



Modeling Anthropogenic Aerosol Sources and Secondary Organic Aerosol Formation: A Wintertime Study in Central Europe

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Abstract.

Anthropogenic aerosol particles remain a significant air quality concern in Central Europe, particularly during winter months. This study employs the COSMO-MUSCAT chemistry transport model to investigate particulate matter sources, with a focus on emissions from residential heating. The model results are compared with winter measurements from sites in Germany and the Czech Republic, where solid fuels are commonly used for heating. A non-reactive tagging method tracking primary organic matter (OM) reveals a high contribution from residential heating. Although the magnitude and temporal changes of the model results mostly agree with total OM values at two measuring stations, it appears to underestimate measurements at a site in the central Czech Republic. This underestimation is partly attributed to the inadequate representation of secondary organic aerosol (SOA) emitted from wood combustion. The study highlights the impact of anthropogenic volatile organic compounds (AVOC) on SOA formation, which are currently underrepresented in air quality models. Sensitivity tests adjusting SOA yields and AVOC emissions increase OM concentrations of up to 40% at the measurement sites. These findings emphasize the need for accurate parameterization of AVOC derived SOA formation and residential heating emissions to better tackle wintertime air quality challenges in Central Europe.

1 Introduction

According to the European Environment Agency's air quality report, 238.000 premature deaths can be attributed to PM_{2.5} (particulate matter of 2.5 µm or smaller aerodynamic diameter) exposure in the European Union (EU) in 2020 (EEA, 2022). In a review summarizing multiple decades of research, Anderson et al. (2011) emphasize the significant impact of PM exposure on global public health and the wider societal costs associated with it. Long-term exposure to PM has been shown to be causally related to cardiovascular disease and mortality (Anderson et al., 2011). The target of the EU's Air Pollution Action Plan is a 55% reduction in premature mortality due to PM_{2.5} by 2030 compared to 2005 levels (EEA, 2022). However, based on self-reported data, 19 EU Member States still fall at least 30% short of their 2030 PM_{2.5} emission reduction targets in 2021. A study by Beloconi and Vounatsou (2023) found that as of 2021, 47.5% of Europeans were living in areas where annual mean PM_{2.5}



concentrations exceeded the new EU limit of $10 \mu\text{g m}^{-3}$, which will come into force in 2030. Ground level measurements of $\text{PM}_{2.5}$ from the European Air Quality Monitoring Network for 2021 and 2022 show a striking gradient between clean and polluted areas. Eastern European regions and the Po Valley in Italy have the highest annual mean concentrations, while central and western Europe have much lower PM levels (EEA, 2019). Simulations of $\text{PM}_{2.5}$ exposure and $\text{PM}_{2.5}$ related mortality for the year 2015 by Gu et al. (2023) also indicate higher concentrations and associated health risks in Eastern Europe.

The inflow of air masses from the east can lead to a transfer of polluted air masses to areas with less emissions. Previous studies for Germany during periods of long-range transport from eastern Europe have identified combustion processes as a major contributor to regional background concentrations (van Pinxteren et al., 2019, 2016). The inflow of air masses from the east was associated with PM_{10} concentration peaks leading to an increase in exceedances of the current daily limit value of $50 \mu\text{g m}^{-3}$ (van Pinxteren et al., 2019). The influence of multiple sources and the transboundary transport of primary and secondary particles contributing to local $\text{PM}_{2.5}$ concentrations is still not fully understood. This needs to be better characterised to enable effective and better targeted mitigation strategies to address the prevailing air quality challenges.

Source apportionment (SA) studies aim to link ambient concentrations of pollutants to their emission sources. Within chemical transport models (CTM), two main methods can be used to do this: the emission reduction impact method and the mass transfer method. The emission reduction impact method, or brute force approach, assesses how pollutant concentrations respond to specific emission changes (Thunis et al., 2019). An extreme case, the 'zero-out' method, sets emissions from selected sources to zero and estimates their maximum possible impact on ambient concentrations. This approach helps to assess the potential impact of emission reductions on air quality (Clappier et al., 2017). Despite its conceptual simplicity, this method is computationally intensive and the results are highly sensitive to the reference scenario chosen.

The mass transfer method, implemented in CTMs as the tagged species approach, estimates contributions from different source sectors and regions by tracing the mass transport of pollutants from emission sources to local concentrations (Thunis et al., 2019). In this method, new tracers are introduced for the pollutants of interest and labeled according to their emission sources, allowing them to be monitored throughout the model run (Kranenburg et al., 2013). This approach facilitates the study of source contributions across both spatial and temporal scales, with source definitions directly linked to the emission inventories used as model inputs (Mircea et al., 2020). Tagged species modules for particulate source apportionment are implemented in several chemistry transport models: e.g. in LOTUS-EUROS (Kranenburg et al., 2013), in DEHM (Brandt et al., 2013), PSAT (Particulate Matter Source Apportionment Technology) in CAMx (Yarwood et al., 2007), TSSA (Tagged Species Source Apportionment) (Wang et al., 2009) and ISAM (Integrated Source Apportionment Method) (Kwok et al., 2013) in CMAQ (US EPA Office of Research and Development, 2024). Tagging approaches are not designed to assess the effectiveness of mitigation measures or the impact of emission reductions because they do not consider indirect chemical effects (Thunis et al., 2019). However, they do provide a direct, additive source attribution of pollutant mass concentrations.

This study aims to improve the understanding of the interaction between dispersion and transformation processes by investigating an area of large $\text{PM}_{2.5}$ concentration gradients in Central Europe. Therefore, we implement a non-reactive tagged species approach into the online Eulerian chemical transport model COSMO-MUSCAT (Wolke et al., 2012). The tagging ap-



proach is applied to identify PM sources with a focus on winter combustion emissions. Online and offline measurements from an extensive campaign in 2021 are used to validate the simulations and to improve the understanding of the local air quality.

2 Observations and modeling

2.1 Sampling sites

The TRACE winter campaign took place from 05 February 2021 to 24 March 2021 at three measurement sites in central Europe: two of the stations, Melpitz (DE) and Košetice (CZ), were already well established as part of ACTRIS (Aerosol, Clouds and Trace Gases Research Infrastructure) and EMEP (European Monitoring and Evaluation Programme), while the third (Frýdlant, CZ) was specifically installed for this project. The sites were selected to capture an important area of transition between polluted and less polluted regions in Central Europe (see Fig. 1).

The research observatory Melpitz (51.54° N, 12.93° E, 86 m a.s.l.) is located 50 km east of Leipzig, Germany, observing atmospheric background conditions in Central Europe. It has been operated by TROPOS for more than 30 years (Spindler et al., 2001; Poulain et al., 2011). The station is surrounded by grassland and flat agricultural land without any notable wind obstacles. About 60% of the time throughout the year, the prevailing wind direction is south-west. These air masses are of maritime origin and reach Melpitz after having crossed Western Europe and, in the immediate vicinity, the city of Leipzig. The second main wind direction is East (about 17% of the time), with dry continental air masses influenced by long-distance transport from Poland, Belarus, Ukraine, Slovakia and the Czech Republic (Spindler et al., 2001, 2012, 2013).

