

# Residential Wood Combustion in Germany: A Twin-Site Study of Local Village Contributions to Particulate Pollutants and Their Potential Health Effects

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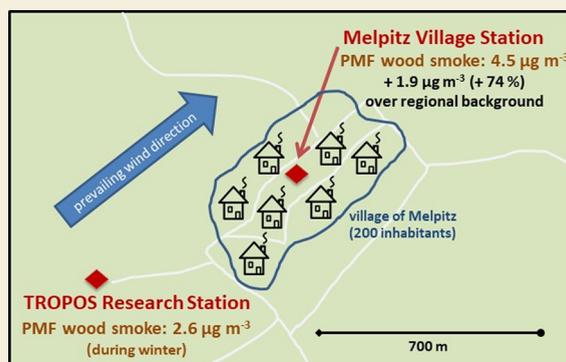
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Supporting Information

**ABSTRACT:** Residential wood combustion contributing to airborne particulate matter (PM<sub>10</sub>) was studied for 1 year at two sites in the village of Melpitz. Significant excess pollution was observed at the Melpitz center compared to that at the TROPOS research station Melpitz reference site, situated only 700 m away. Local concentration increments at the village site for the combustion PM constituents organic carbon, elemental carbon, levoglucosan, and benzo[a]pyrene were determined under appropriate wind directions, and their winter mean values were 0.7  $\mu\text{g m}^{-3}$ , 0.3  $\mu\text{g m}^{-3}$ , 0.1  $\mu\text{g m}^{-3}$ , and 0.4  $\text{ng m}^{-3}$ , representing relative increases over the regional background concentration of 24, 70, 61, and 107%, respectively. Yearly, weekly, and diurnal profiles of village increments suggest residential heating as the dominant source of this excess pollution, mainly originating from wood combustion. Receptor modeling using positive matrix factorization quantified 4.5  $\mu\text{g m}^{-3}$  wood combustion PM at the village site, representing an increment of 1.9  $\mu\text{g m}^{-3}$  and an increase of  $\sim 75\%$  over the 2.6  $\mu\text{g m}^{-3}$  regional background wood combustion PM. This increment varied with season, temperature, and boundary layer height and reached daily mean values of 4–6  $\mu\text{g m}^{-3}$  during unfavorable meteorological conditions. Potential health effects were estimated and resulted in an all-cause mortality from short-term exposure to wood combustion PM of 2.1 cases per 100,000 inhabitants and year for areas with similar wood smoke levels as observed in Melpitz. The excess cancer risk from the concentrations of polycyclic aromatic hydrocarbons was 6.4 per 100,000. For both health metrics, the very local contributions from the village itself were about 40–50%, indicating a strong potential for mitigation through local-scale policies. A compilation of literature data demonstrates wood combustion to represent a major source of PM pollution in Germany, with average winter-time contributions of 10–20%. The present study quantifies the negative impacts of heating with wood in rural residential areas, where the continuous monitoring of air quality is typically lacking. Further regulation of this PM source is warranted in order to protect human health.

**KEYWORDS:** residential heating, biomass burning, air quality, aerosol particles, chemical composition, source apportionment



## 1. INTRODUCTION

Air pollution represents a significant environmental health risk in urban and rural areas,<sup>1,2</sup> and many strategies have been made at different scales to reduce the effects worldwide.<sup>3,4</sup> Air quality policies in Europe have helped reduce emissions of trace gases and particulate matter (PM) including the combustion contributions from the industry and transportation sectors.<sup>5</sup> On the other hand, the promotion to use renewable fuels in the framework of climate change mitigation policies, see EU<sup>6</sup> for such EU measures, which are also criticized, e.g., by Searchinger et al.,<sup>7</sup> as well as the rising in fossil fuel prices have contributed to positioning wood combustion as a significant source of airborne PM in many areas of Europe.<sup>8–11</sup> PM from wood combustion is also associated with toxic

constituents like soot,<sup>12</sup> a large and diverse fraction of organics,<sup>13</sup> including polycyclic aromatic hydrocarbons (PAHs), which are linked with cardiovascular diseases and carcinogenic effects.<sup>14–16</sup>

The major source of primary PM with aerodynamic diameters smaller than 10 or 2.5  $\mu\text{m}$  (PM<sub>10</sub> and PM<sub>2.5</sub>) is residential fuel combustion emissions within Europe, con-

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tributing 40 and 56%, respectively, to the total emissions for the year 2016.<sup>8</sup> The increase in wood combustion in Europe resulted in increases of PM<sub>10</sub> and PM<sub>2.5</sub> emissions by about 7 and 11%, respectively, between 2005 and 2017.<sup>17</sup> For 2018, the countries contributing the most to the annual emission rates for PAHs within Europe were Germany, Poland, Greece, and Spain, with together more than 10% of total European emissions.<sup>18</sup> The contribution of wood combustion to the total PM concentrations is often estimated through the macrotracer levoglucosan, a monosaccharide anhydride produced from the oxidative and dehydrative breakdown of cellulose and thus emitted during biomass burning,<sup>19</sup> or through receptor modeling, including chemical mass balance and positive matrix factorization (PMF).<sup>9</sup> In Europe, higher PM concentrations due to wood burning typically occur during the heating season.<sup>14,20,21</sup> These emissions from residential heating can lead to PAH concentrations during winter time several times higher than during the rest of the year, and accordingly, wood combustion can contribute more to ambient PAH concentrations than other sources like fossil fuel combustion in gasoline and diesel vehicles.<sup>22</sup>

One of the main factors for increasing wood-burning contribution to PM concentrations during the heating season is small appliances used for domestic heating, mainly wood stove systems.<sup>23,24</sup> In Germany, an estimated 11.2 million small heating units generating energy from solid fuels, mostly wood, were reported by 2018.<sup>25</sup> The contributions of their emissions to urban and rural air quality are, however, not well characterized as monitoring stations are typically not located in residential areas and often lack the chemical information needed to identify wood-burning contributions. At the TROPOS research station close to Melpitz, in a rural area in the East of Germany, PMF modeling estimated average wintertime wood-burning contributions of 25 or 45% of the total PM<sub>10</sub> concentration depending on meteorological conditions.<sup>21</sup> In another study, long-range transported pollution, especially during eastern air mass inflow, was reported to explain up to 50% of the wood-burning PM<sub>10</sub> at the Melpitz research station.<sup>11</sup> More locally resolved observations, especially in residential areas, are largely lacking, but only these would allow for better discrimination between background and local contributions.

An approach that addresses the local contributions from specific sources is the analysis of concentration increments from parallel twin-site measurements. It has often been used to deduce local traffic source contributions<sup>26–28</sup> or urban contributions over a regional background<sup>29–31</sup> but less frequently to derive local wood burning contributions in residential areas.<sup>21,32</sup> For urban increment calculations, the parallel monitoring sites are often some tens of kilometers apart, which has raised concerns about the validity of this approach.<sup>33</sup>

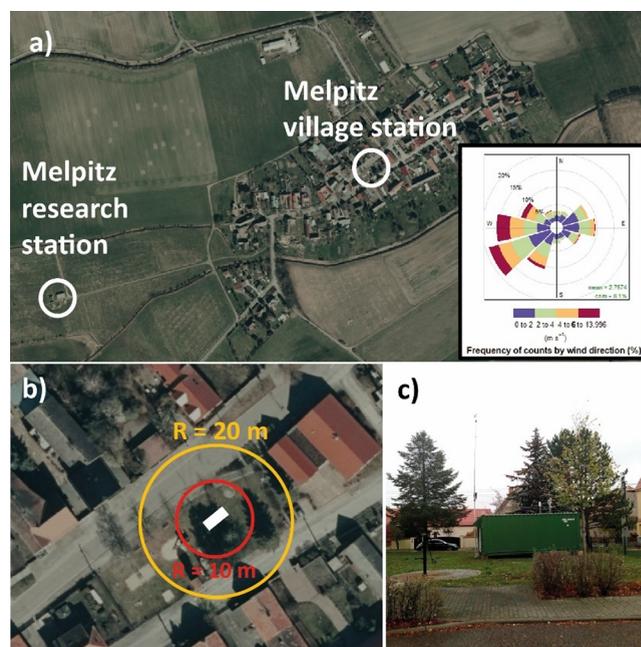
Within the present study, the contribution of local wood combustion to PM<sub>10</sub> mass and its constituents was determined in a rural residential area from twin-site measurements obtained during the course of one full year. The stations were less than 1 km apart from each other, representing the regional background and local village conditions in Melpitz, Germany (Saxony). Careful data filtering was applied to minimize the potential impacts of the village emissions on the background concentrations. Increments in the concentrations of PM<sub>10</sub> and selected combustion tracers as well as from PMF-modeled source contributions were determined for the village

station and discussed with respect to meteorological conditions and seasons. Finally, a health risk assessment evaluated the excess cancer risk and mortality due to residential wood combustion, and the results are discussed in the context of typical wood-burning contributions to PM in Germany.

## 2. EXPERIMENTAL SECTION

### 2.1. Sampling Sites and Campaign Setup

A twin-site sampling concept was implemented in Melpitz (51.54 N, 12.93 E, 86 m.a.s.l.), a small village with approximately 200 inhabitants located approximately 50 km northeast of Leipzig. The Melpitz village is located near to the TROPOS research station Melpitz which was used as the reference station to characterize regional background conditions. The Melpitz research station is a well-established and long-running measurement site located on a meadow west of the village and surrounded by a flat agricultural land.<sup>34</sup> A twin site with identical instrumentation was placed in the village center to characterize local conditions. The village station was approximately 700 m east of the research station, which is windward of the village under the prevailing wind direction. No emission sources are present between the research station and the village boundaries. In Figure 1, the locations of the two sites are shown.



**Figure 1.** Overview of sampling sites with (a) locations of the Melpitz research station and Melpitz village station, including a wind rose showing the main air mass inflow from the southwest during the campaign, (b) location of the measurement container in the village center, and (c) photo of the measurement container at the village center. Source of satellite images in (a) and (b): GeoSN<sup>35</sup> (dl-de/by-2-0).

The distance between the measurement container in the village and the nearest building was at least 20 m in northern, eastern, and southern directions and about twice as much in the western direction. Measurements of particulate and gaseous air pollutants as well as meteorological conditions were conducted in parallel at both sites over the course of one full year, between November 1, 2018, and October 31, 2019. In the following, the two sites are referred to as the “Melpitz research station” and “Melpitz village”, corresponding to the regional background and local village conditions, respectively. Under the prevailing winds, air masses first reach the Melpitz research station and, subsequently, Melpitz village. According to the authors’

knowledge, such Lagrange-type twin station measurements have not yet been designed or applied for a dedicated detailed study and quantification of the wood combustion contribution to local PM.

## 2.2. Instruments

A number of online and offline methods were used in the campaign to measure wood-burning-related air pollutants and auxiliary data, which are briefly described in the following. In addition to these measurements, standard meteorological parameters were obtained at both sites by using a Vantage Pro 2 automatic weather station (Davis Instruments, Hayward, CA, USA).

