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Estimation of Carbonaceous Aerosol Sources under Extremely Cold Weather Conditions in an Urban Environment

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Abstract: The present study investigated the characteristics of carbonaceous species in an urban background site. Real-time measurements of inorganic (sulfate, nitrate, ammonium, chloride, and black carbon [BC]) and organic submicron aerosols (OA) were carried out at the urban background site of Vilnius, Lithuania, during January–February 2014. An aerosol chemical speciation monitor (ACSM, Aerodyne Research Inc., Billerica, MA, USA) and co-located 7- λ aethalometer (AE-31, Magee Scientific, Berkeley, CA, USA) were used to analyze the chemical compositions, sources, and extinction characteristics of the PM₁. Extremely contrasting meteorological conditions were observed during the studied period due to the transition from moderately cold (~ 2 °C) conditions to extremely cold conditions with a lowest temperature of -25 °C; therefore, three investigation episodes were considered. The identified periods corresponded to the transition time from the moderately cold to the extremely cold winter period, which was traced by the change in the average temperature for the study days of 1–13 January, with $T = -5$ °C and $RH = 92\%$, in contrast to the period of 14–31 January, with $T = -14$ °C and $RH = 74\%$, and the very short third period of 1–3 February, with $T = -8$ °C and $RH = 35\%$. On average, organics accounted for the major part (53%) of the non-refractory submicron aerosols (NR-PM₁), followed by nitrate (18%) and sulfate (9%). The source apportionment results showed the five most common OA components, such as traffic and heating, to be related to hydrocarbon-like organic aerosols (HOA_{traffic} and HOA_{heating}, respectively), biomass-burning organic aerosols (BBOA), local organic aerosol (LOA), and secondary organic aerosol (SOA). Traffic emissions contributed 53% and biomass burning 47% to the BC concentration level. The highest BC and OA concentrations were, on average, associated with air masses originating from the southwest and east-southeast. Furthermore, the results of the PSCF and CWT methods indicated the main source regions that contributed the most to the BC concentration in Vilnius to be the following: central-southwestern and northeastern Poland, northwestern-southwestern and eastern Belarus, northwestern Ukraine, and western Russia. However, the potential sources of OA were widely distributed.



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1. Introduction

Carbonaceous aerosols are the major components of fine particulate matter (PM_{2.5}), and they can create challenges for the urban environment due to their variability in chemical and physical properties and originating from a wide range of pollution sources [1,2]. Carbonaceous aerosols have negative impacts on air quality and human health [3]. Carbonaceous PM_{2.5} is five times more hazardous than PM_{2.5} of inorganic origin (nitrate, sulfate, ammonia) [4–6]. Carbonaceous aerosols make up a significant portion (about 20–70%) of atmospheric aerosols, and they play a crucial role in haze formation, visibility degradation, adverse health effects, and atmospheric heating [7]. Carbonaceous aerosols primarily consist of organic carbon (OC), elemental carbon (EC), and water-soluble ions [8]. OC can be divided into two categories: primary organic aerosol (POA) and secondary organic aerosol (SOA), the latter of which is produced by the condensation of oxygenated volatile organic

compounds (OVOCs) or the atmospheric oxidation of primary organic aerosol. Both fossil and non-fossil sources, such as the burning of coal and automobile exhaust, can release the precursors to POA and SOA (e.g., biomass burning, vegetation emission, and cooking) [9]. The refractory light-absorbing fractions of carbonaceous aerosols are commonly elemental carbon (EC) (when assessed using thermal optical methods) and black carbon (BC) (when quantified using light absorption techniques) [10]. OC contributes a major portion of about 90%, while a relatively very low portion of about $\leq 10\%$ is contributed by EC to carbonaceous aerosols [10,11]. Due to having a graphite-like structure, EC is a major contributor to light absorption, while OC is a mixture of thousands of particulate organic compounds, including polycyclic aromatic hydrocarbon (PAHs), polychlorinated dibenzo-p-dioxins, dibenzofurans (PCDD/Fs), and other hazardous components, which can harm human health and increase morbidity and mortality [12–14].

The concentration of OC and EC in atmospheric aerosols is regulated by numerous sources (natural and anthropogenic) as well as by the formation of secondary organic carbon affected via local meteorology. This causes significant seasonal and diurnal fluctuations in the observed concentrations. The examination of carbonaceous aerosols under severely cold weather conditions reveals the effect of the major carbon sources on particulate matter [15]. Studies conducted in Athens (Greece) by Diapouli et al. (2017) [16] revealed that the BC concentrations were greater during the winter season due to wood burning as compared to the warm season. Based on the findings of research conducted by Klejnowski et al. (2017) [17] in Cracow (Poland), it was discovered that the concentrations of carbonaceous material (OC, EC, and levoglucosan) were greater when the weather was severely cold. Additionally, a study by Minderyte et al. (2022) [18] conducted in Vilnius (Lithuania) showed that the carbonaceous aerosol concentration is higher in the cold season compared to the warm season because there is less need for residential heating.

The IPCC's special report, Global Warming of 1.5 °C, recognized the significance of BC as the second-largest anthropogenic contributor to global warming after CO₂ [19,20]. BC is produced as primary particles through incomplete combustion, particularly from gasoline and diesel-fuel vehicles, and it is one of the major sources of pollution in urban environments in many European countries [21–25]. Due to their substantial contribution (90%) to BC emissions, diesel engine vehicles have drawn particular attention in urban areas [26]. In addition to diesel engines from the transportation sector, the primary sources of BC in the atmosphere are residential heating (using fossil or biomass fuels) and biomass burning [27]. In recent years, residential wood burning has become a more popular alternative energy source to fossil fuels. The promotion of CO₂-neutral regulations, greater taxes on heating fuel, and the financial crisis, which has had a considerable impact on the average household income, have all led to a rise in the use of biomass as a heating source recently [28]. In this regard, wood smoke may represent a significant source of gaseous and particle pollution that adversely affects both urban and rural areas. This is crucial in the winter, when the weather and increased use of biomass burning could have a negative and considerable influence on urban air quality, exposing huge populations to pollutant concentrations that are over regulatory limits [29]. Emissions from biomass burning have been identified as a significant source of air pollution in Central and Northern Europe [30]. In addition to burning biomass for home heating, burning agricultural waste is another prevalent practice that contributes significantly to the release of gaseous and particle pollution into the atmosphere [30].

Several regulations and policies have been put in place to help reduce the effects of climate change [31]. These include stricter standards for testing vehicle emissions, such as the EURO 6 European Emission Standards (Commission Regulation (EU) 2016/646 2016), and programs to reduce the use of diesel cars in cities, such as low emission zones, all the way up to the complete phase-out of fossil fuel vehicles by 2035 [32–36].

The European Union has established laws designed to reduce PM emissions in the region, particularly from vehicles and large point sources [37]. Emissions from home solid fuel appliances have been on the rise due to a lack of regulation, although the eco-design

guideline is anticipated to reverse this trend [38]. In the year 2000, transportation was responsible for roughly 45% of all BC emissions in the EU, but by the year 2019, that number had dropped to 30%. Since the year 2000, there has been a significant reduction in PM and BC emissions across the EU-28. Emissions of PM_{2.5}, PM₁₀, and BC decreased by 36%, 32%, and 48%, respectively [32].

In order to enhance air quality and reduce individual exposure to harmful air pollutants, it is imperative to pinpoint specific regional sources of carbonaceous aerosol pollution and create mitigation strategies. The promotion of biofuels as a low-carbon substitute could result in larger black carbon emissions than those from the combustion of fossil fuels, making the exhaust emissions from the burning of biofuels potentially the second largest source of global-warming emissions.

This study aims to investigate the chemical composition of carbonaceous aerosols in an urban environment, to quantify the source contributions, and to evaluate the contribution of remote sources to the mass of carbonaceous species.

2. Materials and Methods

2.1. Site Description

An intensive 2-month field measurement campaign was performed in an urban background site in Vilnius (Lithuania) during the winter of 2014 (January–February). Continuous real-time measurements of the OA composition and BC mass concentrations were carried out on the top floor of the academic building of the Center for Physical Sciences and Technology campus located in Vilnius (54°38' N, 25°10' E, 197 m asl). The inlet of the sampling system was placed on the top floor, about 20 m above ground level, 12 km southwest of the downtown area. The sampling site was relatively far from densely populated residential areas. The station was surrounded by forests to the north/northeast, and by villages to the south/east. The nearest roadway was 300 m to the southwest, while on the opposite side, a low traffic road was 600 m away. The location can be described as an urban background (Figure 1). Under normal meteorological conditions, the potential for the accumulation of vehicle emissions is limited by the site location.

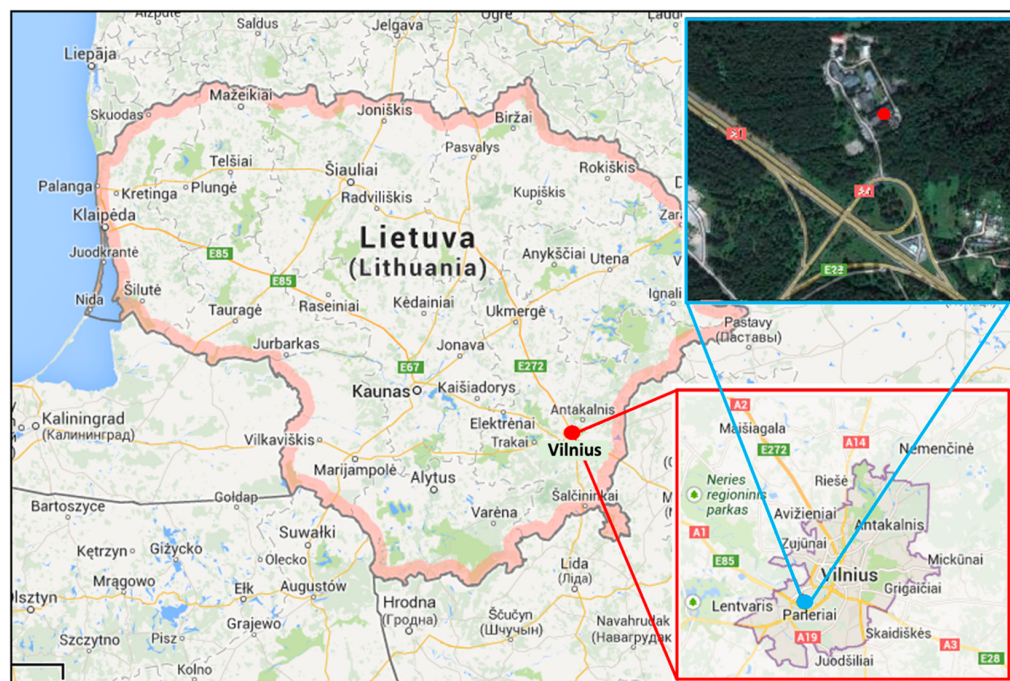


Figure 1. Map of the observation site in Vilnius (indicated by the red marker) (source of maps: free within www.maps.lt (accessed on 10 December 2022)).

2.2. Instrumentation

2.2.1. Organic Aerosol

An Aerodyne Research, Inc. (Aerodyne Research, Inc., Billerica, MA, USA) Aerosol Chemical Speciation Monitor [39] was deployed at an urban background site in Vilnius (Figure 1) with the aim of achieving real-time continuous monitoring of organics, SO_4^{2-} , NO_3^- , NH_4^+ , and Cl^- .