The National Atmospheric Observatory Košetice (49.35° N, 15.05° E, 534 m a.s.l.) is situated 60 km south-east of the Prague metropolitan area in Czech Republic. There are several small settlements in the vicinity of the station, however, the district is one of the least populated in the country (Zíková and Ždímal, 2013). Surrounded mainly by agricultural land and some woodland, a medium-sized timber factory equipped with a biomass furnace is located 7.5 km from the site (Schwarz et al., 2016). In winter, air masses reaching the site predominantly originate from South-West (44%) passing over Central Europe (Pokorná et al., 2022). Similarly, Lhotka et al. (2025) observed that the contribution of continental air masses was higher in winter compared to other seasons, highlighting a distinct seasonal difference.

The Frýdlant temporary measurement site (50.94° N, 15.07° E, 366 m a.s.l.) was set up 2 km north of the centre of Frýdlant, Czech Republic, close to the Polish border. The station is located on the north-western edge of the Jizera Mountains and is surrounded by forests and farmland. The Turów Coal Mine, a large Polish open pit mine, is about 10 km south-west of the site. Lignite from the Turów mine is used to fuel the nearby Turów power station.

During the campaign period, the COVID-19 pandemic continued to affect Europe, with containment measures still in place. In Germany, there was a lockdown from 13 December 2020 to 3 March 2021. Non-essential businesses, schools, and childcare facilities were closed, and employees were required to work from home wherever possible. Essential services such as supermarkets, pharmacies, and healthcare facilities remained open. From 3 March 2021, restrictions were adjusted locally based on infection rates and other factors (BMG, 2023). In the Czech Republic, strict restrictions were in place until 11 April 2021 (Slabá, 2022). In Poland, a partial lockdown was enforced from 28 December 2020 to 14 February 2021. Some restrictions

**Table 1.** Measuring devices used at the three sites to obtain the data for this study.

species	device	resolution		station		
		time	size	Melpitz	Frýdlant	Košetice
OM, SO_4^{2-} , NO_3^- , NH_4^+ , Cl^-	Aerodyne HR-ToF-AMS	2.5 min	PM_{10}	x		
	Aerodyne c-ToF-AMS	5 min	PM_{10}		x	
	Aerodyne ToF-ACSM	5 min	PM_{10}			x
OC, EC	Sunset Lab OC-EC offline Aerosol Analyzer	12 h	$\text{PM}_{2.5}$	x	x	x
	Sunset Lab OC-EC online Aerosol Analyzer	2 h	PM_{10}		x	
	Sunset Lab OC-EC online Aerosol Analyzer	4 h	$\text{PM}_{2.5}$			x
eBC	Magee Scientific AE33	1 min	PM_{10}	x	x	x
PM	Digitel High Volume Aerosol Sampler	12 h	$\text{PM}_{2.5}$	x	x	x
PAH	GC/MS	12 h	$\text{PM}_{2.5}$	x	x	x
Anhydromonosaccharides	Agilent HP 6890 gas chromatograph and HP 5973 mass selective detector	12 h	$\text{PM}_{2.5}$	x	x	x

were eased on 1 January 2021, allowing shops in shopping centers and cultural institutions to reopen. However, on 20 March 2021, stricter measures were reintroduced until 9 April (A3M Global Monitoring GmbH, 2023).

2.2 Measurement data

A multi-device setup for data acquisition was in place at all three stations. The data presented in this study were measured with the instruments listed in Table 1. Instrumentation included, aerosol mass spectrometer for the non-refractory near PM_{10} chemical composition (organic, nitrate, sulfate, ammonium and non sea-salt chloride) and a multi-wavelength aethalometer for the equivalent black carbon (eBC) connected to a dry PM_{10} inlet. The mass concentration of $\text{PM}_{2.5}$ was measured by gravimetric filter sampling using a Digitel high-volume aerosol sampler with pre-heated quartz fiber filters. Samples were collected for 12 hours, covering daytime (5:00 to 17:00 UTC) and nighttime (17:00 to 5:00 UTC). The filters were further analysed with a Sunset Lab thermal-optical transmittance (TOT) instrument according to the EUSAAR2 temperature protocol (Cavalli et al., 2010). Online Sunset OC-EC data are also available for Frýdlant and Košetice. Carbon parallel plate diffusion denuders were used to remove volatile organic compounds to prevent positive sampling artefacts caused by adsorption of gas phase organics onto the filter (Turpin et al., 2000). The AMS/ACSM instruments measure total organic matter (OM), which we can compare directly with our model output, while the two Sunset instruments detect the organic carbon (OC). For better comparability, OC was converted to OM using an OM/OC ratio from literature. Poulain et al. (2011) estimated an OM/OC ratio of 1.64 at the Melpitz station in winter 2009, with almost no diurnal variation. For a winter campaign in 2020 in Košetice Pokorná et al. (2022) found a ratio of $1.51 (\pm 0.36)$. In this study we have applied a factor of 1.6 to the conversion of all Sunset data. Polycyclic aromatic hydrocarbons (PAHs) were detected on the filters by gas chromatography-mass spectrometry (GC-MS). For a detailed description of the measurement campaign see Arora et al. (in preparation).



2.3 Model description

110 All simulations were carried out with the multiscale model system COSMO-MUSCAT. It consists of two online coupled components, the regional numerical weather forecasting model COSMO (Consortium for Small scale MOdelling) (version 5.05, Schättler et al., 2018) in conjunction with the air-chemistry transport model MUSCAT (MUltiScale Chemistry Aerosol Transport) (Wolke et al., 2012), developed at TROPOS. The model system is designed for aerosol-chemistry process studies and air quality applications at the regional scale (Hinneburg et al., 2008; Heinold et al., 2011; Tönnissen et al., 2021; Wolke et al., 115 2012), and participated in model intercomparisons such as the Air Quality Model Evaluation International Initiative (AQMEII; Im et al., 2015; Galmarini et al., 2021). COSMO is a nonhydrostatic atmospheric numerical weather forecasting model based on the primitive thermo-hydrodynamic equations describing compressible flow in a moist atmosphere. The atmospheric equations are solved based on a terrain-following grid with rotated coordinates (Schättler et al., 2018). The meteorological model provides all the necessary meteorological fields (e.g. wind, relative humidity, temperature) to MUSCAT, which then simulates 120 the transport and chemical transformations in the atmosphere for different gas and particle phase species. Transport processes include advection and turbulent diffusion, while physical loss processes are characterised by dry and wet deposition (Wolke et al., 2012).