**2.2.1. Filter Sampling and Chemical Analysis.** Digital high-volume filter samplers with a PM<sub>10</sub> inlet head were used to collect particles on quartz fiber filters with daily samples from 0 to 24 h Central European Time (CET), and mass concentrations were determined by differential weighing.<sup>36</sup> At the research station, the Digital sampler is placed outside and operated under ambient conditions. The Digital sampler at the village station, however, had to be operated at a constant ~20 °C inside the air-conditioned measurement container to avoid noise that would disturb the village population nearby. Given that sampled filters were picked up every 3 to 4 days, the different temperatures during sampling and sample storage until pick-up might alter the PM composition to some extent, especially with respect to semivolatile or thermolabile constituents. To check for this, quality control filters were taken every sixth day in an air-conditioned container at the research site during the entire campaign, leading to approximately 16 filter pairs per season for which the concentrations of inorganic ions and organic carbon were determined. In total, over the campaign, 730 samples at the two sites and 62 quality control filters were taken.

The chemical composition of particles was determined from the filter samples by a range of analytical techniques, which are specified in detail elsewhere.<sup>36–39</sup> Briefly, concentrations of inorganic ions and oxalate were measured from aqueous filter extracts by ion chromatography (IC), the anhydromonosaccharides levoglucosan, mannosan, and galactosan were analyzed by IC coupled with an electrochemical detector, organic and elemental carbon (OC/EC) were quantified according to the EUSAAR2 temperature protocol,<sup>40</sup> water-soluble organic carbon (WSOC) was determined using a Total Organic Carbon (TOC) analyzer, and the determination of the PAHs benzo[a]anthracene (BaA), benzo[a]pyrene (BaP), benzo[b]fluoranthene (BbF), benzo[j]fluoranthene (BjF), benzo[k]fluoranthene (BkF), dibenzo[a,h]anthracene (DBaA), and indeno[1,2,3-cd]pyrene (IndP) was carried out by means of Curie-point pyrolysis gas chromatography–mass spectrometry (CPP-GC/MS).<sup>41</sup> Analytical uncertainties of these methods from repeated analyses are typically in the range of 3–10%.<sup>36–39</sup>

## 2.2.2. Particle Number Concentration and Black Carbon.

Particle number size distributions (PNSDs) were measured by mobility particle size spectrometers (MPSS), type TROPOS-TSMPS (research station) and SMPS (village),<sup>42</sup> and aerodynamic particle size spectrometers (APS), type TSI Model 3321.<sup>43</sup> Both PNSDs were merged at a diameter of 700 nm, assuming compact spherical particles with an aerodynamic shape factor = 1, constant aerosol density, and mobility particle diameter equal to the volume equivalent particle diameter, following Alas et al.<sup>44</sup> At the research station, the time resolution of the PNSD measurements was 10 min, while it was 5 min at the village site. Three measurements of particle number size distribution per hour were made at the research station, while in the remaining 30 min of each hour, the PNSD of nonvolatile particles was measured after a thermal denuder (data not used in this study). All PNSD measurements were aggregated to hourly values for further analysis. PNSD data were used to calculate hourly resolved PM<sub>1</sub> and PM<sub>10</sub> mass concentrations, assuming a particle density of 1.6 g cm<sup>-3</sup><sup>45</sup> across the entire size spectrum.

The concentration of equivalent black carbon (eBC) in PM<sub>10</sub> was derived from the measurement of the absorption coefficient using a multiangle absorption photometer (MAAP)<sup>46</sup> using a reference mass absorption cross-section of 6.6 m<sup>2</sup> g<sup>-1</sup> and a correction factor of 1.05 to account for the nominal and actual wavelength of the light

source.<sup>47</sup> The time resolution of the MAAP was 1 min, and the data was aggregated to hourly values.

## 2.3. Source Apportionment

Gravimetric PM<sub>10</sub> mass concentrations were apportioned to different sources by PMF using the data set of the determined constituents' concentrations. PMF was performed using the "EPA PMF v5" software, which is freely available from the U.S. Environmental Protection Agency.<sup>48</sup> Data preparation was conducted similarly to earlier studies in the Leipzig/Melpitz area.<sup>11,21</sup> Briefly, missing data were replaced with median concentrations and were assigned a high uncertainty of 4 x the median concentration to effectively eliminate their influence on the PMF solution. Data below the limit of detection (LOD) were replaced by 0.5 times the LOD. The uncertainty matrix was calculated from (i) the uncertainty of the LOD, defined as 5/6 of the LOD, and (ii) the analytical uncertainty from repeated standard measurements. Concentrations from both sampling sites were pooled into one data set, and several PMF runs were performed to resolve between 4 and 9 factors. The final solution was chosen based on the mathematical criteria as described in van Pinxteren et al.<sup>21</sup> and especially on the physical plausibility and interpretability of the solutions.

## 2.4. Data Treatment and Analysis

All data processing, analysis, and plotting was performed in R,<sup>49</sup> including the "tidyverse"<sup>50</sup> and "openair"<sup>51</sup> packages. When time-resolved data was aggregated to coarser resolution, e.g., one min to hourly values, a 75% data availability threshold was applied; otherwise, the corresponding individual data point was set to "not available". Data from December 31 and January 1 were excluded from any analyses, while data from December 15, 2018 were excluded in the course of data analysis due to a special event (see Section 3.1). For weekday-dependent observations, public holidays were assigned to "Sunday", regardless of the actual day of the week. All times are given in CET throughout the entire year and indicate the start of the sampling (or aggregation) interval.

## 2.5. Health Risk Assessment

A health risk assessment was done using methodologies as recommended by the World Health Organization (WHO), i.e., the excess cancer risk (ECR) metric<sup>52</sup> and the expected number of mortality cases due to outdoor air pollution.<sup>53</sup>

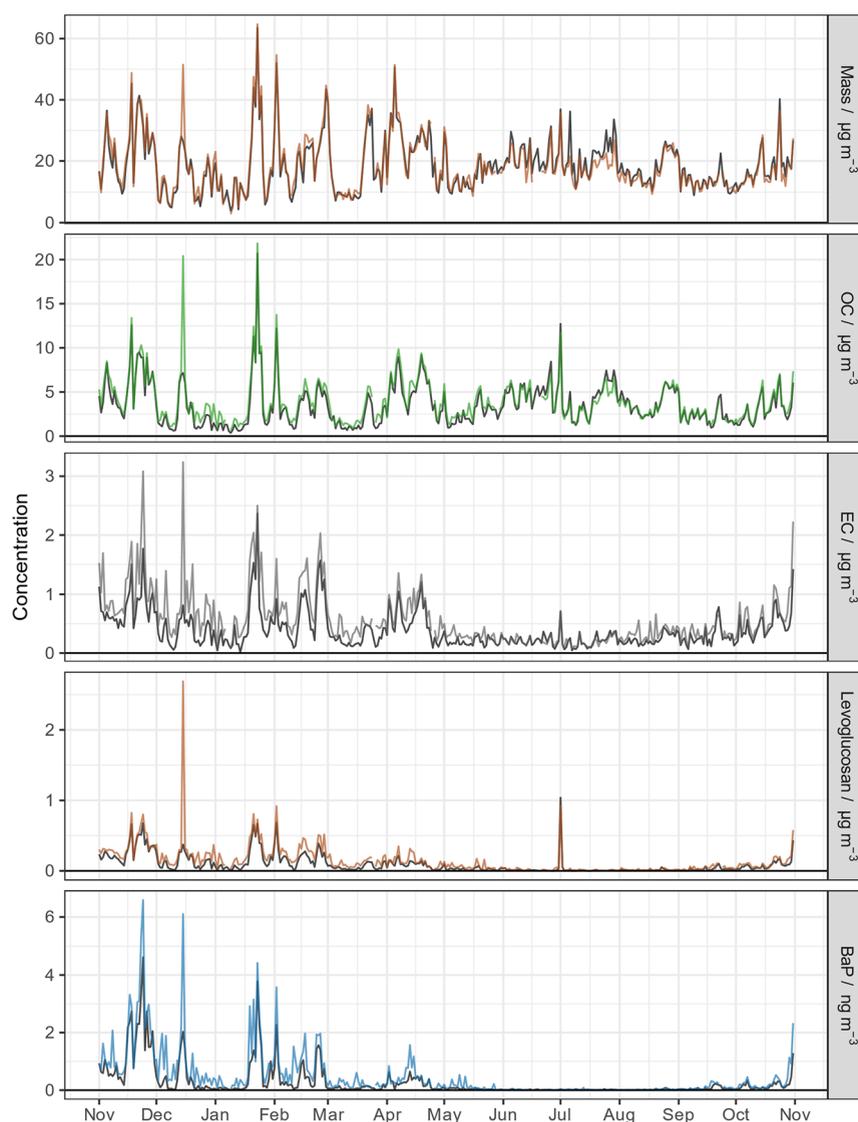
The ECR due to inhalation of a PAH mixture was used to estimate the population health risk using the concentrations of the seven measured PAHs. The ECR from inhalation is obtained from eq 1, where BaP<sub>eq</sub> is the benzo(a)pyrene toxic equivalent concentration of a specific PAH and UR<sub>BaP</sub> is the inhalation unit risk number of exposure to BaP, representing the excess cancer cases in the population with chronic inhalation exposure to 1 ng m<sup>-3</sup> of that PAH over a lifetime of 70 years.<sup>54,55</sup> The WHO uses an UR<sub>BaP</sub> reference value of 8.7 × 10<sup>-5</sup> (8.7 cases per 100,000 people).<sup>52</sup> The total BaP<sub>eq</sub> corresponds to the sum of every BaP<sub>eq</sub>.<sup>22,56</sup>

$$ECR = \sum BaP_{eq} UR_{BaP} \quad (1)$$

A BaP<sub>eq</sub> concentration of a PAH is the product of its ambient concentration and its corresponding toxic equivalent factor (TEF), as shown in eq 2, where C<sub>i</sub> is the PAH concentration in ng m<sup>-3</sup> and TEF<sub>i</sub> is the TEF of each PAH. The TEF, also called the potency equivalent factor, has been widely used to evaluate the carcinogenic risk of PAH mixtures. The TEF for the individual PAHs measured in this study was obtained from the literature.<sup>57–59</sup>

$$\sum BaP_{eq} = \sum (C_i TEF_i) \quad (2)$$

The health risk was estimated with the PAH concentrations measured in the village of Melpitz for the entire period of the measurements and for the winter season. Also, to estimate how much risk is caused by the local residential combustion of wood and coal, the same was done by using only the local annual increments at the village. Additionally, the contributions of each BaP<sub>eq</sub> to the total BaP<sub>eq</sub>



**Figure 2.** Time series of daily PM<sub>10</sub> mass and wood burning related constituents' concentrations (OC, EC, levoglucosan, and benzo(a)pyrene) during the campaign from November 2018 to October 2019. Concentrations at the Melpitz village are given as colored lines, while the reference background concentrations at the Melpitz research station are given in black.

were estimated and compared to the modeled BaP<sub>eq</sub> concentrations for Europe for the year 2018.<sup>60</sup>

