92	111	108	94
135-TPB	PET	100	90	81	94	90	90	76
	PS	100	103	94	94	87	77	67
	LDF	100	88		86	93	98	92
p-QTPH	PET	100	91	85	90	94	91	81
Melamine	LDF	100	91		90	95	93	94
ASS	LDF	100	90		88	91	91	84
SAS	LDF	100			87	96	87	78
SSA	LDF	100	91		89	99	88	76
2BEVT	PET	100	87	71	60	34	21	7

As it can be seen from the Figure 23, the SSS concentration in the PS sample decreased drastically during the first 20 hours, its amount was about 3% of the initial concentration. The SSS concentration from the LDF samples also shows a slight decrease during the aging period but not at the same rate as the PS samples. In LDF sample, the SSS was quite stable (68% remained in the filter after 7 days of aging time). As SSS is also released during the burning of furniture panels, in such cases, HULIS was present in the samples due to the combustion of wood. Consequently, in the case of furniture panels, the conversion of SSS is less pronounced (Fig. 22), likely due to antioxidant property of the HULIS. Therefore, it can be concluded that more reactive compounds like SSS or 2BEVT tend to persist longer in ambient conditions since the particles also contain HULIS.

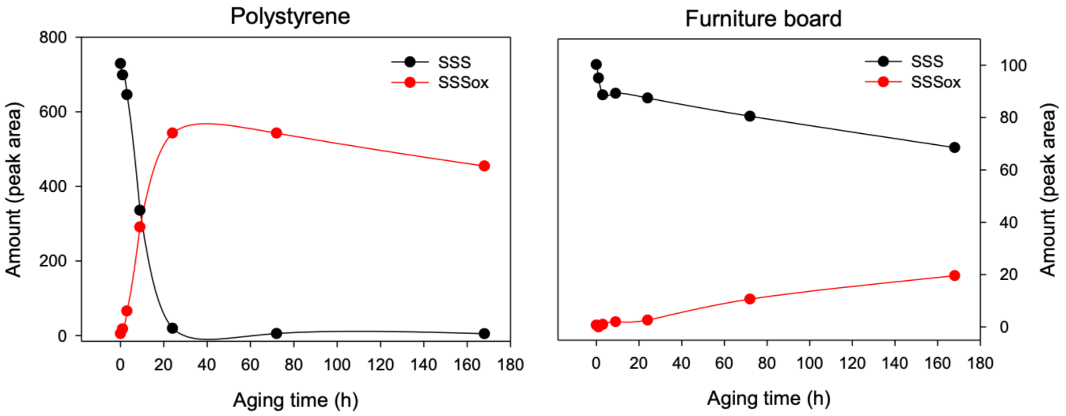

Figure 22. The atmospheric stability of PS(SSS), LDF(SSS) and their oxidation products during aging time.

During the assessment of atmospheric stability, a new compound, 1,3,5-triphenylpentan-1-one (SSSox), resulting from the oxidation of SSS was discovered in the chromatograms. The mass spectra of the SSS and that its oxidation product are shown in Figures 23 and 24, respectively. SSS has a molecular weight of 312 as it can also be seen from the mass spectra (Fig. 23), whereas the newly formed compound has a molecular weight of 314 (Fig. 24). Notable fragment ions included m/z=117, 194, 207 and 105, 194, 91 for SSS and SSSox compounds, respectively. The identified fragment ions align with the proposed structure of SSSox, presenting solid proof of its existence.

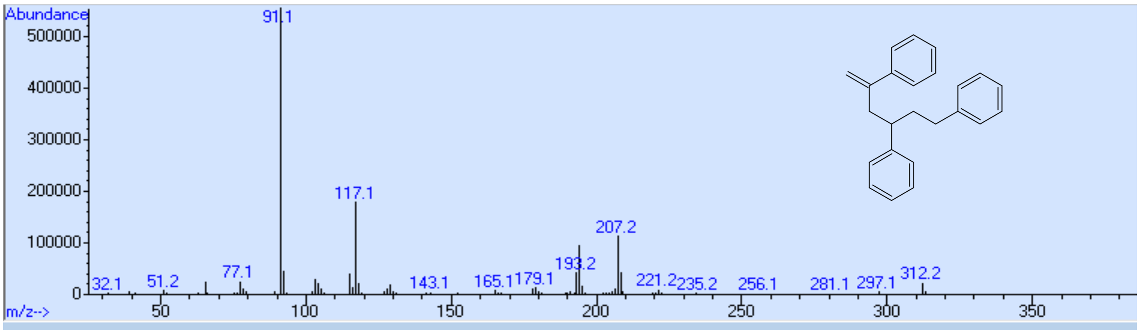

Figure 23. The mass spectra of SSS compound.