Anthropogenic emissions of atmospheric compounds are treated as prescribed point and gridded area sources. Emissions within Germany are provided by the GRETA database of the German Federal Environment Agency (UBA) (Schneider et al., 125 2016) for the year 2019 (resolution: 0.5×1 km). For European emissions outside Germany the CAMS-REG-v5 emission inventory for the year 2018 (resolution: 6 km) is used, provided by the Copernicus Atmosphere Monitoring Service (CAMS) (Kuenen et al., 2022). Emissions are treated according to the Gridded Nomenclature For Reporting (GNFR) (NFR-I, 2023), i.e. they are grouped into different emission sectors representing different source types (e.g. Public Power, Traffic; see Table 2). The temporal variation of emissions (daily, weekly and seasonal cycle) is accounted for by time profiles, which differ according 130 to the emitting sector. These temporal profiles are largely based on those provided with the TNO_MACC-II inventory (Kuenen et al., 2014), with adjustments for livestock and agriculture emissions according to Skjøth et al. (2011). Emissions are provided as aggregated totals for some pollutant groups, which we then break down into individual components. Primary particulate matter (PM) is split into elemental carbon (EC), primary organic matter (OM), sulphate (SO_4^{2-}), sodium and other minerals. A further distinction is made between fine ($< 2.5 \mu\text{m}$) and coarse ($2.5 - 10 \mu\text{m}$) aerosol particles. Non-methane volatile organic 135 compounds (NMVOC) emissions are divided into 23 different hydrocarbon groups. The splitting profiles for PM and NMVOC are based on different literature sources and are also provided by CAMS (Kuenen et al., 2022). In this study, country-specific splitting profiles (based on the year 2017) are applied to the overall emission input.

The emission of biogenic VOC (BVOC) is based on Steinbrecher et al. (2009) and improved for extended land use categories according to Luttkus et al. (2022). The primary natural aerosol components are emitted online in COSMO-MUSCAT. The 140 estimation of desert dust mobilization depends on soil texture and soil size distribution according to Tegen et al. (2002) and preferential source regions (Heinold et al., 2011; Schepanski et al., 2017) using the current wind fields and hydrological conditions provided by COSMO. The emission of sea spray aerosol is based on Barthel et al. (2019).



Natural fire emissions (e.g. EC, OM and primary PM_{2.5}) are provided as point sources for the year 2021 by the Global Fire Assimilation System (GFAS) (Kaiser et al., 2012). These emissions are resolved into 24-hour mean values with a specific injection height for each point source.

Dry deposition is modelled using the resistance approach described by Seinfeld and Pandis (2006). Aerosol particles and trace gases are also removed from the atmosphere through wet deposition, subdivided into in-cloud and below-cloud scavenging. Both processes are parameterized by size-dependent particle capture efficiencies and corresponding gas uptake coefficients (Simpson et al., 2012).

To describe the gas-phase chemistry, an extended version of the Regional Atmospheric Chemistry Mechanism RACM-MIM2-ext (Karl et al., 2006; Stockwell et al., 1997; Karl et al., 2009) is used. The mass-based aerosol population is described using a hybrid bulk-bin scheme. It comprises 25 prognostic aerosol particle tracers, including primary PM_{2.5} and PM₁₀, primary OM, EC, sulphate, nitrate and ammonium, secondary organic aerosol (SOA), as well as six bins for sea salt and primary marine organic particles (diameter range: 0.01–10 µm) and five desert dust bins (0.2–48 µm).

Secondary inorganic aerosol is formed through reactions between ammonia and sulfuric or nitric acid, which are generated from the gaseous precursor species sulfur dioxide (SO₂) and nitrogen oxides (NO_x) (Hinneburg et al., 2008). The partitioning between the particle and gas phases depends on the ambient atmospheric temperature and humidity. The implementation of this particle/gas partitioning follows the equilibrium approach described by Galperin and Sofiev (1998), utilizing the methods proposed by Mozurkewich (1993).

The formation of SOA is described by the module SORGAM (Schell et al., 2001), extended to include additional biogenic volatile organic compound (BVOC) precursors from isoprene, monoterpene and sesquiterpene oxidation and highly oxygenated molecule (HOM) formation from all considered BVOCs (Luttkus et al., 2022). The module uses the two-product approach described by Odum et al. (1996), which splits each SOA product class –comprising reaction products from aromatic precursors, alkanes, alkenes, α-pinene, and limonene– into two pseudo-products. For each, the formation of low volatility products and their gas/particle partitioning is simulated.

Low volatility condensable products are formed through oxidation of organic precursor gases by the OH radical, the nitrate radical NO₃ and ozone. The amount produced is determined by a product species (i) dependent stoichiometric coefficient (α_i) in the specific reaction of the chemical mechanism (Schell et al., 2001). Then the SOA mass resulting from gas-particle partitioning is calculated using a partitioning coefficient K_{om,i} for each low volatile product species following Pankow (1994). The partitioning coefficient depends on temperature and is influenced by the molecular weight and saturation vapor pressure of species i. Each pseudo-product consists of a gas phase and particle phase product with different α_i and K_{om,i}. The total SOA yield (Y) resulting from the two previous steps can be calculated according to the equation (1), where M_o is the total available absorbing organic matter (Odum et al., 1996).

$$Y = \sum_{i=1}^n Y_i = M_o \sum_{i=1}^n \left(\frac{\alpha_i K_{om,i}}{1 + K_{om,i} M_o} \right) \quad (1)$$



175 Over a range of organic mass concentrations M_o , a precursor gas will have a range of aerosol yields Y . The relationship between yield and organic mass concentration can be determined through chamber measurements. To model this relationship, a curve is fitted by selecting the optimal values of α_1 , α_2 , $K_{om,1}$ and $K_{om,2}$ within the two-product framework. The sum of all particle phase products considered gives the total SOA concentration.

2.4 Model setup

180 The domains for the COSMO-MUSCAT simulations were chosen to cover the three measurement sites. To reduce computational costs for the targeted horizontal resolution in the measurement region, the model is nested twice. The innermost domain TraceD1 covers 317×204 grid cells with a horizontal resolution of ~ 2 km (see Fig. 1). The vertical resolution for COSMO in TraceD1 is 50 layers with a maximum height of 22 km, while MUSCAT uses only the lowermost 27 layers, i.e. up to ~ 6 km. A common grid nesting approach is used for the inner domains. The results of the larger domains are used as lateral boundary conditions on the inner domains. The meteorological initial and boundary conditions for the European domain (N0) are provided by reanalysis data of the CAMS global atmospheric composition forecasts (Inness et al., 2019). The simulation covers the period from 1 January to 31 March 2021, including a one month spin-up, with an output resolution of 1 hour. The model system is re-initialized every 48 h using the aerosol and trace gas concentrations at the end of the previous run and a 24 h COSMO pre-run to spin-up the meteorology in order to avoid long-term drifts in the modelled meteorology.