For the village of Melpitz, the expected number of cases of premature mortality ( $E$ ) from all-cause short-term exposure to PM<sub>10</sub> was estimated using the methodology recommended by WHO for assessing the environmental burden of disease due to air pollution on regional and global scales.<sup>53</sup> All-cause mortality refers to the death rate from all causes of death for a population in a given time period. Short-term exposure refers to the acute or more immediate health outcomes.<sup>61</sup>

To assess the mortality  $E$ , the concentration–response function  $RR$  and the impact fraction  $AF$  were calculated for short-term exposure to PM<sub>10</sub> according to eqs 3 and 4.

$$RR = \exp[\beta(X - X_0)] \quad (3)$$

$$AF = \frac{(RR - 1)}{RR} \quad (4)$$

In eq 3, a  $\beta$  coefficient of 0.0008 was used as the recommended value by WHO, with a 95% confidence interval (CI) of 0.0006–0.0010.<sup>53</sup> As intended to estimate the mortality due to the wood-burning PM in the village, the baseline concentration  $X_0$  was set to 0,

i.e., no wood-burning smoke present in ambient air, and the actual concentration  $X$  was set to the PMF-derived source contributions of biomass burning PM<sub>10</sub>. It is noted that these choices implicitly assume the same health impact of wood-burning PM as that of total PM<sub>10</sub>, for which the epidemiological relationships were derived for. While this is not necessarily the case, recent review articles have summarized that there is insufficient evidence to conclude on a significantly different health impact of wood smoke particles than general ambient fine PM<sup>62</sup> or have even suggested a somewhat higher risk for all-cause mortality.<sup>63</sup>

The mortality term ( $E$ ) can then be calculated with eq 5, where ( $B$ ) is the population incidence of the given health effect, i.e., deaths per 1000 people, and ( $P$ ) the relevant exposed population for the health effect.

$$E = AF \times B \times P \quad (5)$$

For these calculations, a total of 11.5 deaths per 1000 inhabitants per year were used for the population incidence parameter  $B$ , considering what was reported in 2018 by the Federal Statistical Office of Germany.<sup>64</sup> The exposed population  $P$  was set to 200 as the approximate number of inhabitants in the village of Melpitz (personal communication municipality city of Torgau).

**Table 1. Concentration Summaries (Mean  $\pm$  Standard Deviation) of PM<sub>10</sub> Mass and Selected Constituents (OC, EC, Levoglucosan, Benzo(a)pyrene) during the Campaign<sup>a</sup>**

		winter (DJF)	spring (MAM)	summer (JJA)	fall (SON)	annual
PM <sub>10</sub> ( $\mu\text{g m}^{-3}$ )	Melpitz village	18.7 $\pm$ 11.7	19.4 $\pm$ 9.3	18.2 $\pm$ 4.8	18.4 $\pm$ 8.4	18.7 $\pm$ 8.8
	research station	17.5 $\pm$ 11.3	19.0 $\pm$ 9.2	20.1 $\pm$ 5.5	18.4 $\pm$ 8.1	18.8 $\pm$ 8.8
OC ( $\mu\text{g m}^{-3}$ )	Melpitz village	3.8 $\pm$ 3.2	3.7 $\pm$ 2.2	4.1 $\pm$ 1.7	4.0 $\pm$ 2.5	3.9 $\pm$ 2.4
	research station	3.2 $\pm$ 3.2	3.2 $\pm$ 2.1	4.3 $\pm$ 1.8	3.7 $\pm$ 2.4	3.6 $\pm$ 2.5
EC ( $\mu\text{g m}^{-3}$ )	Melpitz village	0.83 $\pm$ 0.49	0.49 $\pm$ 0.27	0.25 $\pm$ 0.12	0.72 $\pm$ 0.52	0.57 $\pm$ 0.44
	research station	0.50 $\pm$ 0.41	0.33 $\pm$ 0.24	0.22 $\pm$ 0.1	0.51 $\pm$ 0.33	0.39 $\pm$ 0.32
Levoglucosan ( $\mu\text{g m}^{-3}$ )	Melpitz village	0.25 $\pm$ 0.17	0.10 $\pm$ 0.07	0.03 $\pm$ 0.1	0.18 $\pm$ 0.18	0.14 $\pm$ 0.16
	research station	0.16 $\pm$ 0.15	0.06 $\pm$ 0.05	0.02 $\pm$ 0.11	0.14 $\pm$ 0.16	0.09 $\pm$ 0.14
BaP (ng m <sup>-3</sup> )	Melpitz village	0.87 $\pm$ 0.83	0.25 $\pm$ 0.25	0.02 $\pm$ 0.02	0.76 $\pm$ 1.16	0.47 $\pm$ 0.8
	research station	0.46 $\pm$ 0.64	0.12 $\pm$ 0.14	0.01 $\pm$ 0.01	0.54 $\pm$ 0.86	0.29 $\pm$ 0.58

<sup>a</sup>The three months included in the corresponding seasons are indicated by their first letters.

### 2.6. Melpitz Village Inhabitant Survey

To aid the interpretation of the measurement results presented in the following sections, a survey of the heating systems used by households in the village of Melpitz was conducted during the project time (2018–2019) with the support of the Saxon State Office for the Environment, Agriculture, and Geology (LfULG). The survey was constructed in a simple way with a few multiple-choice options to indicate the heating systems used in households and roughly how frequently they are used. While this level of detail did not allow for a direct connection of the survey results to the ambient measurements, it does give an approximate indication of the heating emission sources present in the village. All 63 households in Melpitz were contacted, and 46% of them replied to the survey. From these responses, it was found that 33% of the continuously running central heating systems are fueled by wood, 7% by coal, 35% by oil, and 11% by liquid petroleum gas (LPG), and 14% use emission-free noncombustion technologies. In addition to central heating, single stoves are installed in many households, contributing 12% to the total nominal heating power in the village. These stoves are mainly fueled by wood (77%) and coal (23%), and households reported to typically use them occasionally and mainly during weekends. Due to the lack of information on the remaining 54% of Melpitz households, the true shares of solid fuels for the entire village might be somewhat different. However, the results of the survey do indicate that wood and, to some extent, coal are common solid fuels in Melpitz, and it can be insightful to study their contribution to the very local air quality in the village.

## 3. RESULTS AND DISCUSSION

In the following, an overview of measured PM<sub>10</sub> mass and selected combustion-related constituent concentrations is first given. Next, the results of the PMF source apportionment are presented, including the source contributions from wood burning. Local village concentration increments are then discussed as indicators of excess pollution regarding individual PM constituents as well as total wood-burning source contributions. Finally, health impacts from exposure to PAHs and wood-burning particle mass are estimated and the wood-burning PM contributions in Melpitz are put into a broader perspective by comparing them to previous studies in Germany.

### 3.1. Concentrations at Both Sites

The concentration time series of PM<sub>10</sub> mass and selected wood-burning-related constituents are shown in Figure 2 for the entire campaign. Concentrations of further constituents are given in Figures S1–S4 in the Supporting Information.

Due to the proximity of the twin sites, the daily means of PM<sub>10</sub> mass, OC, EC, levoglucosan, and BaP generally varied in a very similar manner at both sites. Especially for EC, levoglucosan, and BaP, concentrations were clearly higher

during the colder period of the year from October to April than during the warmer months from May to September. The two main reasons for this tendency are, apparently, heating emissions and reduced atmospheric dispersion, especially due to lower mixing layer heights during the cold season. Particularly from the levoglucosan and BaP time series, the end of the heating season in early May and the beginning of the next one in late September can clearly be recognized. Accordingly, the months of June, July, and August remain with very low levels of levoglucosan and BaP. For PM<sub>10</sub> mass and OC, secondary formation in summer is an important source, which is why they show a less pronounced yearly profile.

On December 15, a large peak in the concentrations of all compounds can be seen, which especially for levoglucosan exceeds all other observed concentrations by more than a factor of 2. The high concentrations were only seen at the Melpitz village and were caused by a small Christmas market with an open fire and barbecue activities close to the measurement container. This day was therefore excluded from any further data analyses.

On July 1, another unusually large levoglucosan peak was observed at both sites in parallel, which to a lesser extent can also be seen for EC, OC, and PM<sub>10</sub> mass concentrations. On this day, a large forest fire in an area of  $\sim 5 \text{ km}^{-2}$  was actively close to Lübbtheen, Germany,<sup>65</sup> a place located approximately 230 km northwest of Melpitz. The prevailing northwestern winds during that day favored the transport of the smoke plume toward the study area.

Mean concentrations of the discussed compounds in different seasons are given in Table 1 and can generally be regarded typical for the Melpitz area.<sup>36</sup>

The concentration of BaP as a lead compound for all cancerogenic PAHs is of special interest. Annual mean values during this study were  $0.5 \pm 0.8$  and  $0.3 \pm 0.6 \text{ ng m}^{-3}$  for the Melpitz village and Melpitz research station, respectively. Both values are below the EU target value of  $1 \text{ ng m}^{-3}$ ; however, these still represent health risks that will be discussed in more detail in Section 3.7.

From both Figure 1 and Table 1, higher concentrations at the Melpitz village than at the research site can be identified, especially during the colder seasons. This observation indicates considerable emissions in the village, which add measurable amounts to the background concentrations of the PM<sub>10</sub> mass and some of its constituents. Given the absence of other strong anthropogenic sources in the small village, the local village excess pollution is likely related to residential heating emissions

and will be quantified and discussed further in the following sections.

### 3.2. PMF Source Apportionment

To quantify source contributions from wood combustion at the twin sites, receptor modeling using PMF was performed as described in Section 2.3. The final PMF solution resolved 7 factors, for which a summary of the main constituents and marker species is given in Table 2. The factor profiles with all mass and species contributions are shown in Figure S5, and a time series of factor contributions at both sites is shown in Figure S6.