The sampled air passed through a Nafion dryer (MD-110-48S-4, PermaPure LLC, Toms River, NJ, USA) and through a ~ 2.65 m long stainless-steel sampling tube (6 mm i.d.) before reaching the ACSM inlet. Aerosol particles with an aerodynamic diameter in the range of 40–1000 nm were sampled at a flow rate of $\sim 1 \text{ l min}^{-1}$ into the ACSM through a 100 μm critical orifice mounted at the inlet of an aerodynamic lens. The particles were then directed onto a resistively heated surface where the NR-PM₁ components were flash vaporized on impaction with the vaporizer, which was typically operated at $\sim 600^\circ\text{C}$. The generated vapors were then ionized by 70 eV electron impact. The ACSM was operated at a time resolution of ~ 28 min for typical aerosol loadings, with a scan rate of 220 ms amu^{-1} from m/z 10 to 140. A detailed description of the ACSM has been given by Ng et al. (2011) [39,40]. The time series of organic mass spectra were processed using multivariate factor analysis methods to extract the distinct factor profiles further associated with aerosol sources. A graphical user interface, SoFi (Source Finder) [41], developed at the Paul Scherrer Institute was used to perform the source apportionment of the NR-PM₁ OA mass spectra collected during January–February 2014. A collection efficiency (CE) of 0.5 was used.

2.2.2. Black Carbon

A Magee Scientific Company Aethalometer (Model AE31, Magee Scientific, Berkeley, CA, USA) was deployed at the site, which provided real-time, continuous measurements of the BC mass concentration. The optical transmission of the carbonaceous aerosol particles was measured at 7 wavelengths ranging from 370 nm to 950 nm (370, 450, 520, 590, 660, 880, and 950 nm). In this regard, 880 nm is considered to be the standard channel for BC measurements [42,43]. Using the assumed mass absorption efficiency, the output of the aethalometer was evaluated directly as the BC mass concentrations. It was investigated whether attenuation in the aethalometer was also affected by the multiple scattering of light from the filter due to the shadowing effect [44,45]. The aethalometer converted the light attenuation into the BC mass using a fixed specific attenuation cross-section (σ) of $16.6 \text{ m}^2 \text{ g}^{-1}$ of BC (*Aethalometer Operations Manual*, Magee Scientific) set for a wavelength of 880 nm. The 5-minute time-step BC measurements were averaged to obtain the 1 h means. The aethalometer was equipped with an impactor that removed the particles with a particle aerodynamic diameter higher than $2.5 \mu\text{m}$. The detection limit of the aethalometer was $\pm 100 \text{ ng BC m}^{-3}$, which is typically considered sufficient for the total BC concentration in urban environments ($1\text{--}10 \mu\text{g m}^{-3}$).

The absorption coefficient b_{abs} for airborne particles is defined by the Lambert–Beer law as follows:

$$I = I_0 \cdot e^{-b_{abs} \cdot x}, \quad (1)$$

where I is the intensity of the incoming light and I_0 is the transmitted light intensity after passing through a medium with thickness x . A power law fit is commonly used to describe the wavelength dependence of the aerosol absorption coefficient:

$$b_{abs} \propto \lambda^{-\alpha} \quad (2)$$

where λ is the wavelength and α is the Ångström exponent of the absorption coefficient (the wavelength's dependence on the optical depth). The meteorological factors (temperature, relative humidity, wind speed, and wind direction [T , RH , WS , and WD , respectively]) were supplied by a site belonging to the Environmental Protection Agency (EPA) less than 1 km from our monitoring site.

2.3. Source Apportionment Techniques

2.3.1. Organic Aerosol Source Apportionment

The ACSM data were analyzed in terms of the aerosol chemical composition and mass concentrations using the standard ACSM data analysis software (version 1.5.3.0) written in Igor Pro (WaveMetrics, Inc., Oregon, USA) $m/z < 120$ [46,47], which was also used for the PMF analysis [48–50]. A positive matrix factorization (PMF) model was used to describe the ACSM measurements (ACSM organic mass spectra as a function of time) as a linear combination of several static factors. The PMF algorithm iteratively solves the equation:

$$X = GF + E \quad (3)$$

where X is the data matrix, G and F represent the factor profile and factor time series matrices, respectively, while E is the residual matrix.

2.3.2. Black Carbon Source Apportionment

The source apportionment technique relies on the observation of distinct absorption spectra at various wavelengths of light from individual particles. For this purpose, Wein-gartner et al. (2003) [44] proposed and implemented a correction to reduce “shadow”, multi scattering, and other effects causing measurement biases. To assign the BC mass concentration to biomass burning (BC_{bb}) and fossil fuels (BC_{ff}), the origin aethalometer model proposed by Sandradewi et al. (2008) [51] was applied. For selected wavelengths (470 and 880 nm), this method uses the absorption Ångström exponent (AAE) as follows:

$$b_{abs,BC_{ff}}(880nm) = \frac{b_{abs}(470nm) - \left(\frac{470}{880}\right)^{AAE_{ff}}}{\left(\frac{470}{880}\right)^{AAE_{bb}} - \left(\frac{470}{880}\right)^{AAE_{ff}}} \quad (4)$$

$$b_{abs,BC_{bb}}(880nm) = \frac{b_{abs}(470nm) - \left(\frac{470}{880}\right)^{AAE_{bb}}}{\left(\frac{470}{880}\right)^{AAE_{ff}} - \left(\frac{470}{880}\right)^{AAE_{bb}}} \quad (5)$$

In this work, the absorption Ångström exponent (AAE) values were selected on the basis of previous studies by Zotter et al. (2017), Quin et al. (2018), Paurait et al. (2020), and Minderytė et al. (2022) [52–54]: absorption Ångström exponent for fossil fuel (AAE_{ff}) = 0.9, absorption Ångström exponent for biomass burning (AAE_{bb}) = 1.68, and absorption Ångström exponent for black carbon (AAE_{BC}) = 1.43. The wavelength dependence method (WDA) of Quin et al. (2018) [53] was used to investigate the AAE of brown carbon (BrC):

$$b_{abs,BC}(370nm) = b_{abs}(880nm) - \left(\frac{880}{370}\right)^{AAE_{BC}} \quad (6)$$

$$b_{abs,BrC}(370nm) = b_{abs}(370nm) - b_{abs,BC}(370nm) \quad (7)$$

This method assumes that light absorption at 370 nm could be related to both BC and BrC light absorption, while light absorption at 880 nm represents only BC.

2.4. Air Mass Backward Trajectories

The TrajStat software includes trajectory statistics and a geographical information system proposed by Wang et al. (2009) [55]. A potential source contribution function (PSCF) model represents the proportion of pollution trajectories in a study area. The higher the value of weighted PSCF (WPSCF), the greater the impact on the concentration of an object [56]. However, it cannot illustrate the trajectories with certain pollution

levels. Therefore, the concentration-weighted trajectory (CWT) method developed by Hsu et al. (2003) [57] was used for this purpose by calculating the weighted trajectory concentration [55]. The aim was to identify the pathways of the 72 h air masses with higher concentrations of OA and BC that arrived at Vilnius in January and February 2014, as well as the geographical source regions of OA and BC, using three trajectory analysis methods: clustering analysis, PSCE, and CWT. The grid covered a study domain between 35–75° north latitude and −30–75° east longitude with a $0.3^\circ \times 0.3^\circ$ resolution. The NCEP/NCAR Reanalysis Archive for global scale analysis provided the meteorological data for the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model [58] in order to calculate the air masses' backward trajectories arriving every hour (00–23 UTC). The arrival height of the backward trajectories was 20 m agl.

3. Results

3.1. Overview

The time series of NR-PM₁ species, aerosol BC mass concentration, PMs, gases, and meteorology parameters for the entire period are shown in Figures 2 and 3. The total PM₁ mass concentration of the non-refractory species measured by the ACSM varied from a few $\mu\text{g m}^{-3}$ to $77.5 \mu\text{g m}^{-3}$. During the campaign, OA comprised the major fraction of NR-PM₁, accounting for 53% ($10.1 \mu\text{g m}^{-3}$) on average, with ammonium and nitrate being the second largest (18% [3.4 and $3.4 \mu\text{g m}^{-3}$, respectively]). Sulfate accounted for 9% ($1.8 \mu\text{g m}^{-3}$) of the total NR-PM₁ mass, while chloride represented a minor fraction, ~2% ($0.3 \mu\text{g m}^{-3}$). The high contribution of ammonia mainly came from the agriculture sector [59]. Thus, it can be concluded that ammonia was long-range transported to the measurement site from more distant locations [51]. The nitrate mainly came from oxidation processes of gaseous NO_x onto pre-existing particles or through new particle formation [58]. It can be concluded that the nitrate content reflects the contribution of transport exhaust to the chemical makeup of aerosol particles, as there are no other significant sources of NO_x in Vilnius [51]. The exceptional meteorological conditions also influenced the increase in pollutant emissions from combustion processes, contributed to the changes in the composition of the carbonaceous particles, and resulted in changes in their transport characteristics. During the monitoring periods, a maximum temperature of 2 °C and a minimum temperature of −25 °C were measured. The last weeks of January 2014 were unusually cold, such that a record low temperature was observed (Figure 3), with a mean relative humidity of 80% (min 32%, max 97%) (Figure 3).

In order to understand the behavior of the atmosphere during the days with a high PM₁ concentration, the measurement period was divided into days with and without episodes. The first two episodes (Ep. 1 and Ep. 2) corresponded to the transition time from a moderately cold winter to an extremely cold winter period, which was evidenced by the change in the average air temperature for the 1–13 January days of campaign, $T = -5^\circ\text{C}$, $\text{RH} = 92\%$, in contrast to the 14–29 January days, $T = -14^\circ\text{C}$, $\text{RH} = 74\%$.

The first episode (Ep. 1) during 1–13 January was characterized by lower concentrations of organic matter (47%, $6.2 \mu\text{g m}^{-3}$). After a slow gradual temperature decrease, on 14 January extremely low temperatures all day long were observed, together with a wind direction from the N–NE–NW sectors with a mean wind speed of 1.5 m s^{-1} (Figure 3). During Episode 1, the mean mass concentration of the pollutants (SO₂— $1.8 \mu\text{g m}^{-3}$, NO_x— $14.0 \mu\text{g m}^{-3}$, PM₁₀— $11.0 \mu\text{g m}^{-3}$, and BC— $1.5 \mu\text{g m}^{-3}$) was significantly lower compared to Episode 2 (SO₂— $3.4 \mu\text{g m}^{-3}$, NO_x— $20.1 \mu\text{g m}^{-3}$, PM₁₀— $18.6 \mu\text{g m}^{-3}$, and BC— $3.5 \mu\text{g m}^{-3}$). The fraction of sulfate, nitrate, and ammonium increased from 8% to 10%, 15% to 16%, and 13% to 20%, respectively, in Episode 2 as compared to Episode 1. According to Calvo et al. (2013) [60], the decrease in ammonium occurred due to the neutralization of HNO₃ and H₂SO₄ with NH₃ likely forming ammonium-derived aerosol in the form of NH₄NO₃ and (NH₄)₂SO₄. During the low pollution episode (Ep. 3), the average relative humidity suddenly dropped to 53%, while the wind speed was relatively high, with a mean value of 2.4 ms^{-1} .