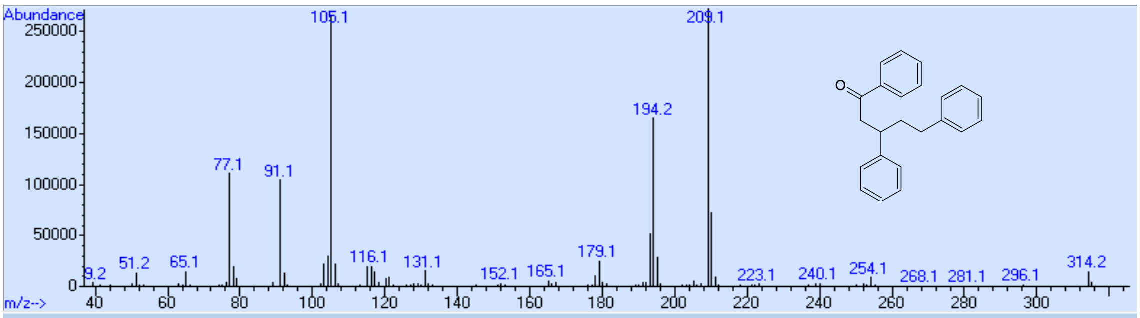

Figure 24. The mass spectra of SSSox compound.

Following the aging experiment, a reanalysis of selected ambient samples collected in Miskolc, Putnok, Deva, and Bucharest was conducted. The selection of these samples was based on variable PM_{10} concentrations (caused by stable atmospheric conditions as well as the change in the meteorological conditions) in winter.

The analysis revealed that the presence of SSSox is notable in the ambient samples collected at these stations. It was also observed that, in most of the samples, the relative proportion of SSS (compared to SSSox) increased as the PM_{10} concentrations rose, signifying the influence of local sources. Conversely, when meteorological conditions changed, and transport processes played a more significant role in the aerosol composition (resulting in a decrease in PM_{10} concentrations as cleaner and aged air arrived at the stations), the relative amount of SSSox increased (see Figure 25). Given the concentrations of these two components, it seems possible to estimate the effect of local waste burning sources on air quality on a relative scale.

Putnok, which is a small town close to Miskolc, experienced a more pronounced impact from transport processes than Miskolc, because the sampling location was at the edge of the settlement. As a result, the concentration of SSSox relative to SSS was higher at the Putnok station than in Miskolc on the same sampling days (Fig. 25).

In the case of Deva, the findings align with the high PM_{10} concentration, indicating a significant presence of local sources. At this station, SSSox levels were also higher when the PM_{10} concentration decreased, likely due to the aged air that was sampled.

In Bucharest, a large variation in some of the tracer compounds has been observed, and this can be observed here as well. Intriguingly, only around 50% of the samples collected in Bucharest contained SSS originating from waste burning. Despite this, the SSS-to-LGS ratio was among the highest, signifying occasional episodes of waste burning events, which affect the SSS/SSSox ratio.