**Table 2. Source Categories of PM<sub>10</sub>, Resolved by PMF Receptor Modeling<sup>a</sup>**

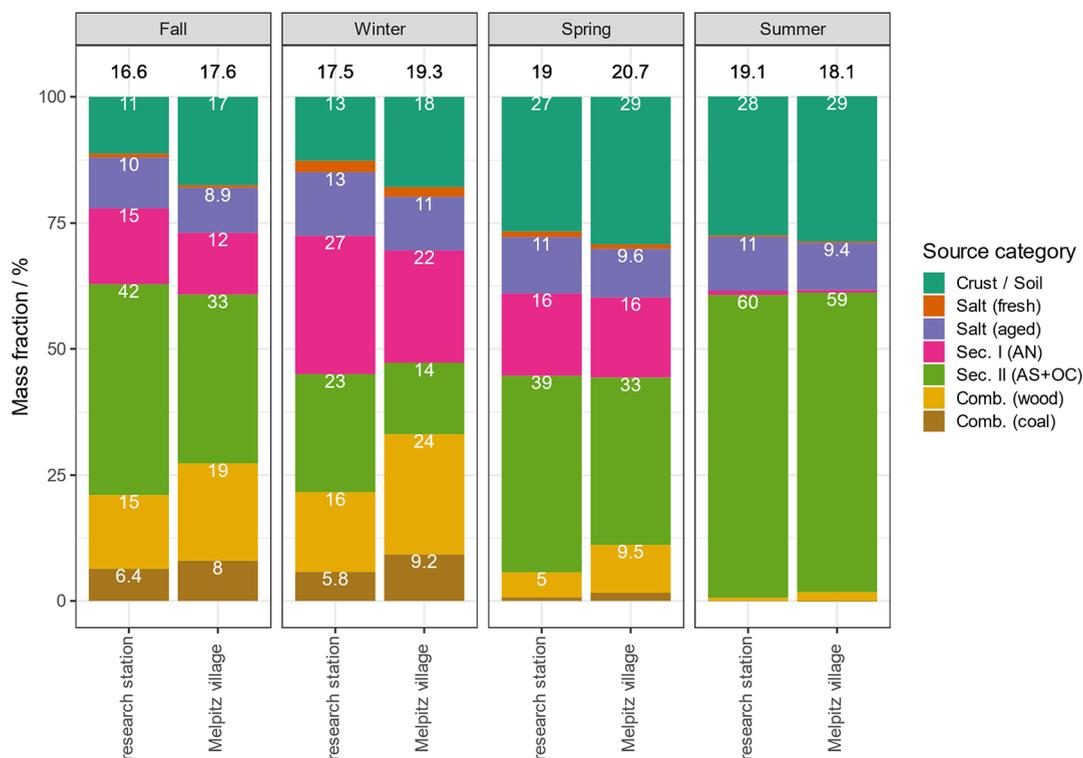
source category	main constituents	marker compounds
crustal material/soil	OC, WSOC	calcium
salt (fresh)	chloride, sodium	chloride, sodium, magnesium
salt (aged)	nitrate, sulfate, sodium	sodium, magnesium
secondary I (AN)	nitrate, ammonium	nitrate, ammonium
secondary II (AS + OC)	OC, WSOC, sulfate	OC, WSOC, sulfate, oxalate
combustion (wood)	OC, WSOC, EC, nitrate	EC, anhydrosugars, potassium, PAHs
combustion (coal)	OC, WSOC, EC, sulfate	PAHs, anhydrosugars

<sup>a</sup>AN: ammonium nitrate and AS: ammonium sulfate.

Based on the main constituents, i.e., high mass contributions in the factor profiles, as well as marker species, i.e., high species

contributions in the profiles, two natural PM sources were identified: resuspension of crustal material or soil and fresh sea salt. Aged sea salt was resolved as a mixed natural/anthropogenic source, and four anthropogenic sources were found, including the secondary formation of ammonium nitrate and mixed ammonium sulfate + organics as well as biomass combustion and coal combustion. The qualitative discrimination between wood and coal combustion might seem to carry some uncertainty as the marker species levoglucosan and PAHs can be emitted from both sources. Especially, lignite was reported to contain relevant fractions of levoglucosan and other anhydromonosaccharides,<sup>66–69</sup> limiting somewhat the usage of these compounds as specific wood-burning tracers, particularly in areas where lignite is still a commonly used fuel. This is known to be the case in some parts of eastern Europe,<sup>70</sup> from where air masses are transported to Melpitz under favorable meteorological conditions.<sup>36</sup> Additionally, the survey results (Section 2.6) suggest an impact of emissions from local coal combustion on the observed particle composition. The PMF solution and its discrimination between the two solid fuels is, however, considered plausible and robust based on (i) the much higher species contribution of potassium in the wood combustion factor, (ii) the high mass contribution of sulfate in the coal combustion factor, and especially (iii) the time series of daily source contributions of both combustion factors, indicating that the coal combustion source is mainly relevant during eastern air mass inflows, while the biomass combustion source shows more homogeneous contributions during the cold seasons (Figure S6).

Overall, the resolved sources are similar to what has previously been reported for the area<sup>11</sup> and what can typically be obtained from similar data.<sup>9,71</sup> It should be noted that for



**Figure 3.** PMF mean source contributions at the two sites during different seasons. Black numbers above the bars indicate the mean modeled PM<sub>10</sub> mass concentration in  $\mu\text{g m}^{-3}$ , while the colored bars with white numbers show the relative fractions of the different source contributions apportioned by the model.

PMF solutions with only 6 or even 8 factors, the profiles for the two combustion-related factors remained very similar to the chosen 7-factor solution. With 6 factors, the two salt-related factors combined into one single factor, and with 8 factors, the crustal/soil factor split into two.

The mean source contributions of these resolved PMF sources during different seasons at both sites are shown in Figure 3 as relative shares of total PM<sub>10</sub>. The mean absolute source contributions in  $\mu\text{g m}^{-3}$  are shown in Figure S7.

Generally, the source contributions at the regional background research station are typical for rural areas in eastern Germany.<sup>11,21</sup> Depending on the season, out of close to 20  $\mu\text{g m}^{-3}$  PM<sub>10</sub> mass concentration,  $\sim 10$ –30% originate from the resuspension of the crustal and soil material,  $\sim 10$ –15% from salt, mainly aged,  $\sim 1$ –25% from secondary particles related to ammonium nitrate,  $\sim 25$ –60% from secondary particles related to ammonium sulfate and organic material,  $\sim 1$ –15% from the combustion of biomass, and  $\sim 0$ –6% from the combustion of coal. At the village station, the contributions from crust/soil and especially from wood and coal combustion were typically higher than those at the background research station, while the contributions from secondary particles were often lower. The higher crust/soil source can likely be explained by dust resuspended from the nearby road, where cars and occasional public transport buses passed the sampling site at a distance of approximately 15 m. The higher wood and coal combustion sources are caused by residential heating in the village and will be further discussed below.

The lower contributions from secondary particles were observed, especially during fall and winter and to some extent also during spring. The differences in relative shares shown in Figure 3 translate into differences in absolute source contributions up to 0.5 and 1.4  $\mu\text{g m}^{-3}$  in the seasonal means of the Secondary I and II source categories, respectively (cf. Figure S7). These differences seem surprisingly large given the proximity of the twin sites. They remain similarly high even when removing days with easterly wind directions and thereby excluding a hypothetical impact of village emissions on the secondary particle formation at the research station (not shown). It is therefore hypothesized that they are related to different sampling artifacts of the quartz filters, operated under ambient vs temperature-controlled conditions. From the quality control filters taken every sixth day in an air-conditioned container at the research site (cf. Section 2.2.1), slightly higher seasonal mean concentrations were observed for ammonium and nitrate from the temperature-conditioned filters, which amounted to 0.26–0.56  $\mu\text{g m}^{-3}$  for the sum of both ions during the three colder seasons. As these concentration differences are very similar to the seasonal differences in Secondary I (AN) PMF source contributions between the twin sites (cf. Figure S7), they suggest that some evaporation of ammonium nitrate from the temperature-conditioned filters at the village station likely explains the different PMF results for this factor.

For the Secondary II (AS + OC) factor, however, temperature effects can likely not explain the rather large PMF source contribution differences, as for both sulfate and OC, the concentrations from the temperature-controlled quality control filters at the research station agreed within  $\pm 0.1 \mu\text{g m}^{-3}$  with the ambient research station filters. Here, differences in relative humidity (RH) together with aqueous-phase chemistry might have played a role. Both quartz fiber filters and deposited hygroscopic PM are known to retain

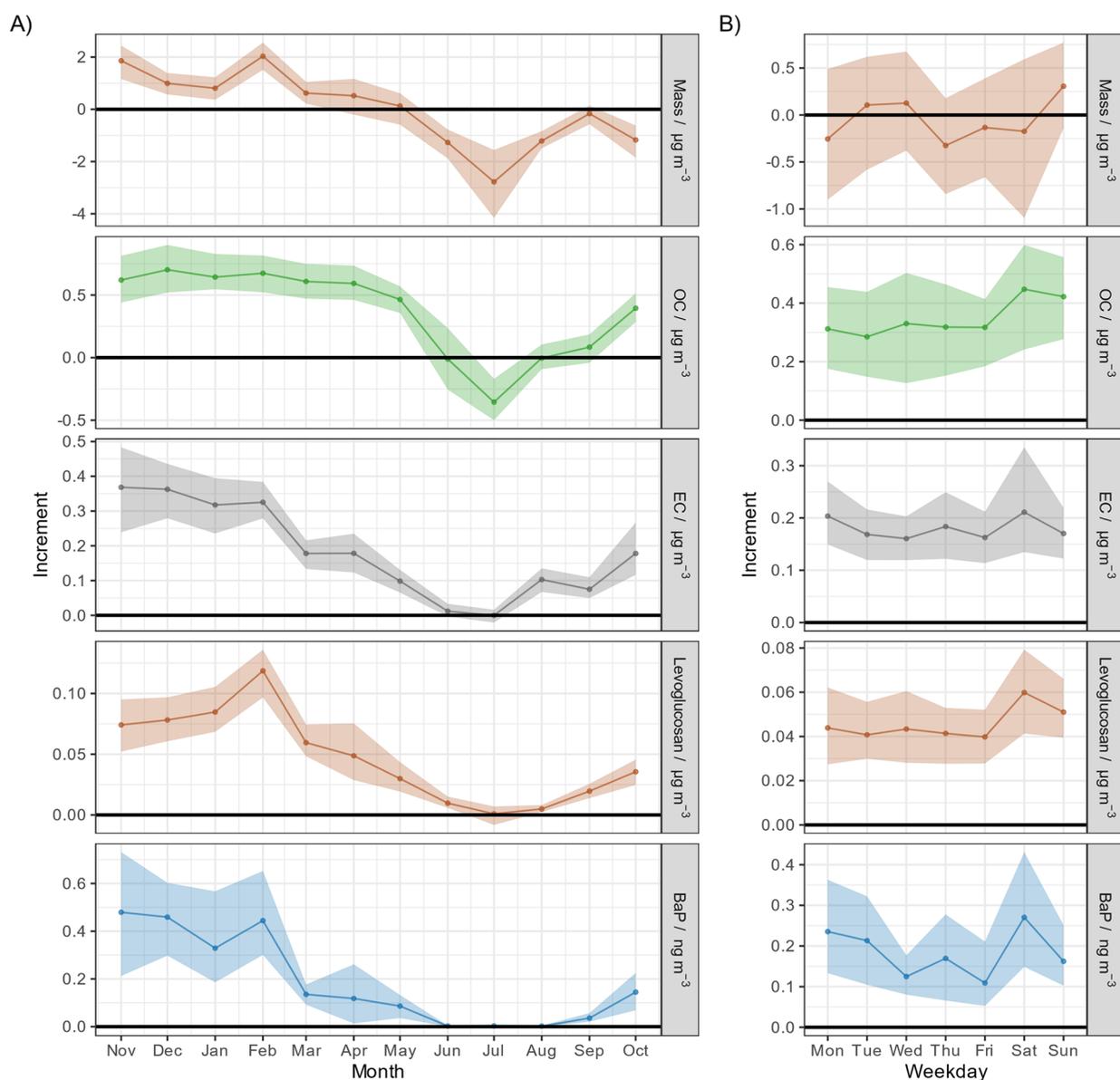
humidity,<sup>72</sup> which can lead to an aqueous phase on the filters where soluble gases such as SO<sub>2</sub> and VOCs can dissolve and potentially react. During the time of sampling or filter storage inside the Digital sampler, the dissolved gases might transform into sulfate and particle organics following well-known atmospheric aqueous phase chemistry pathways<sup>73</sup> and thereby lead to higher concentrations from filters exposed to more humid air. Ambient RH at the village site was always below the ambient RH at the research site (Figure S8) and very high RH with hourly means >90%, where water uptake to the hygroscopic material would be most efficient, occurred  $\sim 2$ –3 times less frequently than at the research site during fall, winter, and spring. When the concentration differences of the Secondary II (AS + OC) source at the two sites are plotted against the daily mean RH, a clear negative dependence can be observed, with differences typically becoming larger with higher humidities (Figure S9). More direct evidence of such an RH-related artifact is lacking in the present study. However, it would be consistent with recent observations of organosulfates strongly increasing in abundance, especially for undenuded filter samples taken at higher ambient RH due to aqueous phase reactions.<sup>74</sup>

It should be noted that the discussed sampling artifacts are related to the secondary PMF sources mainly, while the contributions from primary sources, including the combustion-related ones, are regarded as sufficiently comparable between the two sites to quantitatively assess the impact of residential wood combustion to the local PM pollution in the following sections.