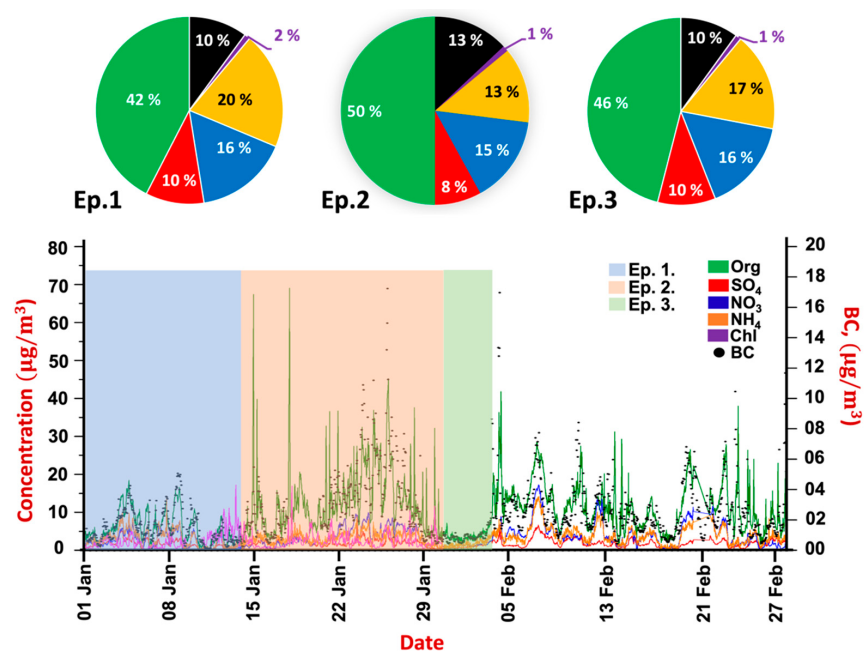


Figure 2. Temporal variation of species (organics, sulfate, nitrate, ammonium, and chloride) and aerosol black carbon. The pie charts show the fractional abundances of individual BC and ACSM species averaged over the episodes.

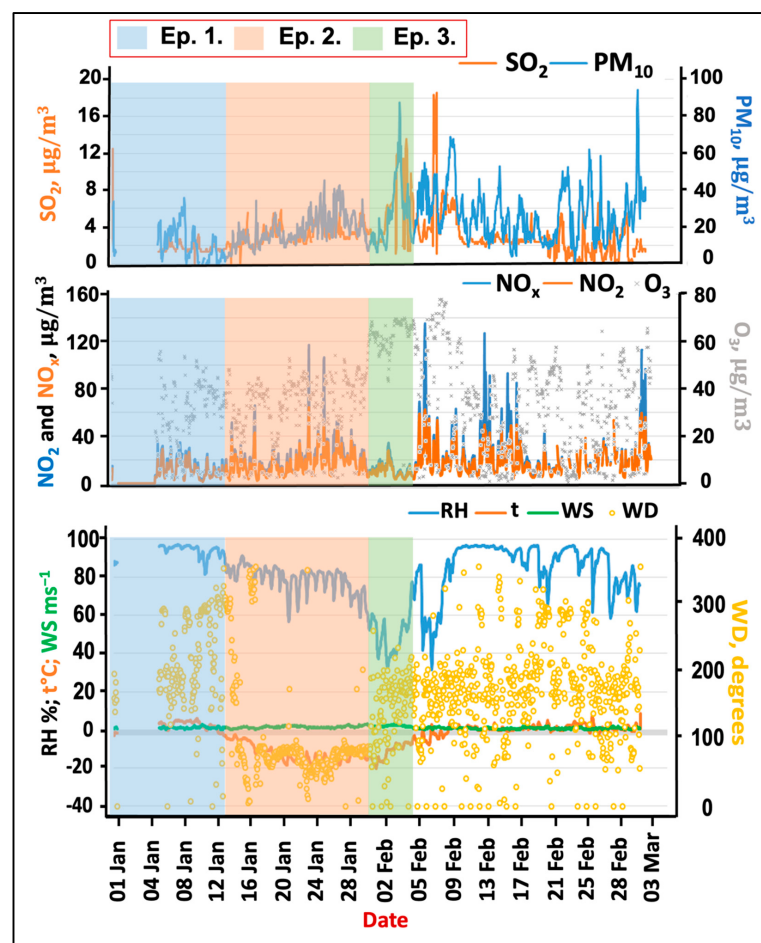


Figure 3. Time series of SO₂, PM₁₀, NO₂, NO₃, NO_x, ambient temperature, wind direction (dots), wind speed, and humidity.

3.2. Source Apportionment of Ambient Black Carbon

The source apportionment of BC from fossil fuel and biomass burning was analyzed using the AAE_{ff} and AAE_{bb} values obtained in Section 2.3.1. Figure 4a depicts the time series of BC_{ff} and BC_{bb} , as well as their contributions to the total BC for the whole measurement campaign. The percentage contribution showed that BC_{ff} and BC_{bb} contributed 53% and 47% to the total BC mass concentration, respectively. The highest contribution of BC_{ff} was observed for the whole measurement campaign, and it was associated with fossil fuel-related pollution from on-road transport.

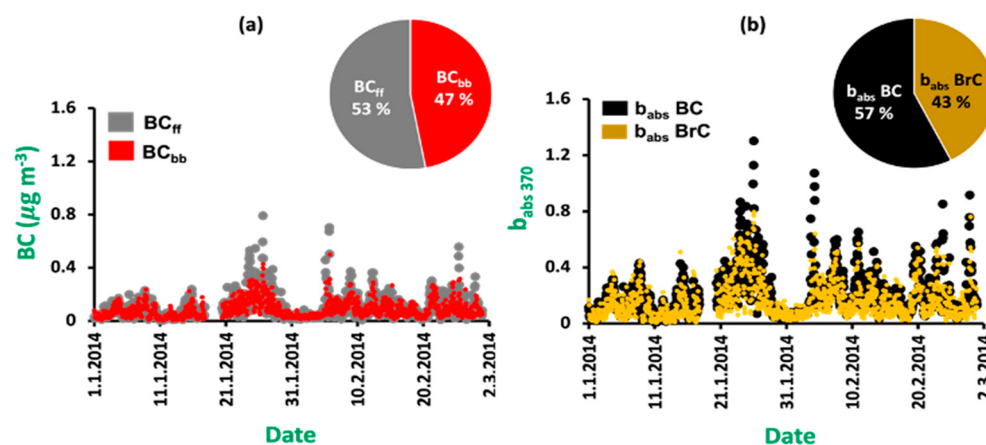


Figure 4. Time series and contributions of the hourly average BC_{ff} and BC_{bb} to the total BC (a), as well as $b_{abs} \text{ BrC}$ and $b_{abs} \text{ BC}$ to the total b_{abs} (Mm^{-1}) (b).

BC and brown carbon (BrC) have different spectral dependences, with brown carbon being expected to strongly absorb in UV range. Figure 4b depicts the time series of the light absorption coefficients of BC and BrC. The light absorption coefficient of BrC at 370 nm ($b_{abs, \text{BrC}}$) varied spatially from 0.02 to 0.80 Mm^{-1} . The $b_{abs, \text{BC}}$ showed slightly higher contribution 57% to light absorption at a lower wavelength (370 nm). According to the studies by Li et al. (2016) [61] and Qin et al. (2018) [49], it is obvious that during the winter season, the major source of BrC was biomass burning.

3.3. Source Apportionment of Ambient Organic Aerosol

The PMF analysis of the ACSM OA mass spectra identified five components, i.e., hydrocarbon-like organic aerosol (HOA) from traffic ($\text{HOA}_{\text{traffic}}$) and heating ($\text{HOA}_{\text{heating}}$) sources, secondary organic aerosol (SOA), local organic aerosol (LOA), and biomass burning organic aerosol. The biomass burning organic aerosols (BBOA) according to Minderytė et al. (2022), can be further divided into BBOA_{WB} from wood burning (levoglucosan) and BBOA_{DH} from domestic heating [55]. Each of those components showed a distinct time series, mass spectral profile, and diurnal variation corresponding to different sources (Figure 5). The BBOA_{WB} profile was characterized by higher contributions at m/z 's 60 ($\text{C}_2\text{H}_4\text{O}_2^+$) and 73 ($\text{C}_3\text{H}_5\text{O}_2^+$), which were associated with the fragmentation of sugars such as levoglucosan [62–64]. The BBOA_{DH} showed similar peaks to the BBOA_{WB} , albeit with additional prominent contributions of m/z 29 and 41. Additionally, the BBOA_{DH} was primarily detected following the beginning of the central household heating season, which supported the notion that the sources were associated with domestic heating [55]. The SOA component had the characteristics of oxygenated organic aerosol with a pronounced relative intensity of m/z 44 (indicating more oxidized aerosol). The intensity of the other fragments in its spectrum was low (<10). The LOA determined in this study showed a spectral pattern, which was characterized by high peaks of m/z 18, m/z 27, and m/z 86. Additionally, it had a highly clear source area because all the larger concentrations were connected to the west wind direction, where many industrial businesses were situated [18].

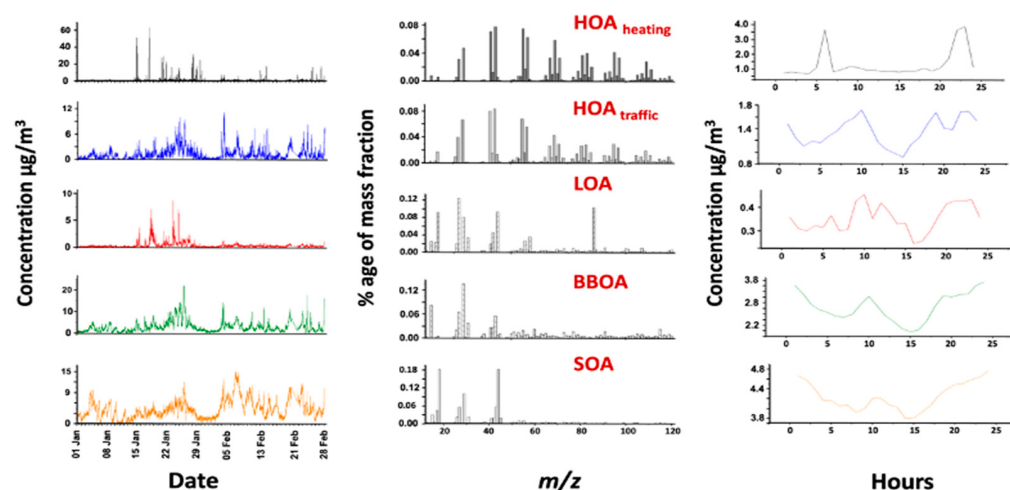


Figure 5. Mass spectra of the five factors from the PMF calculation. The time series (left), mass spectra (middle), and diurnal patterns (right) of the five OA factors determined on the basis of the PMF analysis.

From the initial unconstrained PMF five-factor solution, we could identify mixing between two different HOA sources: one from traffic and one from the additional heating mostly contributing during the coldest period. These two contributions were identified by looking at the diurnal cycle of the HOA, which showed two higher concentration peaks at unusual times for traffic (at c.a. 4:00 am and 10:00 pm) and lower concentration peaks during traffic rush hours (c.a. 7:00 am and 7:00 pm). Increasing the number of factors (>4) did not resolve the additional HOA factors, while the mixing persisted. Therefore, in order to separate the two contributions, we performed an unconstrained PMF analysis using as input the time points included in the hours. In addition to that, $\text{HOA}_{\text{heating}}$ showed two higher concentration peaks for Ep. 2 between (4:30–6:30 am and 7:30–11:30 pm). In total, 100 bootstrap PMF runs were conducted on this subset of the original data [50,65].