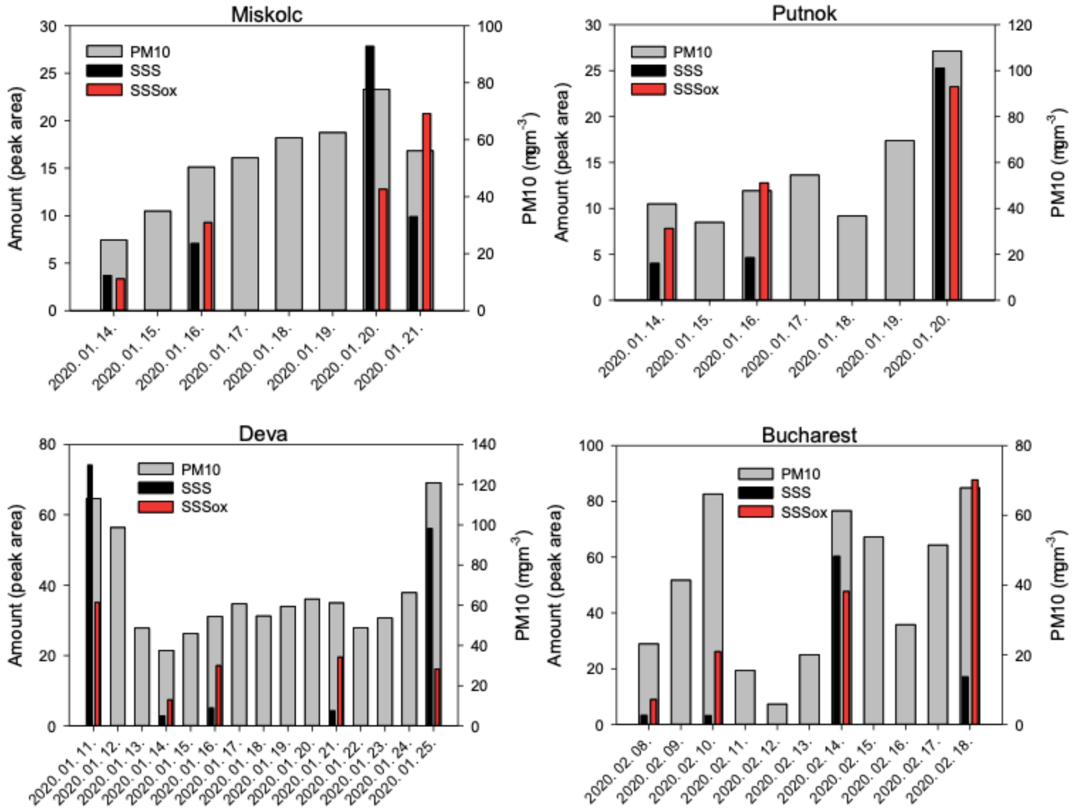

Figure 25. Concentrations of SSS and SSSox compared to PM_{10} in selected ambient samples in 4 settlements in Hungary and Romania.

6. Conclusions

Waste burning in households, although often illegal, remains a common practice, leading to the emission of significant amounts of PM_{10} into the atmosphere.

Understanding the contribution of waste burning to PM_{10} concentrations is highly important, as the emitted particles have a significant environmental and health impact. Laboratory experiments were conducted, where various types of plastics typically found in household waste as well as treated wood samples were burned under controlled conditions. Tracer compounds characteristic for the burning of different waste types were identified and their emission factors as well as their mass ratio to the emitted PM_{10} were calculated.

The findings revealed that the relative emission factor of 1,3,5-TPB (the universal tracer compound initially suggested by Simoneit et al., (2005)), depends on the type of plastic material burned, with plastics containing aromatic rings in their structure emitting higher values. Structural isomers of 1,3,5-TPB (124TPB and quaterphenyls) were also identified in the samples. It was found that the concentration ratio of triphenylbenzenes to quaterphenyls can help to infer the type of burned waste, as burning of PET produces larger amount of linear quaterphenyls. M-terphenyl and p-terphenyl, could serve as additional indicators for waste burning, and their ratio might provide insights into the type of burned material. Specific tracer compounds associated with polymer fragments formed during pyrolysis processes were also identified. The trimer of styrene (SSS) was emitted in large quantities from the burning of polystyrene, low-density fiberboard, and coated paper samples. The burning of low-density fiberboard potentially emitting melamine, a component often used during its production. The compound 2-(benzoyloxy)ethyl vinyl terephthalate (2BEVT) was identified in samples collected during the burning of PET and rag samples. The emission factor of the tracer compounds varied depending on the combustion parameters.

These tracers were then analyzed in atmospheric PM_{10} samples collected in various locations in Hungary and Romania during consecutive heating seasons as well as in summer. The research revealed that PET-containing waste was frequently burned in the northeastern regions of Hungary, with the concentration ratio of p-terphenyl to m-terphenyl supporting this observation. Also, the ratios of such aromatic compounds as m,p-QTPH/124-TPB supported the assumption that the burning of PET-containing waste is burned at these locations. Similar trends were observed in Romanian cities, indicating significant PET burning. The presence of the styrene trimer (SSS) and melamine served as tracers for burning specific types of waste, including furniture panels, household waste with polystyrene (PS), and coated waxy paper. indicated the burning of furniture panels. These tracers were present the most in the capitals of both countries.

The estimated contribution of waste burning emissions to the ambient PM_{10} mass concentration was relatively small, around a few percent. However, taking into account the high emission factors and toxicity of Polycyclic Aromatic Hydrocarbons (PAHs) from waste burning, this contribution posed significant health risks. Using the emission factors and the measured concentrations of tracer compounds for wood burning (levoglucosan) and waste burning in the ambient samples, the mass ratio of the burned waste compared to wood was estimated. By using statistics on firewood consumption and annual produced household waste in Hungary and Romania, it was estimated that approximately 10% of household waste was being illegally burned in household stoves during the heating seasons.

Additionally, this study contains essential information regarding the identification and stability of novel tracer compounds suitable as tracers for waste burning. In the experiments investigating the relative stability of the tracer components, I identified a new compound, the 1,3,5-triphenylpentan-1-one which is the oxidation product of the styrene trimer.