### 3.3. Local Village Concentration Increments

Local concentration increments at the village site were calculated under appropriate meteorological conditions (see below) by subtracting the concentrations observed at the Melpitz research station (background) from those measured at the Melpitz village. The increments indicate additional ambient particulate pollution resulting from very local sources.

**3.3.1. Possible Biases in Increment Calculation.** Potential systematic differences from operating the filter samplers under ambient and temperature-controlled conditions at the two stations were studied with additional temperature-controlled filters at the Melpitz research station, taken in parallel to the ambient ones (Section 2.2.1). These quality control filters tended to show lower concentrations for PM<sub>10</sub> mass during winter and higher ones during summer (Figure S10). On average, the difference was  $-0.75 \mu\text{g m}^{-3}$  ( $-5\%$ ) during winter, likely resulting from partial evaporation of ammonium nitrate. Winter mean ammonium and nitrate were lower by 0.15 and 0.4  $\mu\text{g m}^{-3}$  ( $\sim 12$  and 15%) from the filters operated and stored at approximately constant 20 °C as compared to the ones at colder ambient temperatures. Winter OC was very similar between the two measurements, with only 0.12  $\mu\text{g m}^{-3}$  higher average concentration from the temperature-controlled filters. The higher summer PM<sub>10</sub> mass concentrations (1.2  $\mu\text{g m}^{-3}$  or 6.7% on average) are not related to any of the measured main constituents, i.e., ions, OC, and EC, as none of these showed similarly high discrepancies in summer (Figure S10). They might instead be related to slightly increased very local impacts of the resuspended coarse crustal material from the sand road to the research station. Under some conditions, these might have reached the container inlet more easily than the ambient Digital sampler inlet, which is located 90 m more away from



**Figure 4.** Local concentration increments in the Melpitz village for  $\text{PM}_{10}$  mass, OC, EC, levoglucosan, and BaP as (A) monthly mean values and (B) annual day of week mean values. All values calculated from the wind direction filtered data as explained in the text. Shaded areas indicate the 95% confidence intervals of the mean values.

the road. Overall, these results suggest that the below-discussed  $\text{PM}_{10}$  mass increments might be biased low in the winter and high in the summer, while they should be reasonably accurate for the other combustion-derived constituents.

Another potential systematic bias of the twin-site data could be a direct impact of the village emissions to the background concentrations measured at the Melpitz research station. Since the research station is located to the west of the village, it could be reached by emissions from the village during easterly wind directions, which would distort the calculated increments. Especially during winter, such an effect of clearly lower increments during easterly wind directions was indeed observed for  $\text{PM}_1$  mass increments (Figure S11). Therefore, both the hourly and the daily increments were filtered, and only data with mean wind directions  $>180^\circ$  or mean wind speeds  $<2 \text{ m s}^{-1}$ , i.e., calm conditions, were considered for further analyses of the local increments. While filtering the data

in such a way did have a small impact on the resulting mean increments, it should be noted that the main pattern and features discussed in the following remained very similar both for the filtered and the nonfiltered data.

The discussion of concentration increments is first done for PM mass and constituents (this section) and then for the PMF-derived wood combustion source contributions (Section 3.4).

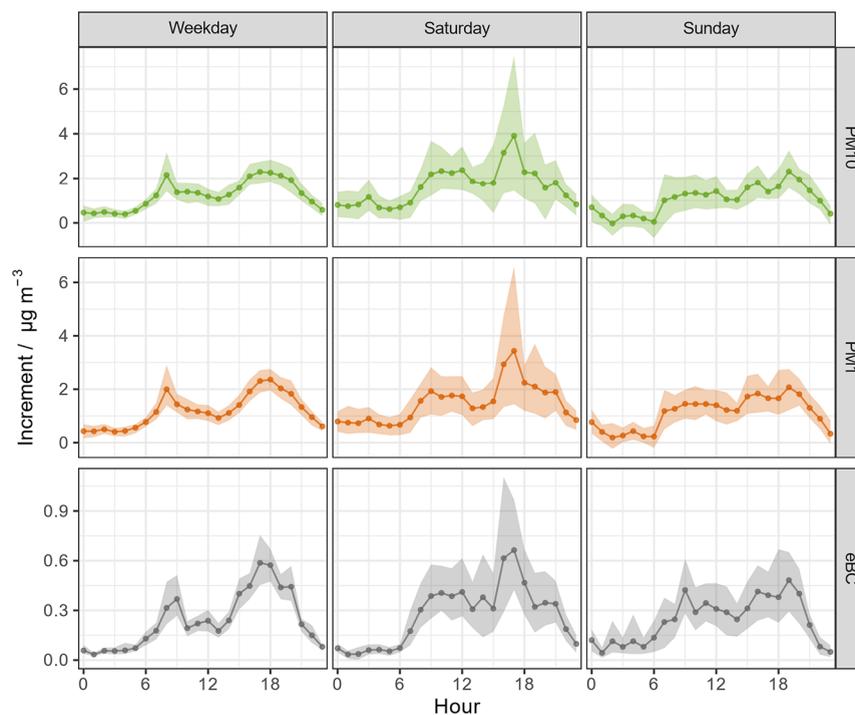
**3.3.2. Overview on Concentration Increments.** In Figure 4A, the calculated local increments in the Melpitz village are shown as monthly mean values with their 95% confidence interval for  $\text{PM}_{10}$  mass and selected constituents. The time series of all calculated daily local PM increments are given in Figure S12 for the selected constituents and in Figures S13–S16 for all the measured constituents.

As can be seen, the resulting concentration increments were positive for most of the year; i.e., the concentrations in the village were higher as compared to the background research

**Table 3. Mean Values  $\pm$  One Standard Deviation of Local Increments in the Melpitz Village for PM<sub>10</sub> Mass and Selected Constituents during Different Seasons and for the Entire Campaign<sup>a</sup>**

	winter (DJF)	spring (MAM)	summer (JJA)	fall (SON)	annual
PM <sub>10</sub> ( $\mu\text{g m}^{-3}$ )	1.28 $\pm$ 1.32	0.41 $\pm$ 1.37	-1.77 $\pm$ 1.86	0.03 $\pm$ 1.86	-0.04 $\pm$ 2.11
OC ( $\mu\text{g m}^{-3}$ )	0.67 $\pm$ 0.44	0.55 $\pm$ 0.36	-0.13 $\pm$ 0.5	0.35 $\pm$ 0.44	0.35 $\pm$ 0.54
EC ( $\mu\text{g m}^{-3}$ )	0.34 $\pm$ 0.20	0.15 $\pm$ 0.12	0.04 $\pm$ 0.09	0.19 $\pm$ 0.24	0.18 $\pm$ 0.21
levoglucosan ( $\mu\text{g m}^{-3}$ )	0.09 $\pm$ 0.06	0.05 $\pm$ 0.04	0.005 $\pm$ 0.016	0.04 $\pm$ 0.04	0.045 $\pm$ 0.051
BaP <sup>b</sup> ( $\text{ng m}^{-3}$ )	0.41 $\pm$ 0.45	0.11 $\pm$ 0.15	0.002 $\pm$ 0.011	0.21 $\pm$ 0.43	0.18 $\pm$ 0.36

<sup>a</sup>All increments are calculated using the data filtered by the wind direction, as explained in the text. <sup>b</sup>Benzo(a)pyrene.



**Figure 5.** Diurnal profiles of seasonal mean PM increments during weekdays (Monday–Friday), Saturdays, and Sundays for PM<sub>10</sub>, PM<sub>1</sub>, and eBC mass concentrations. All values were calculated from the wind direction filtered data. Shaded areas indicate the 95% confidence intervals of the means.

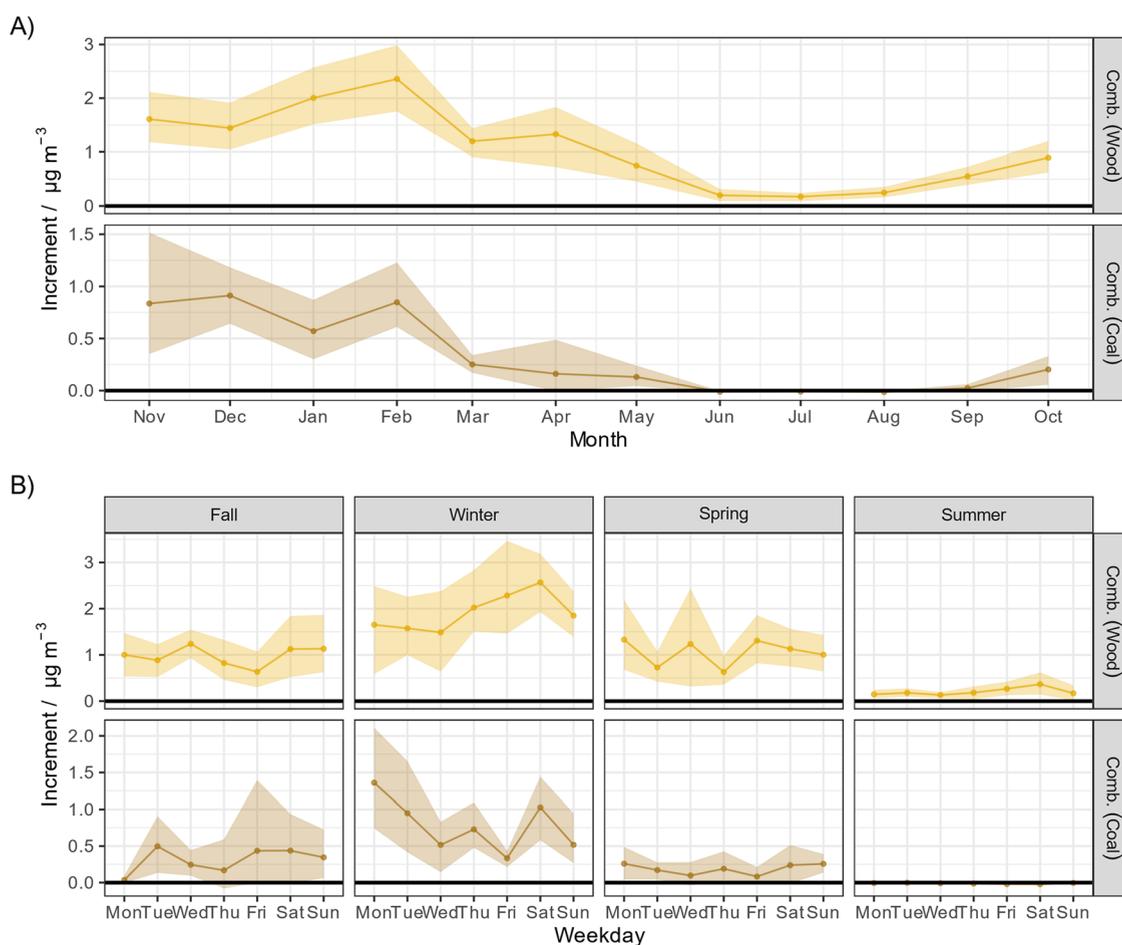
station. Only during summer, they were either not significantly different, i.e., the confidence interval includes 0, or even clearly negative for PM<sub>10</sub> and OC mass concentrations.