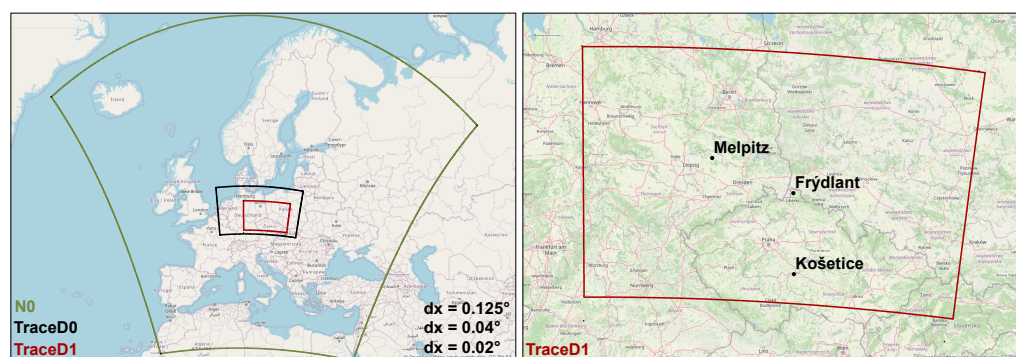


Figure 1. Domains for COSMO-MUSCAT runs and localization of the three rural background sampling sites (OpenStreetMap contributors, 2017).

190 2.5 Source attribution in COSMO-MUSCAT

A source attribution module has been developed for COSMO-MUSCAT 5.05 to analyse the influence of specific source regions, point sources, and emission sectors on primary particulate matter compounds. This new tagging method allows the individual tracking of emitted source-specific species during a single model simulation, thus enhancing the analytical capabilities of the model. Unlike the "zero-out" method, which requires multiple simulations for each source sector or region of interest, this new

**Table 2.** GNFR source categories considered in this study.

GNFR	Source Category	Source Composition
A	Public Power	Public electricity and heat production
B	Industry	Oil and gas refining, coal mining, iron and steel industry, chemical industry, pulp and paper industry, food and beverages industry, cement production
C	other Combustion	Small combustion processes of private households, small businesses, agriculture, forestry and fishing
D	Fugitives	Fugitive emissions from oil and gas, exploration, production, transport and distribution of oil and natural gas
F1	Traffic: Gasoline	Exhaust from gasoline powered vehicles
F2	Traffic: Diesel	Exhaust from diesel powered vehicles
F4	Traffic: Non-Exhaust	Brake wear, tyre wear, gasoline evaporation and road wear
I	Off Road	Railways, off-road vehicles and other machinery, mobile combustion
K	Livestock	Enteric fermentation and manure management
L	Agriculture	Application of manure and fertilizer, indirect emissions from managed soils, storage, handling and transport of agricultural products, use of pesticides
	Other	All other sectors are combined here: Product/solvent use, traffic: LPG/natural gas, shipping, aviation, waste treatment

195 approach eliminates this need. As a result, the analysis is faster and less computationally intensive. To do so, an additional tracer is introduced into the model emissions for each species of interest from each defined source sector or source region, and combinations of both. This additional tracer is labeled with the source information and then processed in parallel. In this way, the concentration of each of the so-called tagged tracers is available in each grid cell of the model and at each time step. This provides detailed spatial and temporal information about the source contribution to local tracer concentrations. In addition to

200 the concentration of each tagged species, the total concentration - representing the cumulative impact of all sources - is also available. This allows the relative contribution of each tagged source to be effectively calculated. An overview of the selected source sectors is given in Table 2. Tags for source regions can be specified via a text-based input file in which each surface grid cell can be assigned a region name. For this study, we have tagged emissions from all countries within the inner domain TraceD1, namely Germany, Poland and the Czech Republic. Additionally, a source region 'Boundary' is introduced referring

205 to the transport from the coarser domains to the inner domain. Input from outside the European domain is not included in the 'Boundary' tagged sector.

Transport (advection, diffusion, sedimentation) and removal (dry and wet deposition) processes are automatically applied to tagged tracers in the same way as for all other tracers. However, gas phase chemistry and aerosol chemistry are not considered at present. Therefore, only chemically passive tracers can be tagged, i.e. non-reactive tagging approach. This enables a high spatial

210 and temporal resolution analysis of the source composition of primary particles. As this study focuses on winter combustion processes, anthropogenic EC and OM emissions are tagged. EC and OM emissions are split into fine and coarse aerosol, therefore the same split is applied for the tagged tracers.



3 Results

3.1 Meteorology

215 During the campaign notable meteorological events and sharp temperature changes occurred in a short period of time. In early February, a low pressure system with cold air in the north and warm air in the south moved southwards, transporting cold air to the Balkans and Greece. On 7 and 8 February, strong easterly winds and heavy snowfall led to significant snow drifts in some areas of Central Europe. This was followed by a week of clear nights with prevailing westerly winds and temperatures dropping to -20°C . The model successfully captured the period of low temperatures and the snow event at all three stations (see 220 Fig. 2). The snow event was followed by a cold episode resulting in more stagnant conditions with a change in wind direction and decreased wind speed at all stations.

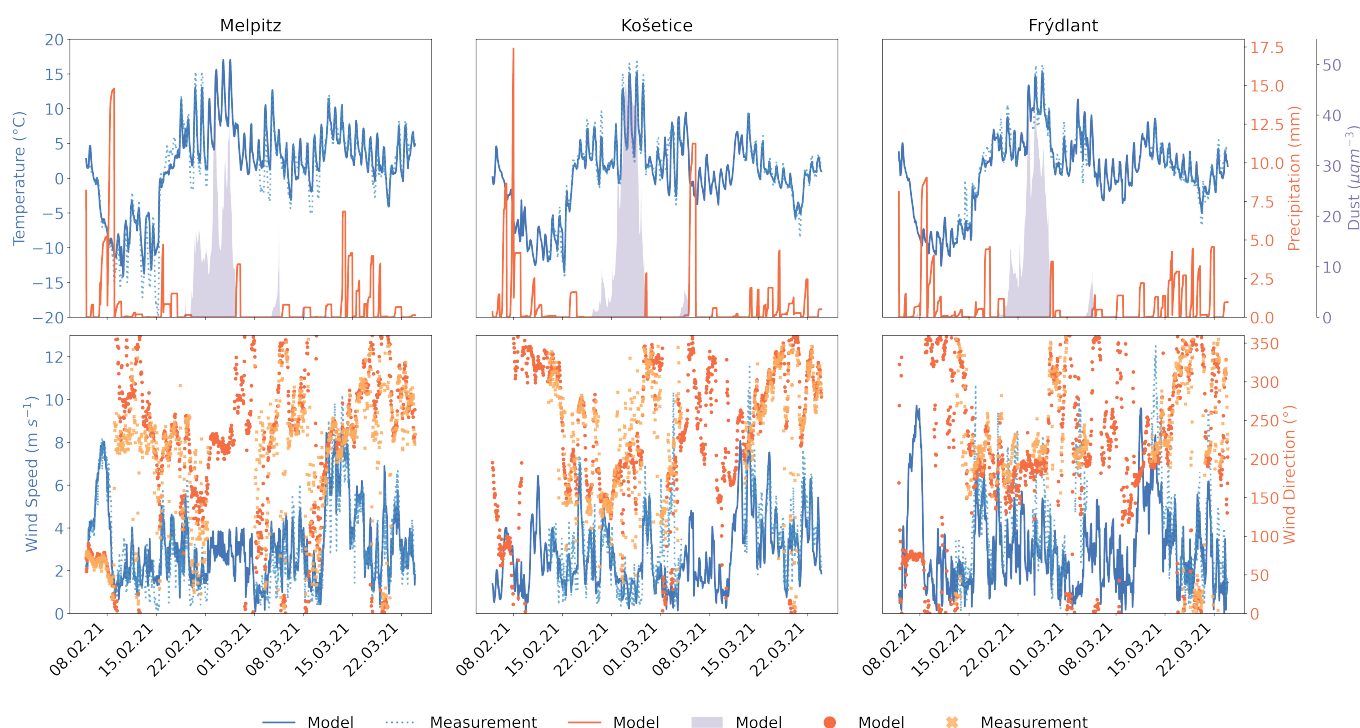


Figure 2. Meteorological parameters for the three stations. The top row shows surface temperature and precipitation, with the shaded area representing surface dust concentration in the size class $< 80 \mu\text{m}$. The bottom row displays the modeled surface wind speed and direction.

