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

THESES OF THE PHD DISSERTATION

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INTRODUCTION AND AIMS

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 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} 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).

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.

THESES

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.

LIST OF PUBLICATIONS AND PRESENTATIONS

Hungarian Scientific Bibliography (MTMT2) Author ID: 10086938

Publications:

Hoffer, A., Tóth, Á., Jancsek-Turóczi, B., Machon, A., Meiramova, A., Nagy, A., ... & Gelencsér, A. (2021). Potential new tracers and their mass fraction in the emitted PM 10 from the burning of household waste in stoves. Atmospheric Chemistry and Physics, 21(23), 17855-17864.

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