Seasonal and annual mean values of the local increments are given in Table 3.

**3.3.3. Seasonal Variability.** From winter to summer, PM<sub>10</sub> mass local increments decreased from an average of 1.28  $\pm$  1.32 to -1.77  $\pm$  1.86  $\mu\text{g m}^{-3}$  (average  $\pm$ 1 standard deviation). Similarly, the OC increments changed from 0.67  $\pm$  0.44 to -0.13  $\pm$  0.50  $\mu\text{g m}^{-3}$ . For the more specific combustion tracers EC, levoglucosan, and BaP, the increments were positive in winter, with mean values of 0.34  $\pm$  0.20  $\mu\text{g m}^{-3}$ , 0.09  $\pm$  0.06  $\mu\text{g m}^{-3}$ , and 0.41  $\pm$  0.45  $\text{ng m}^{-3}$ , respectively, while they were not significantly different from 0 during summer. These patterns clearly indicate an impact of combustion processes in the small Melpitz village, which increases related air pollutant concentrations in a well-observable way. Given the temporal pattern with high increments in winter, lower ones in spring and autumn, and absent ones in summer, as well as the low intensity of other combustion-related activities such as car traffic, these results suggest a significant impact of residential wood and coal combustion on the local air quality at the Melpitz village site.

The concentration increments can be compared to the regional background concentration as measured at the research station outside the village to indicate the concentration increase in relative terms. For winter, this resulted in average increases of 7.7% for PM<sub>10</sub> mass, 24% for OC, 70% for EC, 61% for levoglucosan, and 107% for BaP. As for the increment calculations, these results are based on the background concentration data filtered by the wind direction (see above). It has to be noted that this gives slightly different seasonal mean values than the ones reported in Table 1 for all days. The high relative increases for the more specific combustion tracers reinforce the importance of local wood and coal combustion emissions in the Melpitz village. Relative increases for PM<sub>10</sub> mass and OC were lower because these pollutants have a multitude of other sources in the regional background, and any additional local source in the village will have a lower share of the comparably high background concentrations.

The concentration increase of cancerogenic PAHs in the village merits special attention. Similar as observed for BaP, elevated local increments were also observed during the heating season for all other measured PAHs (Figure S16). They showed very similar patterns relative to each other and



**Figure 6.** Increment of PMF-modeled source contributions for the respective factor (monthly mean values +95% confidence interval), shown as (A) annual cycle and (B) weekly variation as a function of season. All values calculated from the wind direction filtered data.

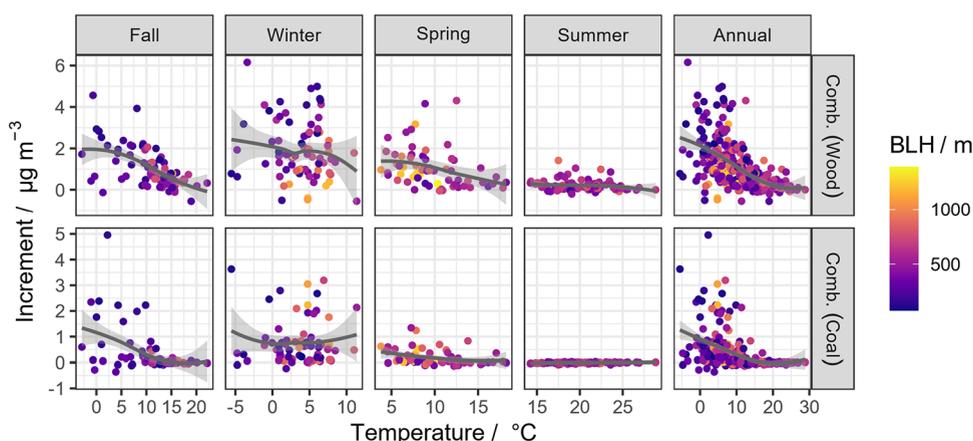
can likely be attributed to the same sources of wood and coal combustion. The increases during the heating season also have strong impacts on the annual mean values. The annual mean local increment of BaP was  $0.18 \pm 0.36 \text{ ng m}^{-3}$ , which is 79% of the mean background concentration that was determined at the research site with  $0.23 \pm 0.51 \text{ ng m}^{-3}$  for the filtered data set the increment calculation was based on. The additional contribution to regional background BaP from local heating with wood and coal in the Melpitz village is thus substantial. It makes up 38% of the total annual mean measured in the village and even the local increment alone, i.e., without contribution from regional background pollution, is 50% higher than the recommended target value by the WHO ( $0.18 \text{ ng m}^{-3}$  local increment vs  $0.12 \text{ ng m}^{-3}$  target value). Similar considerations apply to the other PAHs measured in this study, and their potential health effects are further discussed in Section 3.7.

Negative increments observed in summer for  $\text{PM}_{10}$  and OC, i.e., higher concentrations at the background research station than in the Melpitz village, indicate that contributions from other, not combustion-related sources, can be higher outside the village. Possible candidates for such sources are harvesting and agricultural activities in the vicinity of the research station, which disperse coarse particles of dust, soil, and plant material into the air that, to some extent, reached the filter samples next to the agricultural fields but did not entirely make their way to the samplers in the Melpitz village. Similar effects might have occurred during other seasons as well, which could have

diminished the  $\text{PM}_{10}$  increments during the heating season. Despite the absence of other large  $\text{PM}_{10}$  sources in the Melpitz village, the  $\text{PM}_{10}$  local increments are thus not necessarily representative of local residential heating emissions, and a more robust approach to estimate total local wood-burning contributions based on source apportionment will be discussed in Section 3.4.

Figure 4B shows the increments for the  $\text{PM}_{10}$  mass and constituents as a function of the day of the week.  $\text{PM}_{10}$  mass did not show an explicit dependence on weekdays, possibly due to the impact of other sources, as mentioned above. For OC, EC, levoglucosan, and BaP, however, an increase was observed on Saturdays, when their annual mean increments in the Melpitz village increased by  $\sim 43$ , 20, 43, and 59%, respectively, as compared to their Monday–Friday mean values. Especially for OC and levoglucosan, the local increments stay elevated during Sundays as well,  $\sim 35$  and 22% above their Monday–Friday mean values. These patterns are consistent with a temporary usage of single wood stoves, especially during weekends, as the above-given survey results indicated. Driven by emissions from central heating systems and single stove usage during the week, there are, however, clearly positive local increments for all combustion-related constituents for all days of the week.

**3.3.4. Diurnal Profiles.** Next to the filter-based measurements, hourly  $\text{PM}_1$  and  $\text{PM}_{10}$ , derived from PNSD as described above, together with the eBC measurements allow for studying



**Figure 7.** Temperature-dependence of PMF combustion PM<sub>10</sub> mass increments during different seasons and for all data. The color indicates the mean daily BLH obtained from the GDAS reanalysis data. The gray lines are locally estimated scatterplot smoothers (LOESS) to guide the eye, and the shaded areas indicate the 95% confidence intervals of the smoothing lines.

the diurnal profiles for these pollutants. They are shown in Figure 5 as annual averages during weekdays (Monday–Friday), Saturdays, and Sundays.

A bimodal cycle with higher local increments during morning and evening hours is typically observed, which is smoother during Saturdays and Sundays, presumably due to more continuous operation of the heating units as compared to other weekdays. From the diurnal profiles separated by season (Figure S17), it can be seen that the morning peak is absent in the fall, indicating a preferential usage of single stoves, especially in the evening during the transition period from warm summer to cold winter.

During the summer, there is hardly any variation of local increments throughout the Monday–Friday daytime, confirming the absence of relevant combustion sources in Melpitz other than heating. Slightly elevated increments for PM and eBC during summer Saturday and Sunday evenings (Figure S17) are likely related to recreational open fires or barbecue activities. For PM<sub>10</sub>, it is noted that all mean increments in Figure 5 are positive, opposite to the negative increments obtained from gravimetric filter PM<sub>10</sub> masses, as discussed above. This discrepancy can likely be explained by the different inlets used. The high-volume filter samples are equipped with an aerodynamic PM<sub>10</sub> head, while the APS was connected to a PM<sub>10</sub> cyclone operating at a somewhat higher-than-nominal flow rate, which led to an effective 50% cutoff diameter slightly below 10 μm. The filters might thus have sampled coarse particles occasionally that did not reach the APS.

Altogether, the PM and eBC diurnal profiles suggest a strong and very local influence of wood burning during the heating season, which will be further elucidated in the following section.

**3.3.5. Comparison of Concentration Increments with Literature.** Similar to the findings of this study, elevated daily concentrations of combustion PM components were reported for a small village in Sweden. In a residential wood-burning area, mean concentrations of PM<sub>2.5</sub> (4.9 μg m<sup>-3</sup>), EC (0.34 μg m<sup>-3</sup>), and potassium (0.22 μg m<sup>-3</sup>) during the winter season were higher than those at the reference station (PM<sub>2.5</sub> = 3.9 μg m<sup>-3</sup>, EC = 0.21 μg m<sup>-3</sup>, and K<sup>+</sup> = 0.17 μg m<sup>-3</sup>). The increments found were attributed to domestic wood burning.<sup>32</sup>

Regarding the diurnal profiles, the authors are not aware of other studies analyzing hourly changes of local PM increments

during the day. However, similar profiles as observed in the present study for the local increments are reported for total concentrations of wood-burning BC at several other sites like London<sup>75</sup> and Athens.<sup>76</sup>

### 3.4. Increments of Total Wood and Coal Combustion Source Contributions

Local increments of the PMF source contributions for the two solid fuel combustion factors were calculated in the same way as described above for the marker concentrations, i.e., subtracting research station values from village values for the wind direction filtered data set. The monthly and weekday mean values are shown in Figure 6.