In mid-February, a nearly stationary high pressure system transported warm air from the Sahara into Central Europe, driving a rapid temperature increase of up to 20°C within a week. An omega blocking pattern over Eastern Europe facilitated the inflow of dust that accompanied the warm air, allowing particles to travel as far north as Scandinavia (Hoshyaripour, 2021; 225 Haarig et al., 2022). This event significantly affected all three stations, resulting in elevated surface dust concentrations of up



to $50 \mu\text{g m}^{-3}$ (see Fig. 2). The unusually high, spring-like temperatures persisted until the end of February. Another significant dust event occurred on 3 March, originating from the Sahara and affecting Central Europe. Although our model successfully simulated dust uptake, surface concentrations during this event remained lower than those observed in mid-February. In mid-March, a shift to westerly winds brought low-pressure systems accompanied by widespread precipitation over Germany. This was followed by an intrusion of cold air from the polar regions, resulting in sleet, snow, and gusty winds (DWD, 2021a, b). By the end of March, atmospheric blocking patterns established stable and dry conditions.

3.2 Measurements and model capability

To validate the model's performance, we compared the modeled $\text{PM}_{2.5}$ concentrations and their components with observational data. In our model, $\text{PM}_{2.5}$ comprises mineral dust, sea salt, organic matter (including primary organic carbon and secondary organic aerosol), elemental carbon, ammonium sulfate, and ammonium nitrate. Figure 3 presents the time series of the total $\text{PM}_{2.5}$ concentration. The model does not accurately represent the magnitude of the concentration peaks, especially at Košetice, where the average modelled values are almost $10 \mu\text{g m}^{-3}$ below the observed values (see Table A1 in the Appendix). The Root Mean Squared Error (RMSE) quantifies the error between measured and modeled surface-level mass concentrations. Overall, the model underestimates $\text{PM}_{2.5}$ levels, with RMSE values of $14.26 \mu\text{g m}^{-3}$ for Melpitz, $13.85 \mu\text{g m}^{-3}$ for Košetice, and $10.92 \mu\text{g m}^{-3}$ for Frýdlant. The high RMSE values indicate that the model tends to underestimate concentrations during periods of high concentration peaks, as the RMSE is particularly sensitive to outliers. The Pearson correlation coefficient (R) provides an indication of the overall fit of the trend, ranging from -1 to 1. An absolute value of exactly 1 means that a linear equation perfectly describes the relationship between model and measurement. Among the sites, the $\text{PM}_{2.5}$ trend is most accurately captured in Košetice ($R=0.61$) and least accurately in Melpitz ($R=0.25$).

Stern et al. (2008) conducted a comparative study analysing the performance of five chemical transport models during a high PM_{10} episode over Central Europe in 2003. The study found that all models underestimated both primary and secondary species during peak concentrations. The authors were unable to identify a single source of error, but noted that weather conditions characterised by low inversion heights, high stability and low wind speeds are particularly difficult to simulate with prognostic models. Boundary layer schemes often underestimated inversion strength and overestimated inversion height, leading to excessive vertical mass exchange. Additionally, underestimation of emissions in the applied inventory further contributed to discrepancies. During the first two weeks of February, the TRACE campaign revealed the largest discrepancies between observed and simulated $\text{PM}_{2.5}$ concentrations, with most other tracers also underestimated. Strong easterly winds until 8 February facilitated long-range pollutant transport to Melpitz and Frýdlant. The snow event on 7–8 February caused only a slight decrease in $\text{PM}_{2.5}$ concentrations, indicating limited washout effects. Concentrations rose again after the snow event, peaking around 10 February. The snow event was followed by a cold episode with stagnant conditions, reduced wind speeds, and a shift in wind direction, leading to pollutant accumulation. The model may underestimate residential emissions due to missing temperature dependencies and unaccounted COVID-19 lockdown effects. Increased heating activity due to unusually cold temperatures and limited mobility combined with stagnant meteorology could lead to the observed underestimation of total pollutants. Considering only data from February 15 onwards improves model performance, reducing the RMSE to $9.64 \mu\text{g m}^{-3}$.