Figure 6 displays the computed diurnal pattern of the five OA components ($\text{HOA}_{\text{heating}}$, $\text{HOA}_{\text{traffic}}$, BBOA, LOA, and SOA) for the entire period by dividing it into three episodes. Here, the “Day” and “Night” values were calculated by averaging the factor concentration ($\mu\text{g m}^{-3}$) between 6 am and 7 pm (6:00–18:59 local time) for the “Day” values and from 7 pm to 6 am for the “Night” values (19:00–05:59 local time). The percentage expression of the difference (*Diff*) between the night-time and daytime factors concentration (C_{Night} and C_{Day} , respectively) was calculated using Equation (8) and is presented in Table 1.

$$\text{Diff} = (C_{\text{Night}} - C_{\text{Day}}) / C_{\text{Night}} \times 100\% \quad (8)$$

Table 1. Average of the factor concentration ($\mu\text{g m}^{-3}$) of the three episodes, during daytime (6 am–7 pm) and night-time (7 pm to 6 am).

	$\text{HOA}_{\text{heating}}$			$\text{HOA}_{\text{traffic}}$			LOA			BBOA			SOA		
	Day	Night	Diff	Day	Night	Diff	Day	Night	Diff	Day	Night	Diff	Day	Night	Diff
Ep. 1.	0.55	0.57	3%	0.79	0.69	−15%	0.13	0.14	6%	1.50	1.56	3%	2.73	3.28	17%
Ep. 2.	1.46	3.11	53%	1.60	1.88	15%	0.75	0.66	−13%	3.66	4.72	22%	3.40	3.71	9%
Ep. 3.	0.41	0.38	−9%	0.59	0.53	−12%	0.10	0.10	7%	1.06	1.27	16%	2.60	2.79	7%
Whole period	1.12	1.54	28%	1.33	1.39	5%	0.35	0.37	6%	2.58	3.25	21%	4.06	4.50	10%

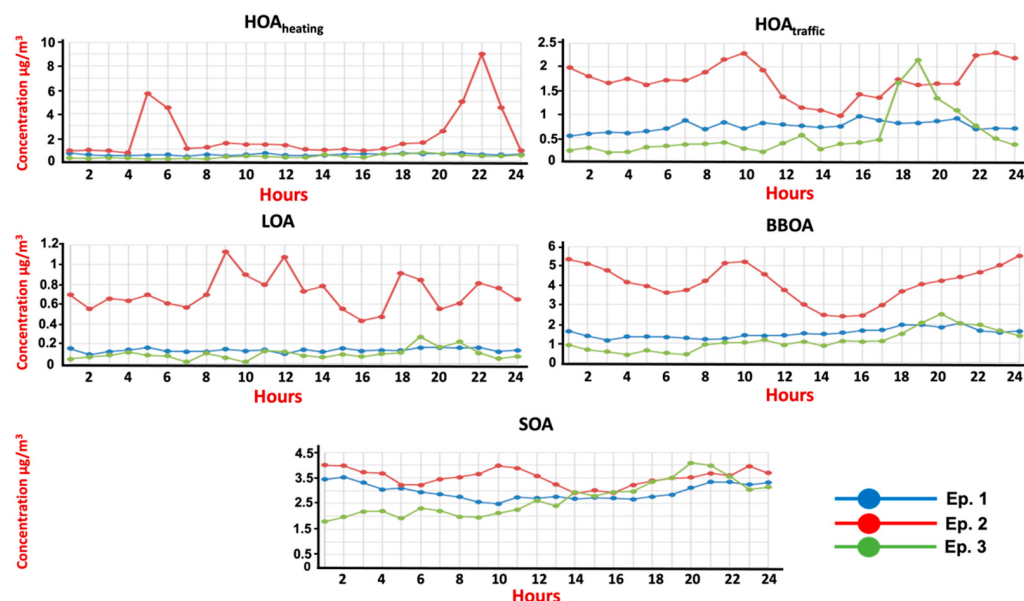


Figure 6. Diurnal patterns of the five OA factors ($\text{HOA}_{\text{heating}}$, $\text{HOA}_{\text{traffic}}$, LOA, BBOA, and SOA) determined based on the PMF analysis.

The $\text{HOA}_{\text{traffic}}$ diurnal pattern strongly suggested that it was associated with rush hours, while the $\text{HOA}_{\text{heating}}$ had a more pronounced diurnal profile. The highest $\text{HOA}_{\text{heating}}$ concentration occurred at 4:30–6:30 am and 7:30–11:30 pm. As expected, BBOA accounted for a large fraction of OA at the site, which was presumably associated with intensive domestic heating, and its contribution to the total OA mass varied on average between 26 and 77% (corresponding to 0.1–22.0 $\mu\text{g m}^{-3}$). The SOA factor represented a large fraction of the OA. On average, its relative contributions to the total OA spanned from 2 to 80%. Finally, LOA contributed on average 2%, from 1% to 40% (corresponding to 0.3–8.0 $\mu\text{g m}^{-3}$), to the total OA mass. The LOA factor did not show any regular diurnal pattern, although the contribution variation suggested that LOA mostly influenced (up to 40% [corresponding to 8.7 $\mu\text{g m}^{-3}$]) the OA concentration during extremely cold periods. This behavior allowed the identification of SOA, BBOA, and LOA emissions as the sources that were responsible for reaching high levels during the winter season in Vilnius. Extremely low temperatures were the primary meteorological factor that caused these high pollution episodes.

It is important to note that during Ep. 2., when the site was affected by extremely low temperatures, the $\text{HOA}_{\text{heating}}$ could be a product of the combustion of residual oil. The observed $\text{HOA}_{\text{heating}}$ diurnal cycle (Figure 6) was comparable to the reserved intensity of residual fuel combustion in the Vilnius Combined Heat and Power Plant (CHPP) during working days (during Monday–Friday only). The main fuels used in the Vilnius CHPP were natural gas and secondary (reserve) fuel—residual fuel oil. The Vilnius CHPP capacity was 603 MW heating power and 360 MW electric power. It was the most polluting electricity generating power plant in Lithuania and, therefore, it was closed at the beginning of 2016. The $\text{HOA}_{\text{heating}}$ mass concentration was strongly higher when the prevailing wind was from the N–NW (same as the direction of the plant). Therefore, it appears that a large portion of the HOA was transported to the measurement site over a period of 30 min. Thus, the diurnal cycle of the $\text{HOA}_{\text{heating}}$ was more likely driven by the emissions from the CHPP, which is consistent with a number of previous studies' findings that HOA is mainly associated with transport-related emissions [60,61].

The plot shows that the primary organic components, $\text{HOA}_{\text{heating}}$ and BBOA, had higher night-time concentration values during Ep. 2. If the daytime average was calculated from 6 am to 7 pm and the night average was calculated from 8 pm to 5 am, then the night average was 53% higher for $\text{HOA}_{\text{heating}}$ and 29% higher for BBOA. On the other hand, SOA showed a smaller difference in concentration during all the episodes and exhibited an increase in daytime during Ep. 2, although its contribution was higher over the whole

measurement period, particularly during the cleaner periods. Consequently, secondary pollutants play a major role in this part of the city. The hydrocarbon-like OA factor from heating contributed on average 11% to the total OA mass, with concentration ranges between 0.1 and 63.0 $\mu\text{g m}^{-3}$ depending on the measurement period, while HOA from traffic accounted for on average 13% ($\sim 1.4 \mu\text{g m}^{-3}$) during the whole measurement period.

3.4. Impact of Air Masses on the Concentration Level of OA and BC

For the cluster analysis of OA and BC, the 72 h backward trajectories were grouped into five clusters (Figures 7 and 8).

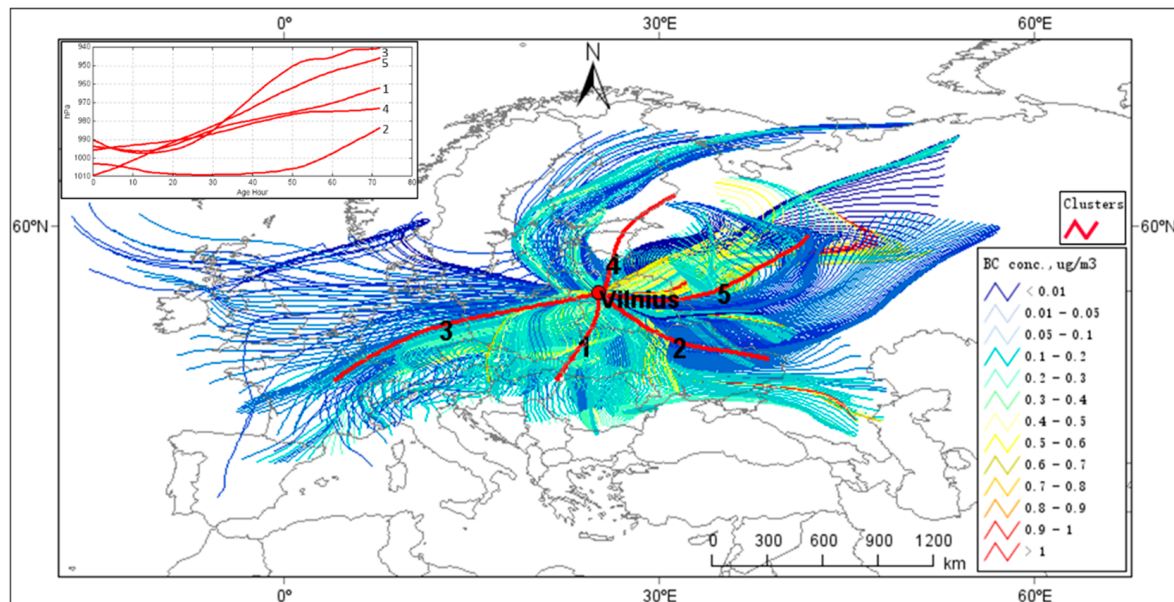


Figure 7. Air mass backward trajectories and clusters with associated BC mass concentration in January and February 2014.