In the annual cycle, the increments of the two sources have a very similar pattern, with the highest values from November to February and lowest values in summer. For coal combustion, the summer increments are zero, while for wood combustion, they remain slightly positive even during the nonheating season. As indicated by the survey results given above, only a few percent of central heating systems, which are typically used for hot water generation as well, are fueled by coal in the village of Melpitz, while about one-third of them run on wood. Emissions from these boilers are one likely reason for the small but observable summer increments of wood combustion. Another reason is the popularity of recreational fires, e.g., barbecues, especially during summer weekend nights. The impact of these can be observed from the summer weekday cycle in Figure 6B, where wood-burning increments during Fridays and especially Saturdays are clearly increased over other days of the week. A similar weekly pattern can also be observed during the winter, indicating a preferential usage of single stoves as additional heating systems during the weekend. During fall and spring, the weekend effect is less obvious, and for coal burning, it is absent as well.

### 3.5. Influence of Meteorology

In addition to seasonal and day-of-week effects, local increments from residential heating can be expected to depend on meteorological conditions, which might favor or impede the enrichment of local emissions in the village. In Figure 7, the PMF-derived combustion increments are plotted against daily mean temperatures (measured) and boundary layer heights (BLHs) (obtained from 1° × 1° reanalysis data of the NOAA Global Data Assimilation System, GDAS1).

**Table 4. Mean Values of PMF-Derived Local PM<sub>10</sub> Mass Increments for Wood and Coal Combustion in the Melpitz Village (Local Increment), Together with Corresponding Values at the Research Station (Background Concentration), Both Calculated from the Wind Direction Filtered Data Set<sup>a</sup>**

source	season	local increment / $\mu\text{g m}^{-3}$	background concentration / $\mu\text{g m}^{-3}$	local increase over background /%
wood combustion	fall	0.80	1.28	62
	winter	1.91	2.59	74
	spring	1.12	0.69	163
	summer	0.23	0.08	293
	annual	1.07	1.24	86
coal combustion	fall	0.15	0.06	241
	winter	0.78	0.66	119
	spring	0.20	0.01	1416
	summer	0 <sup>b</sup>	0 <sup>b</sup>	NA
	annual	0.32	0.22	146

<sup>a</sup>The last column indicates the mean increase of source contributions caused by local combustion emissions over the corresponding background wood or coal smoke concentrations. <sup>b</sup>Minimally negative PMF values, set to zero.

As can be seen from the annual data, both wood and coal combustion PM<sub>10</sub> increments tend to increase at daily mean temperatures below  $\sim 20$  °C, consistent with increasing emissions from residential heating at colder temperatures. The temperature effect is most pronounced during fall and spring as the demand of heating during these two transition seasons is especially variable with cold days in late fall and early spring and warm nonheating days in early fall and late spring. The winter season lacks a clear temperature dependence and shows a high variability of combustion increments instead. With mean temperatures on basically all days below 10 °C, central heating systems typically run continuously throughout winter, while single stove usage shows some day-of-week dependence as discussed above, which may explain some part of the variability. Another part might be explained by the prevailing meteorological conditions during the winter season. Except for the coldest days, winter days with high mean BLHs tend to show much smaller combustion increments than days with low BLHs, possibly because the vertical mixing of heating emissions is suppressed. However, the BLH obtained from the coarse reanalysis data set is highly correlated to locally measured wind speeds with Pearson correlation coefficients of  $\sim 0.9$  during fall, winter, and spring. Therefore, in addition to the vertical mixing, horizontal transport and dilution of emissions are likely much lower during days with a low boundary layer. Next to variable emissions, such unfavorable conditions might be a further reason for the measured wood combustion PM increments reaching daily means of  $\sim 4$   $\mu\text{g m}^{-3}$  and even higher during several winter days. Such high wood combustion increments often corresponded to  $\sim 100\%$  or more of the background wood smoke concentration resolved by the PMF for the research station, indicating that residential wood combustion, together with unfavorable meteorological conditions, can easily double the mass concentration of wood smoke in Melpitz village ambient air. For coal combustion, the local enrichment in the village was often higher even as source contributions at the research station were often very low as compared to the village increments. Clear dependencies with meteorological variables beyond temperature and BLH were not observed.

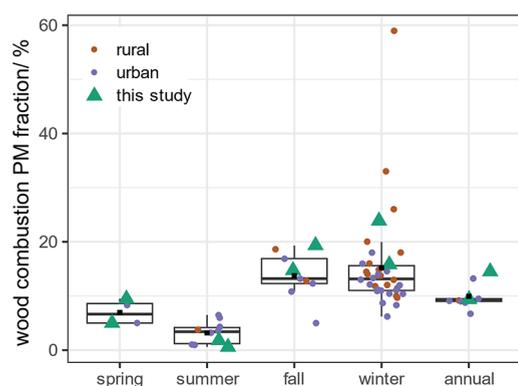
Seasonal mean values of the combustion PM increments in the village are given in Table 4 together with the relative increases they represent over the regional background source contributions.

Residential wood combustion caused an increment of  $1.9$   $\mu\text{g m}^{-3}$  PM<sub>10</sub> mass during winter 2018/19 in the village of Melpitz, which translates into a  $\sim 75\%$  increase over the  $2.6$   $\mu\text{g m}^{-3}$  PM<sub>10</sub> wood smoke source contribution observed at the regional background research station. For coal combustion, the absolute increment was  $\sim 1$   $\mu\text{g m}^{-3}$  lower than for wood combustion due to the lower share of coal as a solid fuel for heating, and the relative increase over background conditions was higher, with a value of  $\sim 120\%$ . For other seasons, the local combustion increments were lower or even absent, but relative increases over the background were similar or even higher than during winter.

### 3.6. Comparison of Wood Combustion Contributions to PM in Germany

To compare the results observed in the village of Melpitz on a larger scale, wood combustion contributions to PM in Germany were compiled from the peer-reviewed scientific as well as the gray literature, comprising mainly project reports and nonpeer-reviewed professional journals. Studies estimating wood-burning contributions to PM mass from observations in Germany only were analyzed.<sup>11,21,77–88</sup> Methods of estimation included receptor modeling, mainly PMF, or macrotracer approaches, mainly based on levoglucosan multiplied by a factor to derive total wood-burning PM. Typically, mean values for campaigns or during longer periods of the year were considered. Most studies reported data for PM<sub>10</sub>, while a few values for PM<sub>2.5</sub> were included as well. The results of the literature survey are shown in Figure 8 and represent an updated version of a similar compilation published earlier in a German journal.<sup>89</sup> It is noted that the data compilation carries substantial uncertainties as estimation methods, number of individual observations, and integration times of mean values differed substantially among the data sources. No attempt to rank or weigh the data was made. Despite these uncertainties, the compilation allows for the best-possible assessment of observation-derived wood-burning contributions to PM mass in Germany. Detailed information on the 15 selected, including the year of measurements, location, station type, season, and average wood combustion contributions, can be found in the Supporting Information (Table S1).

In Germany, the contribution of wood burning to the total PM mass ranges from an average of 3% during summer to 15% in winter, with most winter values lying between  $\sim 10$  and 20%. The average fall and spring contributions are 14 and 7%, and the annual mean is 10%. Rural values tend to be somewhat



**Figure 8.** Mean contributions of wood combustion to PM in Germany during different seasons or for the entire year. Individual mean values from different studies are shown as colored symbols and summarized by box-whisker plots, where the whiskers indicate values within the 1.5-fold interquartile range. The black squares indicate arithmetic means over all of the individual data points.

higher than urban values, with a rural mean winter contribution of  $\sim 18$  vs  $\sim 12\%$  in urban areas, which is likely related to higher  $PM_{10}$  mass concentrations in cities, especially in the coarse particle fraction from road dust resuspension or nonexhaust traffic emissions. However, the range of the values indicates that heating with wood represents a concern for air quality both in rural and in urban areas. As can be seen from Figure 8, the contribution of wood combustion to  $PM_{10}$  as measured in this study is generally in line with the available literature for Germany.

The wood-burning contributions for Germany fit well within the values compiled for different European sites.<sup>89</sup> The authors reported median contributions between 1 and 18% of the total PM, depending on the season, and annual median contribution of 12%, independent of a rural or urban station character. Of course, in individual studies, the contributions can be much higher depending on local conditions and characteristics.

While this brief literature overview corroborates the well-known and significant impact of residential wood combustion on air quality in Germany and Europe, most source apportionment studies do not allow for distinguishing between wood smoke contributions from the regional background and those from more local sources. Emissions from wood

combustion typically disperse in a larger region, and even transport over long distances can have a strong impact on the locally observed concentrations.<sup>11</sup>

In addition, monitoring sites both in cities and in rural areas are typically not located in areas where the strongest impacts of residential wood combustion can be expected because European and German policies have traditionally focused on characterizing air quality at traffic sites and in the urban and rural background (EU directive 2008/50/EC, German ordinance 39. BImSchV). In the present study, wood combustion contributions during winter were 16% in the rural background but 24% directly in the village of Melpitz (Figure 8), showing that contributions can be significantly higher within even small residential areas. In order to better estimate peoples' exposure to wood combustion PM, more measurements directly in residential areas, where people spend most of their time, would be beneficial. Ideally, such measurements would be accompanied by detailed surveys on the applied combustion technology, fuels, and activity profiles, which would facilitate extrapolation of the local results to other regions.

### 3.7. Health Risk Assessment

Given the elevated levels of wood smoke in the village, negative health impacts for the population of Melpitz and many other similar villages in the area can be expected. Such health risks are assessed in the following: first related to cancer risks based on PAH concentrations and then related to mortality based on PMF source contributions. It should be noted that the applied methodology, while suggested by the WHO, should be taken with care due to its uncertainties related to, for example, the epidemiological response functions or the exposure being estimated from ambient concentrations. Nevertheless, the results provide an indication of the possible magnitudes of health effects and are thus considered valuable. As shown above, wood combustion contributions in Melpitz were similar to those of other sites in Germany. A quantification of health effects based on the present study can thus be expected to also broadly cover the general conditions in Germany at least insofar wood burning  $PM_{10}$  contributions are available from literature.