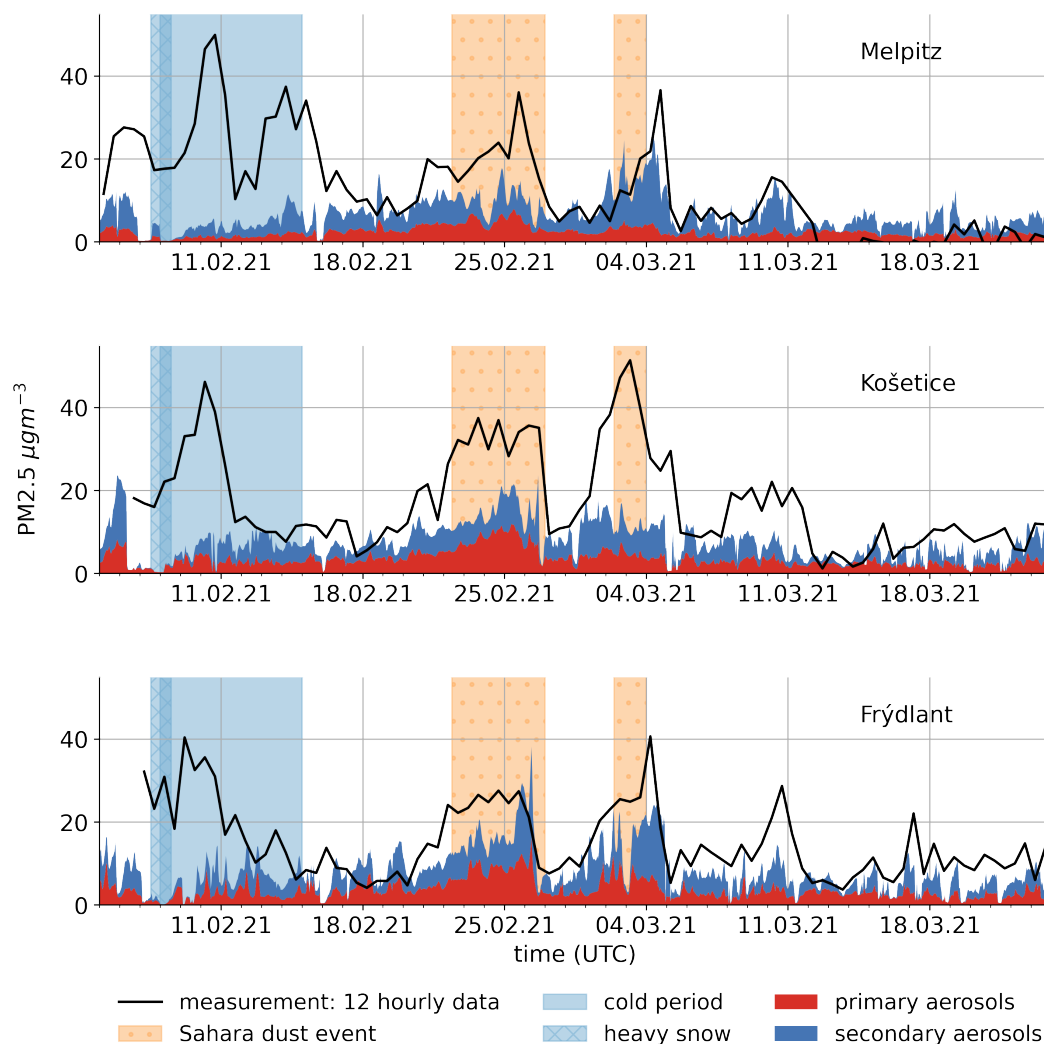


Figure 3. Time series for PM_{2.5} mass concentration for the three stations. Filter data compared with modelled primary and secondary aerosol mass concentration. The timestamp for the filter data corresponds to the time of filter collection.

260 for Melpitz, 12.44 µgm⁻³ for Košetice, and 7.63 µgm⁻³ for Frýdlant. Additionally, the overall trend is better captured, with R increasing to 0.79 in Košetice, 0.65 in Frýdlant, and 0.61 in Melpitz.

To gain a better understanding of the remaining discrepancies between modelled and measured PM_{2.5}, we can evaluate the accuracy for each individual PM_{2.5} component (see Figure 4).

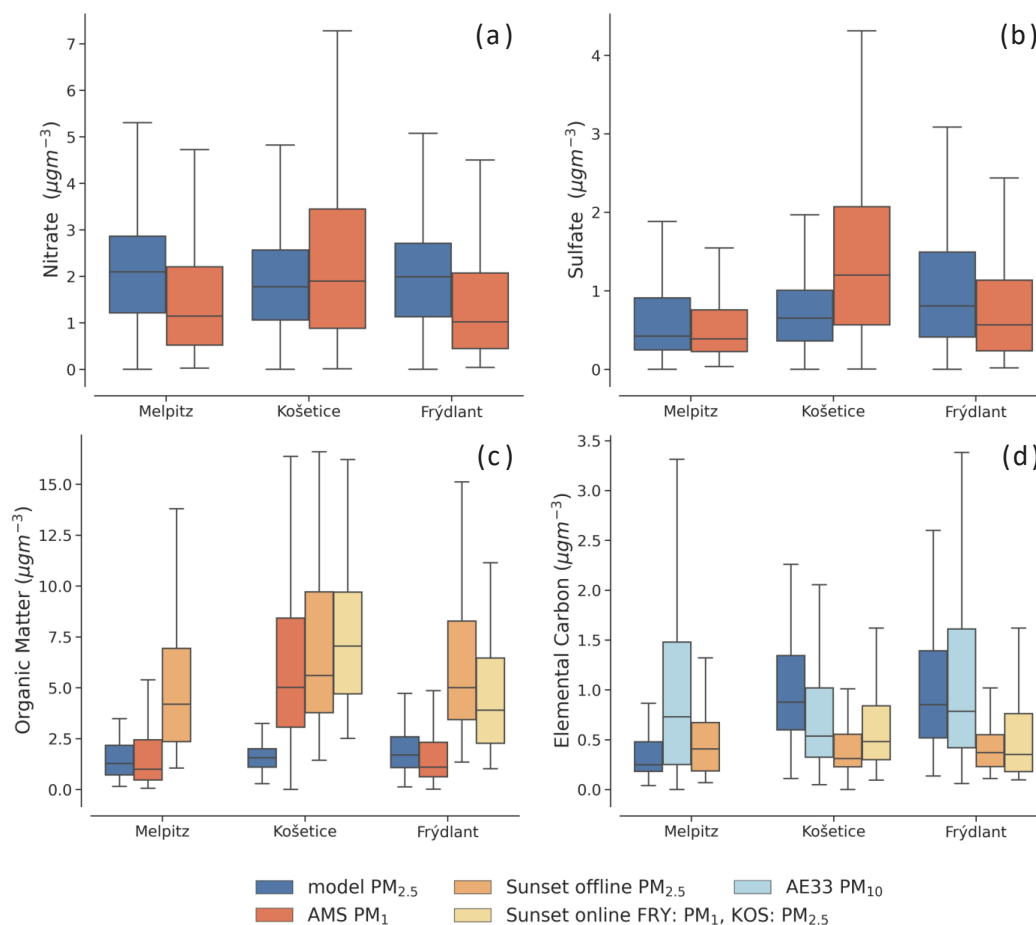


Figure 4. Box and whisker plots for observations and modelled data at corresponding times during the campaign period. Rectangular boxes display the first and third quartiles. The line within each box represents the median value. Outliers are excluded. For OM and EC all data was averaged to 12 hours. For comparability with AMS measurements, a factor of 1.6 was applied to the OC measured by the Sunset instruments.

Mineral Dust

265 The Saharan dust outbreaks likely influenced the total PM_{2.5} concentrations during the TRACE campaign. In the model, the February event brought high dust loads for several days and led to dust deposition at all three stations (see Fig. 2). Lidar measurements in Leipzig recorded pure dust conditions, but below 3 km, aerosol from continental Europe was likely mixed into the Saharan dust plumes (Haarig et al., 2022). This event had a rather short travel time (less than two days) before reaching Leipzig. For the March event, the model also shows dust reaching the three stations, though the loads were not as high as

270 during the second event. Observations by Haarig et al. (2022) detected mixed pollution-dust conditions after air masses were



transported over Spain and France, reaching Leipzig after 3-4 days. It is possible that the model underestimated surface $\text{PM}_{2.5}$ during these events, potentially due to limitations in the model domain or insufficient vertical mixing to the surface.

Nitrate and Sulfate

275 Secondary inorganic aerosol is well captured by our model (see Figure 4 (a) and (b) and Fig. A3/A4 in the Appendix) and are not likely to cause the discrepancy between predicted and measured aerosol mass concentrations. Košetice shows the lowest correlations and a slight underestimation of nitrate and sulfate concentrations. In Melpitz and Frýdlant the results for sulfate are reasonable while nitrate is slightly overestimated.