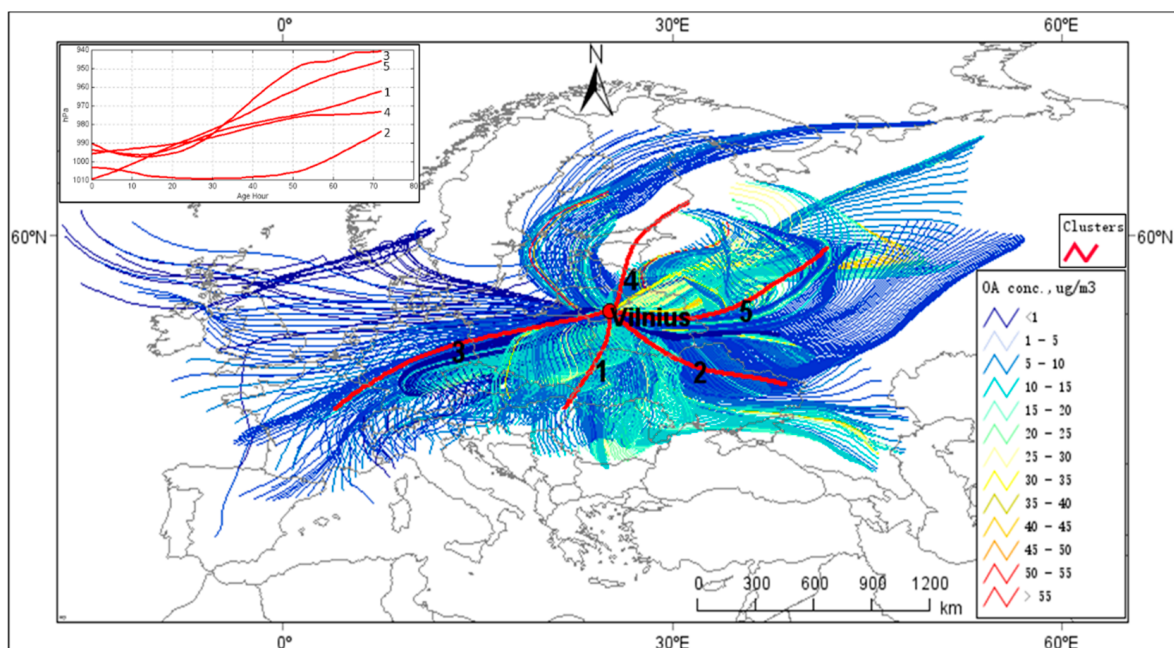


Figure 8. Air mass backward trajectories and clusters with associated OA concentration in January and February 2014.

In the trajectory analysis, the air masses assigned to clusters 1 and 3 came from the southwest and were observed in 33.83% and 13.56% of the cases, respectively; the air masses assigned to clusters 2 and 5 came from the east–southeast and accounted for 18.79% and 24.51% of the analyzed trajectories, respectively; while the air masses in cluster 4 arrived from the north and were composed of 9.32% of all the trajectories (Table 2). The clusters were also characterized by fluctuating atmospheric pressure (hPa) (subgraphs in Figures 7 and 8).

Table 2. Statistics of the clusters.

General Statistics of Clusters			Trajectories of BC			Trajectories of OA		
Cluster no.	No. of trajectories	Ratio to all trajectories	No. of trajectories	Mean conc., $\mu\text{g}/\text{m}^3$	Standard deviation	No. of trajectories	Mean conc., $\mu\text{g}/\text{m}^3$	Standard deviation
1	479	33.83%	479	0.21	0.12	473	11.15	5.85
2	266	18.79%	264	0.19	0.19	254	9.60	6.26
3	192	13.56%	192	0.11	0.09	164	4.90	3.59
4	132	9.32%	120	0.12	0.09	132	8.04	8.49
5	347	24.51%	297	0.25	0.21	346	12.27	8.03
Total	1416	100.01%	1352	0.19	0.16	1369	10.10	7.00

In the beginning, the air masses in cluster 1 reached ~962 hPa, while later at the receptor site the pressure increased to 994 hPa. The pressure in the air masses in cluster 2 fluctuated the least—from ~983 hPa in the beginning to ~1003 hPa at the receptor site. The pressure of cluster 3 fluctuated from almost 940 hPa to 990 hPa, while cluster 4 reached a pressure level of 973 hPa in the beginning and rose up to ~995 hPa at the end. Meanwhile, cluster 5 had the biggest amplitude of all the clusters, as it was characterized by a lower pressure of ~945 hPa in the beginning and reached almost 1009 hPa passing over the site area.

According to the statistical analysis of the clusters (Table 2), the highest BC concentration on average was calculated in clusters 5 and 1, with $0.25 \mu\text{g m}^{-3}$ and $0.21 \mu\text{g m}^{-3}$, respectively (Table 2). The lowest BC concentration in cluster 3 was $0.11 \mu\text{g m}^{-3}$, which was 1.1–2.27 times smaller than that of the other clusters.

Meanwhile, the air masses in clusters 5 and 1 had the highest OA concentration on average— 12.27 and $11.15 \mu\text{g m}^{-3}$, respectively. The average OA concentration in cluster 3 was 1.6–2.5 times lower compared with the average OA concentrations in the other clusters. In the WPSCF of the BC plot (Figure 9), the colors reflect the conditional probability that a cell is a source of BC, while in the WCWT plot (Figure 10), the colors express the weighted average concentration of BC in the cells [66]. As shown in Figure 9, the highest WPSCF values of the BC concentration were associated with air masses from northeastern Poland, northern–central Slovakia, northwestern–western–southern and southeastern Belarus, northwestern Ukraine, and western Russia, with the potential contribution to the BC being up to 1.1.

From these areas, the atmospheric BC arrived with air masses to the receptor site along clusters 1, 2, and 5 and affected the local BC concentration. The distribution of the WCWT values represents the contribution of potential sources to the BC mass concentration in Vilnius (Figure 10).

The highest concentrations of BC arrived with air masses from western Russia (which contributed up to $0.4 \mu\text{g m}^{-3}$ to the BC), northern–western–southwestern–eastern–northeastern Belarus (up to $0.35 \mu\text{g m}^{-3}$), northwestern–western–central–southwestern–southern Ukraine (up to $0.29 \mu\text{g m}^{-3}$), northwestern–southern Romania (up to $0.24 \mu\text{g m}^{-3}$), southwestern and northern Slovakia (up to $0.25 \mu\text{g m}^{-3}$), northeastern and southern Czech Republic (up to $0.27 \mu\text{g m}^{-3}$), and northeastern–eastern–southeastern–southern–southwestern Poland (up to $0.25 \mu\text{g m}^{-3}$). According to the WCWT analysis results, these territories made the biggest impact on the increased BC concentration in Vilnius. However, the lowest concentrations were associated with air masses passing from further southwestern, northern,

southeastern, and further eastern Lithuania; therefore, the WCWT values varied within the range of $0.03\text{--}0.06\ \mu\text{g m}^{-3}$. These results indicate that these regions made the smallest contribution to the BC concentration in Vilnius. Meanwhile, the highest WPSCF values of OA (Figure 11) were calculated for central Slovakia, eastern–northeastern Poland, western–southern and eastern Belarus, northwestern Ukraine, and western Russia. The range of the WPSCF values was also 1.0–1.1.

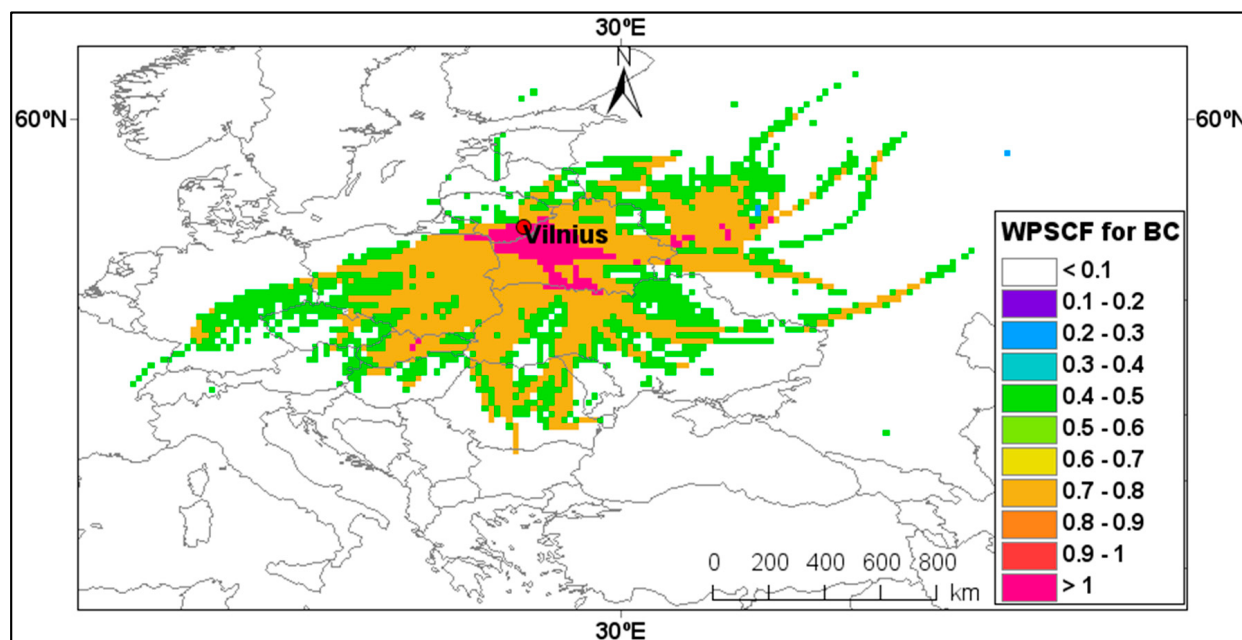


Figure 9. Weighted potential source contribution function of BC in Vilnius in January and February 2014.

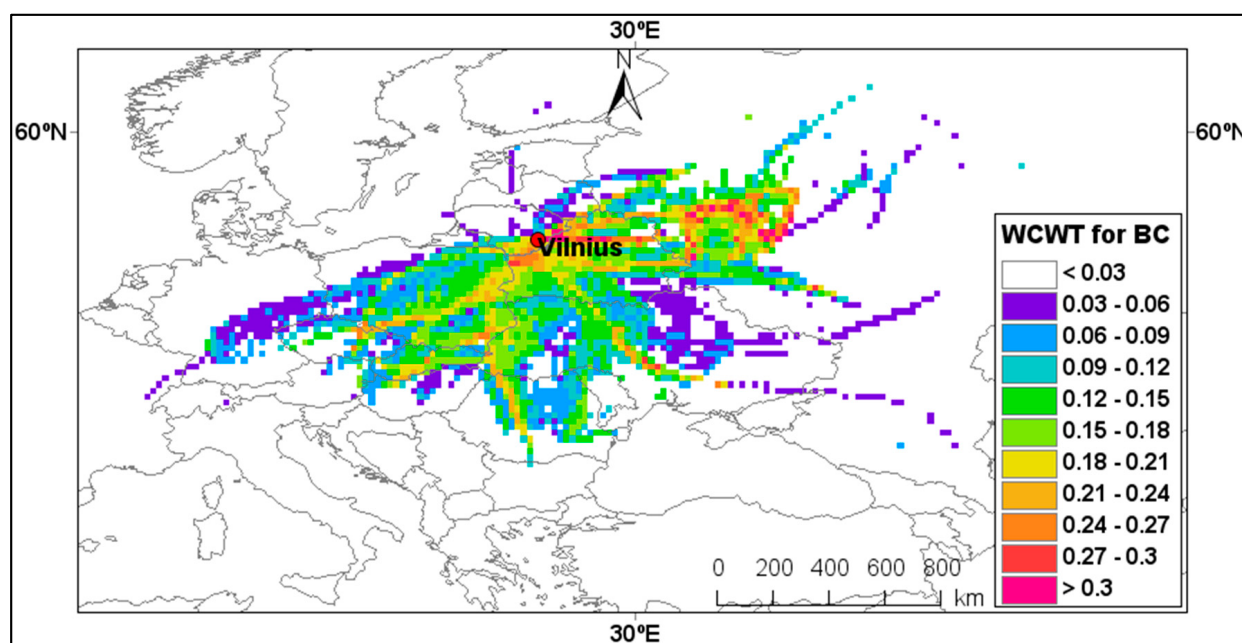


Figure 10. Weighted concentration weighted trajectory of BC in Vilnius in January and February 2014.