Thesis points:

1. Identification of novel and specific tracers for residential waste burning

Tracer compounds that can potentially be used to monitor waste burning in ambient particulate matter were identified by gas chromatography-mass spectrometry of PM_{10} aerosol samples collected under controlled laboratory burning of different waste types in a household stove. I have detected novel pyrolysis products of waste polymers that had never been observed and described as tracers in atmospheric aerosol. When burning PET (polyethylene terephthalate) containing wastes (PET bottles and rags) I have identified 2-(benzoyloxy)ethyl vinyl terephthalate in high concentrations. Upon the burning of furniture boards containing melamine-formaldehyde resin, I have found free melamine as an abundant pyrolysis product in the collected PM_{10} particles. Similarly, the trimer of styrene (5-hexene-1,3,5-triyltribenzene) has been identified in the PM_{10} samples collected during the burning of styrene containing wastes (insulation boards, glossy coated papers, furniture boards). During the burning of ABS (acrylonitrile butadiene styrene)-containing waste types (housing of electronic equipment, furniture panels) I have detected hybrid trimer compounds consisting of various combinations of styrene and acrylonitrile monomers (ASS, SSA, and SAS). I have also detected and in most cases quantified all of these tracers in ambient PM_{10} samples collected in several locations during the heating season.

2. Application of non-specific tracers for the assignment of waste types in residential waste burning emissions

I have found the universal tracer for waste burning, the 1,3,5-triphenylbenzene, in the PM samples from the controlled burning of all waste types, at the highest concentrations in waste emissions containing larger number of aromatic rings in their structure (PS and PET). I have also confirmed that the burning of PET-containing wastes, as well as of PS, ABS, and glossy papers are significant sources of terphenyls and quaterphenyls. I have hypothesized that these compounds are not specific to the type of waste burned, but their relative mass ratios are. I have proven that straight-chain quaterphenyls and para-terphenyl are formed in higher amounts during the burning of PET and PET-containing waste types than branched quaterphenyls and meta-terphenyl.

3. Determination of the normalized mass emission factors of specific tracers for residential waste burning

I have determined the relative mass ratio of specific tracers to PM_{10} in particulate emissions from controlled waste burning, as well as their absolute mass emission factors. As a base case I have assumed residential waste composition for plastics consisting of 42.3% PE, 28.2% PET, 14.1% PP, 14.1% PS, 0.7% PVC, and 0.7% ABS. I have calculated the emission factors for waste mixtures with varying proportions of furniture boards (ranging from 10% to 91.7%) and mixed plastics from household waste (ranging from 1.8% to 65.2%). The relative mass emission factors were the highest for melamine (up to 19 mg g^{-1} PM_{10}), while for other tracers it ranged between 100 and 1800 µg g^{-1} PM_{10}. The absolute emission factors of the waste burning tracers ranged from 0.5–51 mg kg^{-1}.

4. Assessment of the contribution of residential waste burning emissions to ambient PM_{10} mass concentrations in selected Hungarian and Romanian settlements during the heating seasons

Based on the quantification of specific and non-specific tracers in both controlled laboratory experiments and ambient measurement campaigns in Hungary and Romania I have assessed the contribution of residential waste burning particulate emissions to the atmospheric PM_{10} levels in several settlements during the heating season. I have presented evidence on the mass burning of scrap furniture panels in big cities such as

Budapest, Bucharest, and Cluj, and of plastic wastes (such as PET and rag) in Miskolc, Putnok, Deva, and Focsany.

5. Assessment of the total mass of residential waste burned relative to firewood burning

By using the absolute emission factors of the specific tracers for residential waste burning as well as that of the widely used wood burning tracer levoglucosan I have estimated the total mass of residential solid waste that possibly ended up in household stoves. For the calculations, I have used statistical reports on firewood sales and waste production in both countries, as well as other publicly available relevant information. I have found that up to 10% of solid waste produced in households may be burned in stoves during the study period.

6. Assessment of the atmospheric stability of novel tracers for residential waste burning

Based on the estimated vapor pressures of the tracers I have shown that they are predominantly partitioned into the aerosol phase. By experimentally studying the chemical stability of the tracers, I have concluded that terphenyls, quaterphenyls, and melamine have longer residence times in the atmosphere than pyrolysis products containing at least one double bond. I have hypothesized that due to the antioxidant properties of atmospheric humic-like substances also present in combustion-derived particulates even the latter compounds are more resistant to photooxidation and thus have longer residence time. For the first time in atmospheric science, I have identified a novel compound (1,3,5-triphenylpentan-1-one) formed by the photooxidation of the primary burning tracer styrene trimer (5-hexene-1,3,5-triyltribenzene), which might be considered as an indicator (i.e. secondary tracer) for residential waste burning and subsequent atmospheric aging processes.

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