**3.7.1. Morbidity: Excess Cancer Risk by PAHs.** Table 5 summarizes the ECR and BaP equivalent ( $BaP_{eq}$ ) values for the

**Table 5.** Summary of the PAH Health Risk Assessment, Including Mean Total PAH Concentrations, BaP Equivalents ( $BaP_{eq}$ ), and ECR Values from the Annual Concentrations and Local Increments for the Village of Melpitz for the Entire Period of Measurements

PAHs	carcinogenic group <sup>a</sup>	TEF <sup>b</sup>	annual concentrations			annual increments in the village of Melpitz			concentrations in winter		
			conc. ( $ng\ m^{-3}$ )	$BaP_{eq}$ ( $ng\ m^{-3}$ )	$BaP_{eq}$ (%)	incr. ( $ng\ m^{-3}$ )	$BaP_{eq}$ ( $ng\ m^{-3}$ )	$BaP_{eq}$ (%)	conc. ( $ng\ m^{-3}$ )	$BaP_{eq}$ ( $ng\ m^{-3}$ )	$BaP_{eq}$ (%)
BaP	1	1	0.47	0.47	63.9	0.19	0.19	66.56	0.87	0.87	61.60
BaA	2B	0.1	0.37	0.04	5.1	0.12	0.01	4.48	0.82	0.08	5.87
BjF	2B	0.1	0.33	0.03	4.5	0.12	0.01	4.39	0.68	0.07	4.82
BkF	2B	0.1	0.32	0.03	4.3	0.12	0.01	4.16	0.65	0.07	4.63
BbF	2B	0.1	0.62	0.06	8.5	0.20	0.02	7.31	1.30	0.13	9.27
DbahA	2A	1	0.06	0.06	8.2	0.02	0.02	7.66	0.12	0.12	8.50
IcpP	2B	0.1	0.41	0.04	5.6	0.15	0.02	5.44	0.75	0.07	5.30
total $BaP_{eq}$ ( $ng\ m^{-3}$ )			0.73			0.28			1.41		
ECR			$6.4 \times 10^{-5}$			$2.4 \times 10^{-5}$			$1.2 \times 10^{-4}$		

<sup>a</sup>As classified by IARC.<sup>90</sup> <sup>b</sup>TEF (toxic equivalent factor).<sup>91–93</sup>

population in the village of Melpitz, calculated with the seven PAHs measured in the present study.

The annual ECR value based on total PAH concentrations in the village is  $6.4 \times 10^{-5}$ , i.e., 6.4 cases per 100,000 people. The annual ECR value calculated from local PAH concentration increments only is  $2.4 \times 10^{-5}$ , i.e., about 40% of the total excess cancer risk in Melpitz. These ECR values are below the reported ones for Oporto, Portugal ( $2.06 \times 10^{-4}$ ),<sup>94</sup> in the same range as reported for Athens, Greece ( $4.6 \times 10^{-5}$ ),<sup>22</sup> and Florence, Italy ( $8.8 \times 10^{-5}$ ),<sup>94</sup> and above the values reported for Brno, Czech Republic, and Ljubljana, Slovenia), which ranged from  $1.83 \times 10^{-10}$  to  $2.94 \times 10^{-6}$ .<sup>95</sup>

During winter, the ECR value is  $1.2 \times 10^{-4}$ , i.e., 12 cases per 100,000 people, which is about twice as high as for the annual value and similar to the mean reported values for winter season at other European sites like Oporto ( $3.10 \times 10^{-4}$ ), Florence ( $1.21 \times 10^{-4}$ ),<sup>94</sup> Dattenhausen, Germany ( $2.26 \times 10^{-4}$ ),<sup>96</sup> and Grenoble-France ( $1.22 \times 10^{-4}$ ).<sup>97</sup>

The main contributors to the total  $BaP_{eq}$  were BaP, BbF, DbahA, and BaA, contributing to the annual average of 64, 8.5, 8.2, and 5.1%, respectively. The same pattern in the contributions was observed for winter; nevertheless, for the local increment contribution, the order was BaP, BaA, DbahA, IcpP, and BbF. Despite this, these results are consistent with the annual mean modeled  $BaP_{eq}$  emissions for Europe estimated for 2018, calculated with 16 PAHs, which shows that the most significant contribution to total  $BaP_{eq}$  is made by DbahA, followed by BbF and BaP.<sup>60</sup> It is also consistent with the values reported in studies for Athens, Greece.<sup>22</sup>

It has to be noted that ECR values aim at estimating the lifetime health risk due to long-term PAH exposure, while the present study quantifies 1 year mean values only. As in similar earlier applications, which are cited above, the presented results therefore assume long-term PAH concentrations as measured for this study's year. In reality, this is not necessarily the case, and thus, the ECR will likely be lower or higher depending on past and future developments.

**3.7.2. Mortality due to Wood Combustion PM.** An estimation of the mortality due to wood smoke in the village of Melpitz was done following the WHO methodology, as described in Section 2.6. Based on the annual PMF-derived wood combustion contribution of  $2.3 \mu\text{g m}^{-3}$  (Table 4), it results in a mortality E of 0.004, i.e., 0.004 deaths per year in the village that would be caused by wood smoke. Assuming that other villages and cities would have the same wood smoke pollution as measured in Melpitz, this would translate to 2.1 death cases per 100,000 inhabitants per year in such larger regions. This value compares in a broadly reasonable way to the PAH-derived annual excess cancer risk of 6.4 cases per 100,000 people, even though this direct comparison must be taken with care due to the different underlying epidemiological functions. The local contribution, i.e., the wood combustion concentration increment (Table 4), makes up 46% of the total mortality, while regional contributions from the background contribute 54%. Using winter-time wood smoke concentrations instead of annual mean values would roughly double the mortality to 4.1 deaths per 100,000 inhabitants with similar shares of 42% mortality caused by very local village sources and 58% from the regional background wood smoke.

**3.7.3. Health Effects in Perspective.** The ECR observed for the village of Melpitz are as high as in cities like Athens and Florence, where wood burning has been reported as a large contributor to the total PM. ECR at a rural village can thus be

the similar to large urban agglomerations where residential heating by wood burning is important.<sup>22,98</sup>

To put the estimated mortality into perspective, it is compared to the number of deaths as reported by Lelieveld et al.<sup>99</sup> These authors applied a different methodology, i.e., a global atmospheric chemistry model linked to exposure response functions to derive the premature mortality attributable to  $PM_{2.5}$  for different regions of the world and different source categories. For Germany and the year 2010, they related 8% of 34,000 deaths to the residential energy sector. Using a total population of 81.7 Mio in 2010 Germany,<sup>100</sup> this translates to a rate of 3.3 deaths per 100,000 inhabitants. Considering the broader source sector, i.e., residential energy vs residential wood combustion, the different scales, i.e., entire Germany vs local Melpitz, and the different methodologies, this rate seems reasonably close to the value of 2.1 per 100,000 derived from Melpitz and corroborates our above-given conclusion on the wood smoke pollution in Melpitz being not too far off of other regions in Germany.

Lelieveld et al.<sup>99</sup> report premature mortality in Germany for other source categories as well, including agriculture, power generation, and land traffic with mortality rates of 18.7, 5.4, and 8.4 per 100,000, respectively (converted from the published data as described above). Accidental deaths due to traffic accidents were reported for Germany in 2018 as 4.4 per 100,000 inhabitants.<sup>64</sup> Compared to the present study, this indicates that residential wood smoke at the level as measured in Melpitz would, statistically, cause 25% of premature mortality from average traffic pollution (in 2010) or about 50% of the traffic accident mortality in 2018.

About half of the mortality from wood smoke in the village of Melpitz is derived from very local sources, as discussed above. This implies that policies even at the municipal level could lead to a significant improvement, which is in contrast to most other sources, where abatement on small local scales is either not possible (e.g., agriculture or power generation) or hardly practicable (e.g., traffic).

Biomass burning PM is well documented to have further health effects, beyond the cancer risk and all-cause mortality considered here.<sup>62,63</sup> Many of the health studies are based on large wildfire burning events, however, and health risks from exposure to residential heating PM in developed countries are very scarce.<sup>62,63</sup> More detailed assessments of the toxicity of residential wood smoke particles in contrast to other PM sources and total PM would be beneficial for more accurate assessments of its health impacts.

## 4. CONCLUSIONS

The increasingly popular usage of wood as an allegedly climate-friendly and relatively cheap fuel for residential heating poses a substantial challenge to air quality control as emissions especially of particulate pollutants are typically orders of magnitude higher than for modern gas- or even oil-fueled heating devices. Evidence of wood combustion contributions to total PM mass in the range of  $\sim 10$ –20% during winter has been reported for Germany in many mainly episodic measurements. The full extent of the problem is difficult to assess because, following European regulation, air quality monitoring stations typically represent either heavily traffic-impacted or urban and rural background conditions. Long-term operating stations located in residential areas, where people live and the impacts of domestic heating are highest, are

largely lacking. In addition, the chemical PM speciation needed to estimate wood combustion contributions is largely absent in monitoring networks, as well.

The present study quantifies the impacts of residential heating with wood on the local concentrations of PM<sub>10</sub> mass based on a carefully designed twin-site approach. The results indicate substantial excess pollution in a residential area over the background conditions, measured just outside the residential area. This suggests that the exposure of people to wood combustion PM is likely larger than can be estimated from the predominantly available background measurements alone.

Estimations of health impacts in the present study demonstrate an excess cancer risk from PAHs similar to that of larger urban agglomerations, and calculations based on total PM response functions indicate a mortality from short-term exposure to wood smoke PM about half as high as from traffic accidents. About 40–50% of these health risks originate from the very local excess pollution and would not necessarily need national or international policies to be effectively mitigated.

The annual local wood combustion PM concentration increment was determined to be  $\sim 1 \mu\text{g m}^{-3}$  in the village of Melpitz. Considering that wood combustion emits mainly fine particle mass,<sup>101</sup> this exclusively locally originated pollution alone would make up 20% already of the recently lowered WHO long-term AQG annual levels for PM<sub>2.5</sub>.<sup>102</sup> Together with the contribution from the rural background, wood combustion PM in the village of Melpitz reached nearly 50% of the WHO recommendation of a maximum of  $5 \mu\text{g m}^{-3}$  PM<sub>2.5</sub> annual mean concentration.

More long-term, chemically as well as spatially resolved PM monitoring in residential areas would be beneficial for obtaining a more accurate picture of this emerging PM source. This applies to both rural and urban residential areas, where especially small stoves as secondary heating devices have seen widespread application as well. Such monitoring would allow for better assessments of emissions from such devices and especially their impact on local air quality under real-world usage as opposed to laboratory conditions during device certification, often leading to optimistic emission values. It could also help in improving the validation of emission inventories and associated source-oriented spatial modeling as an important prerequisite for larger scale exposure calculations. Finally, it would allow for dedicated health effect studies of wood smoke particles from residential heating, which might differ from more widely studied wildfire biomass burning events due to different concentration levels and exposure times. Longer-term twin-site studies similar to the present one would constitute a useful approach to accompany such future developments, allowing for realistically evaluating their impacts both in residential areas on a smaller scale and under representative background conditions on a larger spatial scale.

Overall, measures to reduce residential wood combustion emissions are desirable and of considerable health interest since they occur mainly where people are present or at home. Further mitigation efforts are needed, which might include refinements of existing legislative regulations and technological advances.

## ■ ASSOCIATED CONTENT

### Data Availability Statement

The data underlying this study are available from the corresponding author upon reasonable request.

### SI Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acsenvironau.3c00035>.

Additional figures on concentrations, increments, PMF results, and meteorological data and wood-burning contributions from earlier studies in Germany (PDF)

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### Notes

The authors declare no competing financial interest.

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