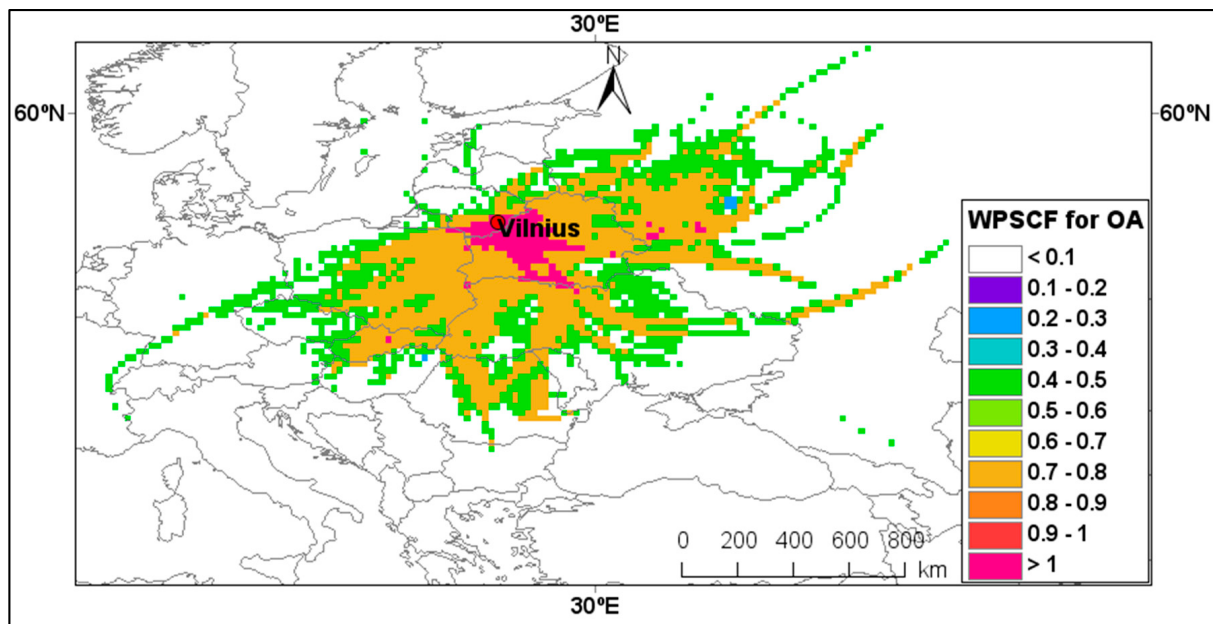


Figure 11. Weighted potential source contribution function (WPSCF) of OA in Vilnius in January and February 2014.

Atmospheric BC from these areas arrived at the receptor site with air masses in clusters 1, 2, and 5, and it had a noticeable impact on the BC concentration level. The contribution of potential sources to the OA concentrations in Vilnius is represented in Figure 12. The highest concentrations of OA associated with air masses originated from Central Europe (western–central–northeastern Czech Republic, Slovakia, Poland [except western parts], northeastern Hungary), Southeastern Europe (northeastern–central–southern and eastern Romania), Eastern Europe (northwestern–western–southwestern–southern Ukraine, northwest Moldova, Belarus [except the southeastern part]), southeastern Latvia, and western Russia. The range of the highest WCWT values ranged from 9.7 to 16.37 $\mu\text{g m}^{-3}$. As shown in Figure 12, these territories, which had the greatest impact on the concentration of OA in Vilnius, are geographically widely distributed.

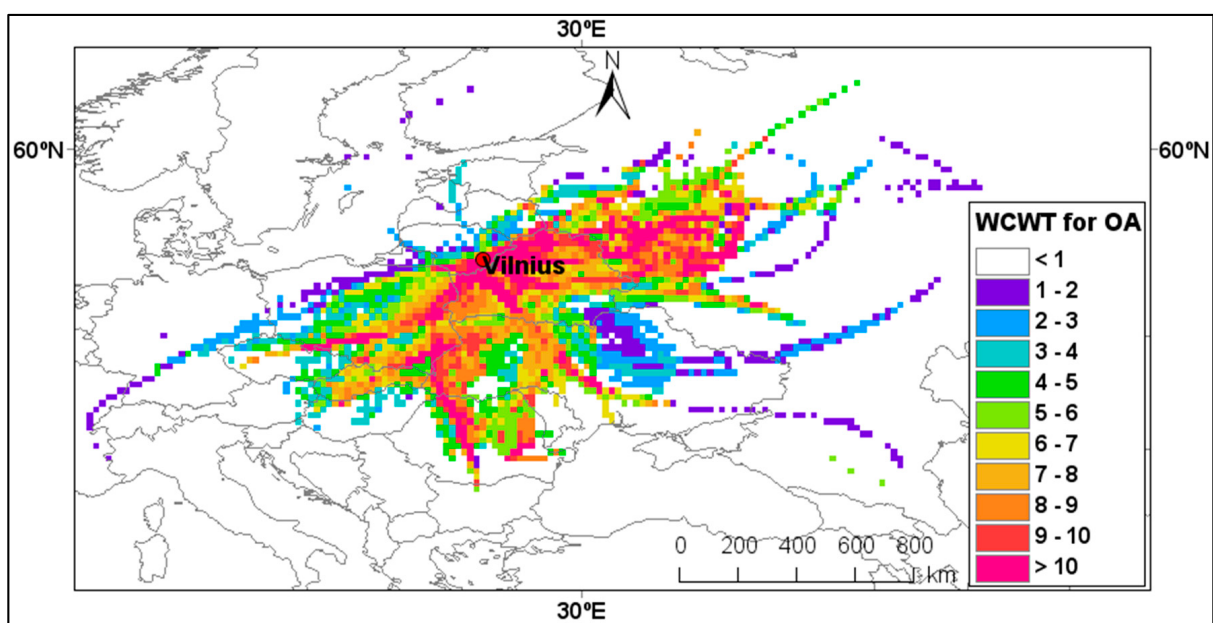


Figure 12. Weighted concentration weighted trajectory (WCWT) of OA in Vilnius in January and February 2014.

Meanwhile, the regions that had the weakest influence on the OA concentration at the receptor site were further west–southwest, southeast, and north of Lithuania and much further east–southeast in the territory of Russia. The trajectories of the air masses arriving over Vilnius territories were evaluated for the third period (Ep. 3) due to the extremely low temperature event (Figure 13). A 96 h backward trajectory was computed every 3 h for altitudes fixed at 500 m, 1000 m, and 1500 m above the observation site. The analysis disclosed that the receptor site was affected by widely distributed air masses that originated from different regions in Eurasia. In Figure 13, the red trajectory distinguishes the height of which, on 27 January, reached more than 3000 m agl over the territory of northern Siberia. Its height gradually dropped to 1500 m agl on 29 January. Later, the height of the trajectory descended less quickly to 500 m agl at the receptor site. There is noticeable compliance between the movement of a high-pressure system towards the Baltic region and the red trajectory that came from northern Siberia to Vilnius on the same days in Ep. 3. Although the air temperature dropped below $-10\text{ }^{\circ}\text{C}$ on almost all the days of Ep. 3 and no precipitation was recorded (these meteorological conditions are favorable for an increase in air pollution), the hourly average BC and OA concentrations in Vilnius gradually decreased during this period. Such an effect could have been caused by the increased wind during these days.

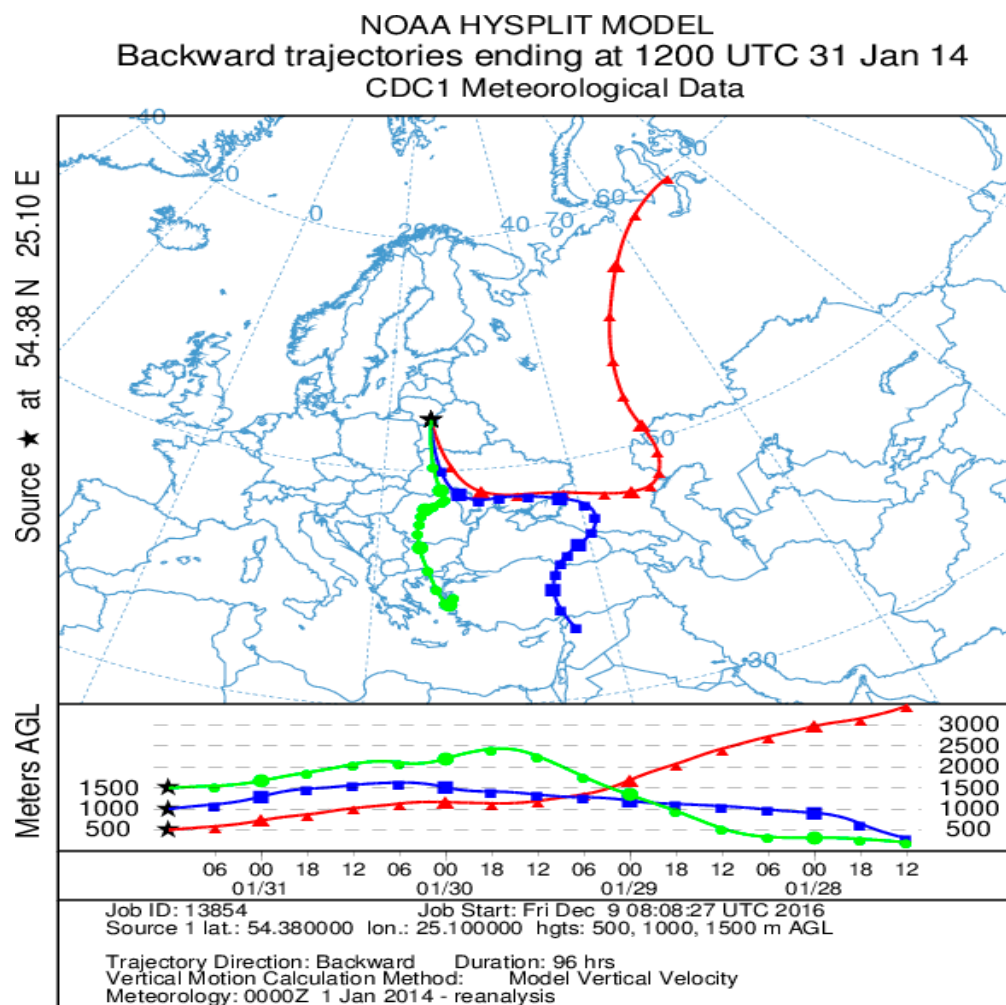


Figure 13. The 96 h backward trajectories of air masses arriving at Vilnius when the pollutant mass concentrations were the lowest. Trajectories were evaluated at each 3 h intervals.

In addition, on 1 February, no precipitation was recorded. The temperature dropped to minus $-14\text{ }^{\circ}\text{C}$ at night and the wind speed reached $8\text{--}13\text{ ms}^{-1}$, with wind gusts up to $15\text{--}18\text{ ms}^{-1}$. The increased windiness, coupled with the air masses that arrived to the

receptor site from different altitudes (ranging from below 500 m agl in the source regions to 1000–1500 m agl above the receptor site) and directions, were expected to improve the air quality in Vilnius by dispersing the air pollution.