Elemental Carbon

EC concentrations show an overall good agreement with observations (see Fig. 4 (d)). Our model aligns more closely with the
280 Aethalometer data in Košetice (RMSE: $0.66 \mu\text{g m}^{-3}$) and Frýdlant (RMSE: $0.93 \mu\text{g m}^{-3}$) than in Melpitz, where it agrees well with the offline Sunset measurements (RMSE: $0.37 \mu\text{g m}^{-3}$). The discrepancy between Aethalometer and Sunset measurements arises from the different carbon fractions they detect: Aethalometers measure optically absorbing carbon (black carbon) in PM_{10} , while Sunset instruments quantify elemental carbon (see Fig. A2 in the Appendix). In Košetice and Frýdlant, our model slightly overestimates EC concentrations. For winter 2019, Aethalometer measurements reported $0.98 \pm 0.76 \mu\text{g m}^{-3}$ BC in
285 Košetice (Lhotka et al., 2025), while Pokorná et al. (2022) found $0.92 \pm 0.77 \mu\text{g m}^{-3}$ for winter 2020. In comparison, our averaged model result for 2021 was $0.76 \mu\text{g m}^{-3}$. In Melpitz, literature data show significant variability in BC concentrations. Atabakhsh et al. (2023) reported a value of $1.38 \mu\text{g m}^{-3}$ converted to PM_1 using a multi-angle absorption photometer (MAAP) during winter 2016/2017. Later, van Pinxteren et al. (2023) observed a marked decrease to $0.5 \pm 0.41 \mu\text{g m}^{-3}$ in winter 2018/2019, likely reflecting reduced emissions and meteorological influences.

290 Organic Matter

The modelled OM that we refer to further is the sum of the fine primary organic aerosol (OM in $\text{PM}_{2.5}$), the total SOA and OM from outside the European simulation domain. Primary OM accounts for approximately half of the total OM, with mean contributions of 44% in Melpitz, 48% in Frýdlant and a slightly higher share of 57% in Košetice (see Fig. A1 in the Appendix). Panel (c) in Fig. 4 compares all available OM values for our campaign period. The underestimation of these values by our model
295 seems to have a large contribution to the total $\text{PM}_{2.5}$ underestimation. A comparison of the modelled concentrations with the AMS/ACSM PM_1 values gives a similar picture to that for the $\text{PM}_{2.5}$ mass concentration (see Fig. A1 in the Appendix). The model underestimates the OM concentrations in Košetice (RMSE: $6.48 \mu\text{g m}^{-3}$) while for Melpitz and Frýdlant the overall fit is good (RMSE: 1.17 and $2.01 \mu\text{g m}^{-3}$). The model matched the measurements well, except for the first two weeks of February. Compared to the offline Sunset measurements, the RMSE is higher for Melpitz and Frýdlant (4.95 and $5.18 \mu\text{g m}^{-3}$), but
300 improves notably when considering only data from February 15 onwards, decreasing to 2.87 and $3.85 \mu\text{g m}^{-3}$, respectively. The discrepancy between AMS and Sunset in Melpitz and Frýdlant may partly be caused by the different size classes. In



Košetice the ACSM and both Sunset instruments give consistent results, while the modelled data is noticeably lower. For Košetice, the same concentration levels for PM₁ and PM_{2.5} size class OM indicate a dominance of fine aerosol, while there is few coarse mode organic aerosol. The correlation coefficient of the model concentrations against the AMS measurement is lower (0.39) than that of the filter samples (0.63). If only the SOA components of the modelled OM are taken into account, the correlation coefficient compared to the AMS for Košetice decreases further to 0.18. For Frýdlant and Melpitz, calculating the correlation coefficient using only SOA gives similar results to using the total modelled OM concentration. The reduced correlation at Košetice when isolating SOA implies that the model underestimates secondary aerosol at this site, thereby negatively affecting the overall correlation.

Previous Sunset filter measurements taken at Melpitz in winter 2018/2019 found an averaged value of $3.2 \pm 3.2 \mu\text{g m}^{-3}$ (van Pinxteren et al., 2023). AMS data for winter 2009 also gives comparable values $2.08 \pm 1.6 \mu\text{g m}^{-3}$ (Poulain et al., 2011) while measurements with an ACSM in winter 2016/2017 show higher values of $6.21 \mu\text{g m}^{-3}$ (Atabakhsh et al., 2023). For Košetice, a good characterisation of the site is also given by various previous studies. AMS measurements provide average values of $3.13 \mu\text{g m}^{-3}$ in winter 2019 (Lhotka et al., 2025) and $4.55 \pm 4.40 \mu\text{g m}^{-3}$ in winter 2020 (Pokorná et al., 2022). Mbengue et al. (2018) found an average OC concentration in PM_{2.5} of $2.85 \pm 1.91 \mu\text{g m}^{-3}$ for the period 2013 - 2016.

The discrepancy in modelled PM_{2.5} concentrations does not appear to be driven by deviations in elemental carbon (EC), sulfate, or nitrate concentrations. Instead, the underestimation of organic matter (OM) accounts for a significant portion of this mismatch. The overall good agreement between modelled and observed EC concentrations indicates a reliable simulation of primary aerosol emissions. In contrast, the substantial discrepancy between modelled and measured OM concentrations likely stems from secondary aerosol.

A spatial variation in the model's performance is apparent, with similar trends observed in Melpitz and Frýdlant, whereas Košetice exhibits distinct behaviour. The dominance of fine particles in OM, suggested by nearly identical concentrations in the PM₁ and PM_{2.5} size fractions, points to elevated levels of secondary particles. Given the general agreement with previous campaigns, we can also conclude that our campaign, although characterised by unusual meteorological conditions during the first weeks, represents typical aerosol conditions at these sites. Therefore, the underestimation could indicate a general underrepresentation of SOA during winter in this area in COSMO-MUSCAT.

3.3 Source attribution for elemental carbon and primary organic matter

Since the model accurately reproduces EC concentrations, which represent a primary anthropogenic aerosol component, we conclude that anthropogenic sources are well represented in the model, enabling reliable identification of source contributions. Additionally, with approximately half of the total OM comprised of POA, we infer that overall source profiles can be effectively identified by analysing primary OM and EC using the non-reactive tagging approach. The results, shown as relative contributions to primary OM and EC for the cold and warm period (Fig. 5), underline the importance of long-range transport of particles. The source sector 'Boundary' represents transported particles from the outer model domains into the innermost domain where tagging is applied. During the warm period, long-range transport accounts for about 38% of both EC and OM in Melpitz, illustrating the significant influence of particles originating outside the domain. In Košetice the contribution is 23.8%



for OM and 22.8% for EC, while Frýdlant has the lowest influence with 16.6% for OM and 14.6% for EC, respectively. The prevailing wind regime and the basin-like topography of the Czech Republic reduce the influence of long-range transport at Košetice compared to the other two stations. The 'Boundary' contribution to fine OM and EC is only slightly higher than for Frýdlant, which is located in the middle of the domain (see Fig. 8). Backward trajectory analyses (HYSPLIT; Stein et al., 2015) indicate that during the high PM peak event in early February, stationary meteorological conditions resulted in minimal air mass transport to all sites. This effect is particularly pronounced in Košetice, where strong local stagnation can be observed.