4. Conclusions

This study investigated the chemical components of NR-PM₁ and the BC mass concentration and sources in an urban environment in Vilnius during January–February 2014. The organic fraction was apportioned into five organic aerosol factors representing HOA, which were likely to be related to traffic and residual fuel oil combustion for central household heating, biomass burning that mostly originated from residential wood burning and domestic heating, local OA factor and secondary organics. The diurnal five-factor behavior allowed us to identify HOA_{heating} and BBOA as the sources that were responsible for the high night-time PM₁ concentrations reached during the extremely low ambient temperature episode in Vilnius. Secondary organic aerosols played a major role during the whole measurement campaign in this part of the city, contributing on average from 2 to 80% to the total OA mass in Vilnius. Combinations of the AAE values AAE_{ff} = 0.9 and AAE_{bb} = 1.68 were used for the BC source apportionment in an urban background environment. After completing the BC source apportionment using the predetermined AAE values (0.9; 1.68), it was discovered that BC_{ff} contributed more (53%) than BC_{bb} (47%) in 2014. The fossil fuel-related BC contribution was further supported by an analysis of brown carbon (BrC). The b_{abs} BC presented a higher contribution of 57% and b_{abs} BrC presented a lower contribution of 43%, which showed that the transportation sector is a significant contributor to rising BC emissions in Vilnius. The reduction in pollution was significantly and directly impacted by the low relative humidity of the air. The clustering analysis revealed that the highest BC and OA concentrations on average were observed in air masses arriving from southwest–south–southeast–east and northeast. However, the potential sources of OA were widely distributed—the highest WCWT values were seen in some countries in Central, Southern, Southeastern, and Eastern Europe and western Russia.

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References

1. Oh, H.J.; Ma, Y.; Kim, J. Human inhalation exposure to aerosol and health effect: Aerosol monitoring and modelling regional deposited doses. *Int. J. Environ. Res. Public Health* **2020**, *17*, 1923. [[CrossRef](#)] [[PubMed](#)]
2. Ouidir, M.; Seyve, E.; Rivi re, E.; Bernard, J.; Cheminat, M.; Cortinovis, J.; Ducroz, F.; Dugay, F.; Hulin, A.; Kloog, I.; et al. Maternal ambient exposure to atmospheric pollutants during pregnancy and offspring term birth weight in the nationwide elfe cohort. *Int. J. Environ. Res. Public Health* **2021**, *18*, 5806. [[CrossRef](#)] [[PubMed](#)]
3. WHO. WHO global air quality guidelines. *Coast. Estuar. Process.* **2021**, 1–360.
4. Lelieveld, J.; Evans, J.S.; Fnais, M.; Giannadaki, D.; Pozzer, A. The contribution of outdoor air pollution sources to premature mortality on a global scale. *Nature* **2015**, *525*, 367–371. [[CrossRef](#)] [[PubMed](#)]

5. Cao, J.J.; Lee, S.C.; Ho, K.F.; Zhang, X.Y.; Zou, S.C.; Fung, K.; Chow, J.C.; Watson, J.G. Characteristics of carbonaceous aerosol in Pearl River Delta Region, China during 2001 winter period. *Atmos. Environ.* **2003**, *37*, 1451–1460. [\[CrossRef\]](#)
6. Zou, C.; Wang, J.; Hu, K.; Li, J.; Yu, C.; Zhu, F.; Huang, H. Distribution characteristics and source apportionment of winter carbonaceous aerosols in a rural area in Shandong, China. *Atmosphere* **2022**, *13*, 1858. [\[CrossRef\]](#)
7. Chung, C.E.; Ramanathan, V.; Decremer, D. Observationally constrained estimates of carbonaceous aerosol radiative forcing. *Proc. Natl. Acad. Sci. USA* **2012**, *109*, 11624–11629. [\[CrossRef\]](#)
8. Ancelet, T.; Davy, P.K.; Trompeter, W.J.; Markwitz, A.; Weatherburn, D.C. Carbonaceous aerosols in a wood burning community in rural New Zealand. *Atmos. Pollut. Res.* **2013**, *4*, 245–249. [\[CrossRef\]](#)
9. Pachauri, T.; Singla, V.; Satsangi, A.; Lakhani, A.; Kumari, K.M. Characterization of carbonaceous aerosols with special reference to episodic events at Agra, India. *Atmos. Res.* **2013**, *128*, 98–110. [\[CrossRef\]](#)
10. Safai, P.D.; Raju, M.P.; Rao, P.S.P.; Pandithurai, G. Characterization of carbonaceous aerosols over the urban tropical location and a new approach to evaluate their climatic importance. *Atmos. Environ.* **2014**, *92*, 493–500. [\[CrossRef\]](#)
11. Bisht, D.S.; Dumka, U.C.; Kaskaoutis, D.G.; Pipal, A.S.; Srivastava, A.K.; Soni, V.K.; Attri, S.D.; Sateesh, M.; Tiwari, S. Carbonaceous aerosols and pollutants over delhi urban environment: Temporal evolution, source apportionment and radiative forcing. *Sci. Total Environ.* **2015**, *521*, 431–445. [\[CrossRef\]](#)
12. Pani, S.K.; Wang, S.H.; Lin, N.H.; Chantara, S.; Lee, C.T.; Thepnuan, D. Black carbon over an urban atmosphere in Northern Peninsular Southeast Asia: Characteristics, source apportionment, and associated health risks. *Environ. Pollut.* **2020**, *259*, 113871. [\[CrossRef\]](#)
13. Lin, W.; Dai, J.; Liu, R.; Zhai, Y.; Yue, D.; Hu, Q. Integrated assessment of health risk and climate effects of black carbon in the Pearl River Delta Region, China. *Environ. Res.* **2019**, *176*, 108522. [\[CrossRef\]](#) [\[PubMed\]](#)
14. Mauderly, J.L.; Chow, J.C. Health effects of organic aerosols. *Inhal. Toxicol.* **2008**, *20*, 257–288. [\[CrossRef\]](#) [\[PubMed\]](#)
15. Daniele, C.; Vechhi, R.; Viana, M. Carbonaceous aerosols in the atmosphere. *Atmosphere* **2018**, *9*, 181. [\[CrossRef\]](#)
16. Diapouli, E.; Kalogridis, A.C.; Markantonaki, C.; Vratolis, S.; Fetfatzis, P.; Colombi, C.; Eleftheriadis, K. Annual variability of black carbon concentrations originating from biomass and fossil fuel combustion for the suburban aerosol in Athens, Greece. *Atmosphere* **2017**, *8*, 234. [\[CrossRef\]](#)
17. Klejnowski, K.; Janoszka, K.; Czaplicka, M. Characterization and seasonal variations of organic and elemental carbon and levoglucosan in PM10 in Krynica Zdroj, Poland. *Atmosphere* **2017**, *8*, 190. [\[CrossRef\]](#)
18. Minderytė, A.; Pauraitė, J.; Dudoitis, V.; Plauškaitė, K.; Kilikevičius, A.; Matijošius, J.; Rimkus, A.; Kilikevičienė, K.; Vainorius, D.; Byčenkienė, S. Carbonaceous aerosol source apportionment and assessment of transport-related pollution. *Atmos. Environ.* **2022**, *279*, 119043. [\[CrossRef\]](#)
19. Goss, M.; Swain, D.L.; Abatzoglou, J.T.; Sarhadi, A.; Kolden, C.A.; Williams, A.P.; Diffenbaugh, N.S. Climate change is increasing the likelihood of extreme autumn wildfire conditions across California. *Environ. Res. Lett.* **2020**, *15*, 094016. [\[CrossRef\]](#)
20. Kirchmeier-Young, M.C.; Gillett, N.P.; Zwiers, F.W.; Cannon, A.J.; Anslow, F.S. Attribution of the influence of human-induced climate change on an extreme fire season. *Earth's Future* **2019**, *7*, 2–10. [\[CrossRef\]](#) [\[PubMed\]](#)
21. Hamilton, R.S.; Mansfield, T.A. Airborne particulate elemental carbon: Its sources, transport and contribution to dark smoke and soiling. *Atmos. Environ. Part A Gen. Top.* **1991**, *25*, 715–723. [\[CrossRef\]](#)
22. Briggs, N.L.; Long, C.M. Critical review of black carbon and elemental carbon source apportionment in Europe and the United States. *Atmos. Environ.* **2016**, *144*, 409–427. [\[CrossRef\]](#)
23. Popovicheva, O.; Ivanov, A.; Vojtisek, M. Functional factors of biomass burning contribution to spring aerosol composition in a Megacity: Combined ftir-pca analyses. *Atmosphere* **2020**, *11*, 319. [\[CrossRef\]](#)
24. Chernyshev, V.V.; Zakharenko, A.M.; Ugay, S.M.; Hien, T.T.; Hai, L.H.; Kholodov, A.S.; Burykina, T.I.; Stratidakis, A.K.; Mezhev, Y.O.; Tsatsakis, A.M.; et al. Morphologic and chemical composition of particulate matter in motorcycle engine exhaust. *Toxicol. Rep.* **2018**, *5*, 224–230. [\[CrossRef\]](#)
25. Chernyshev, V.V.; Zakharenko, A.M.; Ugay, S.M.; Hien, T.T.; Hai, L.H.; Olesik, S.M.; Kholodov, A.S.; Zubko, E.; Kokkinakis, M.; Burykina, T.I.; et al. Morphological and chemical composition of particulate matter in buses exhaust. *Toxicol. Rep.* **2019**, *6*, 120–125. [\[CrossRef\]](#)
26. Reddington, C.L.; McMeeking, G.; Mann, G.W.; Coe, H.; Frontoso, M.G.; Liu, D.; Flynn, M.; Spracklen, D.V.; Carslaw, K.S. The mass and number size distributions of black carbon aerosol over Europe. *Atmos. Chem. Phys.* **2013**, *13*, 4917–4939. [\[CrossRef\]](#)
27. Ceolato, R.; Bedoya-Velásquez, A.E.; Fossard, F.; Mouysset, V.; Paulien, L.; Lefebvre, S.; Mazzoleni, C.; Sorensen, C.; Berg, M.J.; Yon, J. Black carbon aerosol Number and mass concentration measurements by picosecond short-range elastic backscatter lidar. *Sci. Rep.* **2022**, *12*, 8443. [\[CrossRef\]](#) [\[PubMed\]](#)
28. Weagle, C.L.; Snider, G.; Li, C.; Van Donkelaar, A.; Philip, S.; Bissonnette, P.; Burke, J.; Jackson, J.; Latimer, R.; Stone, E.; et al. Global sources of fine particulate matter: Interpretation of PM2.5 chemical composition observed by SPARTAN using a global chemical transport model. *Environ. Sci. Technol.* **2018**, *52*, 11670–11681. [\[CrossRef\]](#) [\[PubMed\]](#)
29. Kwon, H.S.; Ryu, M.H.; Carlsten, C. Ultrafine particles: Unique physicochemical properties relevant to health and disease. *Exp. Mol. Med.* **2020**, *52*, 318–328. [\[CrossRef\]](#)
30. Wang, C.; Ye, Z.; Yu, Y.; Gong, W. Estimation of bus emission models for different fuel types of buses under real conditions. *Sci. Total Environ.* **2018**, *640*, 965–972. [\[CrossRef\]](#)

31. Shan, X.; Chen, X.; Jia, W.; Ye, J. Evaluating urban bus emission characteristics based on localized MOVES using Sparse GPS data in Shanghai, China. *Sustainability* **2019**, *11*, 2936. [\[CrossRef\]](#)
32. Byčenkienė, S.; Khan, A.; Bimbaitė, V. Impact of PM_{2.5} and PM₁₀ emissions on changes of their concentration levels in Lithuania: A case study. *Atmosphere* **2022**, *13*, 1793. [\[CrossRef\]](#)
33. Jonidi Jafari, A.; Charkhloo, E.; Pasalari, H. *Urban Air Pollution Control Policies and Strategies: A Systematic Review*; Springer International Publishing: Berlin/Heidelberg, Germany, 2021; Volume 19, ISBN 0123456789.
34. Triantafyllopoulos, G.; Dimaratos, A.; Ntziachristos, L.; Bernard, Y.; Dornoff, J.; Samaras, Z. A Study on the CO₂ and NO_x emissions performance of Euro 6 diesel vehicles under various chassis dynamometer and on-road conditions including latest regulatory provisions. *Sci. Total Environ.* **2019**, *666*, 337–346. [\[CrossRef\]](#) [\[PubMed\]](#)
35. Brewer, T.L. Black carbon emissions and regulatory policies in transportation. *Energy Policy* **2019**, *129*, 1047–1055. [\[CrossRef\]](#)
36. Grigoratos, T.; Fontaras, G.; Giechaskiel, B.; Zacharof, N. Real world emissions performance of heavy-duty euro vi diesel vehicles. *Atmos. Environ.* **2019**, *201*, 348–359. [\[CrossRef\]](#)
37. Lurkin, V.; Hambuckers, J.; van Woensel, T. Urban low emissions zones: A behavioral operations management perspective. *Transp. Res. Part A Policy Pract.* **2021**, *144*, 222–240. [\[CrossRef\]](#)
38. Trojanowski, R.; Fthenakis, V. Nanoparticle emissions from residential wood combustion: A critical literature review, characterization, and recommendations. *Renew. Sustain. Energy Rev.* **2019**, *103*, 515–528. [\[CrossRef\]](#)
39. Ng, N.L.; Herndon, S.C.; Trimborn, A.; Canagaratna, M.R.; Croteau, P.L.; Onasch, T.B.; Sueper, D.; Worsnop, D.R.; Zhang, Q.; Sun, Y.L.; et al. An aerosol chemical speciation monitor (ACSM) for routine monitoring of the composition and mass concentrations of ambient aerosol. *Aerosol Sci. Technol.* **2011**, *45*, 780–794. [\[CrossRef\]](#)
40. Middlebrook, A.M.; Bahreini, R.; Jimenez, J.L.; Canagaratna, M.R. Evaluation of composition-dependent collection efficiencies for the aerodyne aerosol mass spectrometer using field data. *Aerosol Sci. Technol.* **2012**, *46*, 258–271. [\[CrossRef\]](#)
41. Canonaco, F.; Crippa, M.; Slowik, J.G.; Baltensperger, U.; Prévôt, A.S.H. SoFi, an IGOR-based interface for the efficient use of the generalized multilinear engine (ME-2) for the source apportionment: ME-2 application to aerosol mass spectrometer data. *Atmos. Meas. Tech.* **2013**, *6*, 3649–3661. [\[CrossRef\]](#)
42. Helin, A.; Niemi, J.V.; Virkkula, A.; Pirjola, L.; Teinilä, K.; Backman, J.; Aurela, M.; Saarikoski, S.; Rönkkö, T.; Asmi, E.; et al. Characteristics and source apportionment of black carbon in the Helsinki Metropolitan area, Finland. *Atmos. Environ.* **2018**, *190*, 87–98. [\[CrossRef\]](#)
43. Liu, C.; Chung, C.E.; Yin, Y.; Schnaiter, M. The absorption Ångström exponent of black carbon: From numerical aspects. *Atmos. Chem. Phys.* **2018**, *18*, 6259–6273. [\[CrossRef\]](#)
44. Weingartner, E.; Saathoff, H.; Schnaiter, M.; Streit, N.; Bitnar, B.; Baltensperger, U. Absorption of light by soot particles: Determination of the absorption coefficient by means of aethalometers. *J. Aerosol Sci.* **2003**, *34*, 1445–1463. [\[CrossRef\]](#)
45. Zotter, P.; Herich, H.; Gysel, M.; El-Haddad, I.; Zhang, Y.; Mocnik, G.; Hüglin, C.; Baltensperger, U.; Szidat, S.; Prévôt, A.S.H. Evaluation of the absorption Ångström exponents for traffic and wood burning in the aethalometer-based source apportionment using radiocarbon measurements of ambient aerosol. *Atmos. Chem. Phys.* **2017**, *17*, 4229–4249. [\[CrossRef\]](#)
46. Paatero, P. Least squares formulation of robust non-negative factor analysis. *Chemom. Intell. Lab. Syst.* **1997**, *37*, 23–35. [\[CrossRef\]](#)
47. Crippa, M.; Canonaco, F.; Lanz, V.A.; Äijälä, M.; Allan, J.D.; Carbone, S.; Capes, G.; Ceburnis, D.; Dall'Osto, M.; Day, D.A.; et al. Organic aerosol components derived from 25 AMS data sets across Europe Using a consistent ME-2 based source apportionment approach. *Atmos. Chem. Phys.* **2014**, *14*, 6159–6176. [\[CrossRef\]](#)
48. Sandradewi, J.; Prévôt, A.S.H.; Weingartner, E.; Schmidhauser, R.; Gysel, M.; Baltensperger, U. A study of wood burning and traffic aerosols in an Alpine valley using a multi-wavelength aethalometer. *Atmos. Environ.* **2008**, *42*, 101–112. [\[CrossRef\]](#)
49. Qin, Y.M.; Bo Tan, H.; Li, Y.J.; Jie Li, Z.; Schurman, M.I.; Liu, L.; Wu, C.; Chan, C.K. chemical characteristics of brown carbon in atmospheric particles at a suburban site near Guangzhou, China. *Atmos. Chem. Phys.* **2018**, *18*, 16409–16418. [\[CrossRef\]](#)
50. Pauraite, J.; Mainelis, G.; Kecorius, S.; Minderytė, A.; Dudoitis, V.; Garbarienė, I.; Plauškaitė, K.; Ovadnevaite, J.; Byčenkienė, S. Office indoor PM and BC level in Lithuania: The role of a long-range smoke transport Event. *Atmosphere* **2021**, *12*, 1047. [\[CrossRef\]](#)
51. Sun, Y.L.; Zhang, Q.; Schwab, J.J.; Demerjian, K.L.; Chen, W.N.; Bae, M.S.; Hung, H.M.; Hogrefe, O.; Frank, B.; Rattigan, O.V.; et al. Characterization of the sources and processes of organic and inorganic aerosols in New York City with a high-resolution time-of-flight aerosol mass spectrometer. *Atmos. Chem. Phys.* **2011**, *11*, 1581–1602. [\[CrossRef\]](#)
52. Saarikoski, S.; Niemi, J.V.; Aurela, M.; Pirjola, L.; Kousa, A.; Rönkkö, T.; Timonen, H. Sources of black carbon at residential and traffic environments obtained by two source apportionment methods. *Atmos. Chem. Phys.* **2021**, *21*, 14851–14869. [\[CrossRef\]](#)
53. Paatero, P.; Tapper, U. Positive matrix factorization: A non-negative factor model with optimal utilization of error estimates of data values. *Environmetrics* **1994**, *5*, 111–126. [\[CrossRef\]](#)
54. Paglione, M.; Gilardoni, S.; Rinaldi, M.; Decesari, S.; Zanca, N.; Sandrini, S.; Giulianelli, L.; Bacco, D.; Ferrari, S.; Poluzzi, V.; et al. The impact of biomass burning and aqueous-phase processing on air quality: A multi-year source apportionment study in the Po Valley, Italy. *Atmos. Chem. Phys.* **2020**, *20*, 1233–1254. [\[CrossRef\]](#)
55. Wang, Y.Q.; Zhang, X.Y.; Draxler, R.R. TrajStat: GIS-Based software that uses various trajectory statistical analysis methods to identify potential sources from long-term air pollution measurement data. *Environ. Model. Softw.* **2009**, *24*, 938–939. [\[CrossRef\]](#)
56. Guo, Y.; Lin, C.; Li, J.; Wei, L.; Ma, Y.; Yang, Q.; Li, D.; Wang, H.; Shen, J. Persistent pollution episodes, transport pathways, and potential sources of air pollution during the heating season of 2016–2017 in Lanzhou, China. *Environ. Monit. Assess.* **2021**, *193*, 852. [\[CrossRef\]](#)

57. Hsu, Y.K.; Holsen, T.M.; Hopke, P.K. Comparison of hybrid receptor models to locate pcb sources in Chicago. *Atmos. Environ.* **2003**, *37*, 545–562. [[CrossRef](#)]
58. Draxler, R.R.; Hess, G.D. An overview of the hysplit_4 modelling system for trajectories, dispersion and deposition. *Aust. Meteorol. Mag.* **1998**, *47*, 295–308.
59. Teinilä, K.; Aurela, M.; Niemi, J.V.; Kousa, A.; Petäjä, T.; Järvi, L.; Hillamo, R.; Kangas, L.; Saarikoski, S.; Timonen, H. Concentration variation of gaseous and particulate pollutants in the helsinki city centre—Observations from a two-year campaign from 2013–2015. *Boreal Environ. Res.* **2019**, *24*, 115–136.
60. Calvo, A.I.; Alves, C.; Castro, A.; Pont, V.; Vicente, A.M.; Fraile, R. Research on aerosol sources and chemical composition: Past, current and emerging issues. *Atmos. Res.* **2013**, *121*, 1–28. [[CrossRef](#)]
61. Li, X.; Zhao, Q.; Yang, Y.; Zhao, Z.; Liu, Z.; Wen, T.; Hu, B.; Wang, Y.; Wang, L.; Wang, G. Composition and sources of brown carbon aerosols in Megacity Beijing during the winter of 2016. *Atmos. Res.* **2021**, *262*, 105773. [[CrossRef](#)]
62. Weitzel, K.; Chemie, F.; Rev, M.S.; Introduction, I.; Reference, C. Bond-dissociation energies of cations—Pushing the limits to quantum state resolution. *WHO Libr. Cat. Data* **2011**, 221–235. [[CrossRef](#)] [[PubMed](#)]
63. Thepnuan, D.; Chantara, S.; Lee, C.T.; Lin, N.H.; Tsai, Y.I. Molecular markers for biomass burning associated with the characterization of PM2.5 and component sources during dry season haze episodes in upper South East Asia. *Sci. Total Environ.* **2019**, *658*, 708–722. [[CrossRef](#)] [[PubMed](#)]
64. Weimer, S.; Alfarra, M.R.; Schreiber, D.; Mohr, M.; Prévôt, A.S.H.; Baltensperger, U. Organic aerosol mass spectral signatures from wood-burning emissions: Influence of burning conditions and type. *J. Geophys. Res. Atmos.* **2008**, *113*, D10304. [[CrossRef](#)]
65. May, N.; Kuo, L.; Caby, E.A. Bootstrap Methods and Their Application. *ASQ* **2014**, *42*, 216–217. [[CrossRef](#)]
66. Berriban, I.; Azahra, M.; Chham, E.; Ferro-García, M.A.; Milena-Pérez, A.; Nouayti, A.; Orza, J.A.G.; Brattich, E.; Tositti, L.; Piñero-García, F.; et al. PSCF and CWT Methods as a Tool to Identify Potential sources of 7Be and 210Pb aerosols in Granada, Spain. *J. Environ. Radioact.* **2022**, *251*, 106977. [[CrossRef](#)] [[PubMed](#)]

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