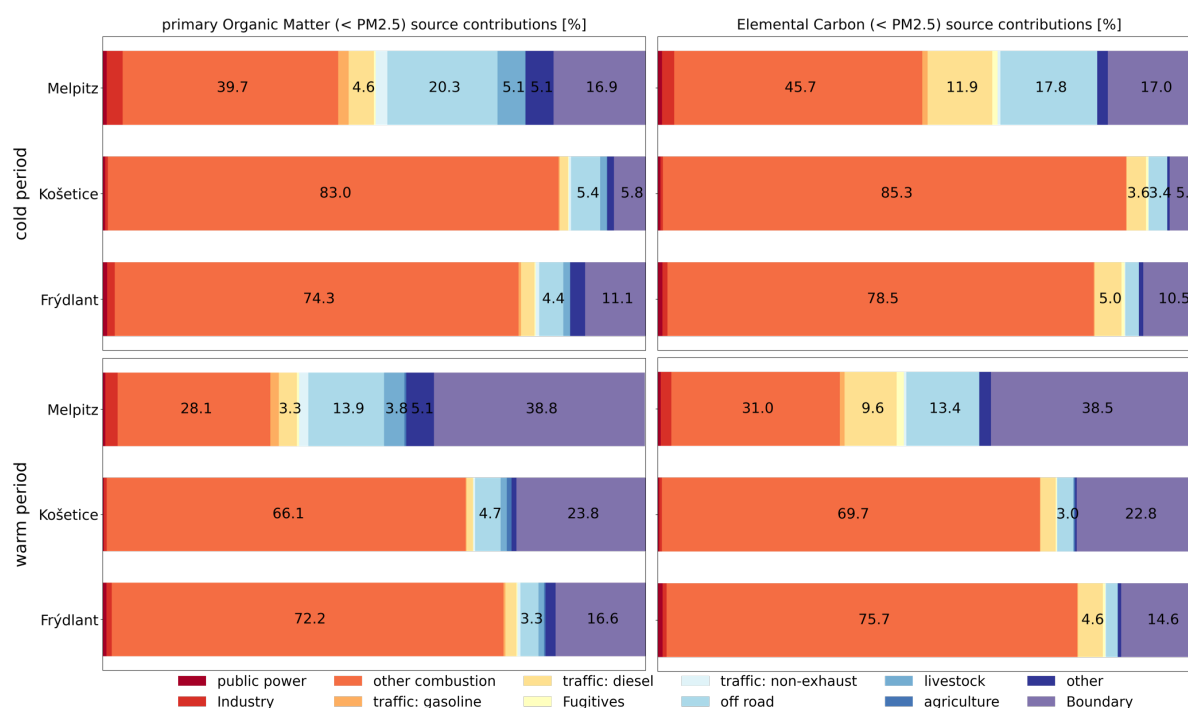


Figure 5. Relative source contributions to primary organic matter < PM_{2.5} and elemental carbon < PM_{2.5}. Top: cold period (05.02.2021 - 16.02.2021), bottom: warm period (16.02.2021 - 23.03.2021)

The study region, spanning parts of Germany, Poland, and the Czech Republic, is characterized by a high density of active lignite mines (see Fig. 6). Lignite, a particularly emissions-intensive fuel, is the energy source for many large power plants in this area. Germany and Poland host the largest number of coal-fired power plants with the highest total capacities in the EU (Alves Dias et al., 2018). Emissions from power plants used for electricity and heat production are categorized under the source sector 'Public Power'. Despite its proximity to areas with a high number of coal-fired power plants, the 'Public Power' sector contributes only a small share to the overall concentration of primary OM and EC. Tagging results for this sector, split by country of origin, are shown in Figure 7, indicating that the peaks at Frýdlant are predominantly driven by Polish emissions. The proximity of the Turów lignite power plant largely explains the observed peaks, especially during periods of low wind speeds. During other periods, emissions from German and Czech sources dominate. The influence of coal burning on air

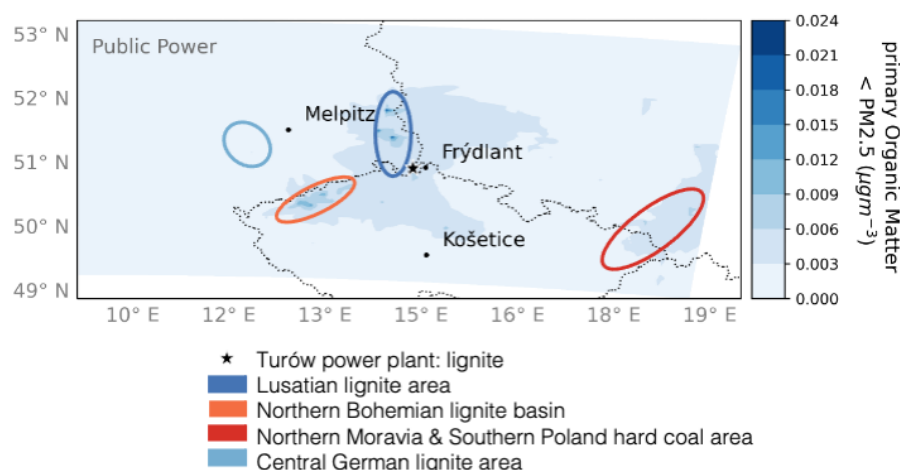


Figure 6. Spatial distribution of the average absolute contribution of emissions from the source sector Public Power to the concentrations of primary organic matter $< \text{PM}_{2.5}$. Areas with many coal-fired power plants are highlighted.

quality in Frýdlant is further amplified by its use in domestic heating. In 2017, 47.7% of Polish households with individual heating relied on coal (Macuk, 2019).

The sector 'other Combustion' includes combustion processes of private households, in particular domestic heating processes with all fuel types. This sector has the biggest contribution with up to 76.3% for EC and 72.6% for primary OM in Frýdlant. Contributions to fine OM from the 'other Combustion' sector are highest in the Czech Republic and in urban agglomerations in Poland and around Berlin, Germany (see Fig. 8). Unlike the 'Public Power' sector contributions, the main contributors to the concentrations observed at the stations are emissions originating within the country where the station is located. However, Melpitz stands out with the highest proportion of contributions from cross-border emissions. Atabakhsh et al. (2023) carried out a positive matrix factorization (PMF) analysis over a period of one year at Melpitz. They found the highest coal combustion contribution to POA under the influence of easterly continental air masses. Furthermore, they found a temperature and RH dependence for the factor consisting of aged SOA and highly oxidised OA in winter, with the highest concentrations observed at temperatures below 0°C and RH above 80%. They concluded that increased precursor emissions due to higher heating activities and amplified aqueous phase chemistry lead to increased SOA formation. This could suggest a potential additional underestimation of the SOA formation rate in early February, as strong easterly winds were observed, followed by a subsequent cold period.

Chen et al. (2022) conducted a multi-year PMF source apportionment study across various locations in Europe. They identified a coal combustion factor of primary OA at only two sites: Melpitz (data collected in 2016/2017) and the urban location Kraków (data collected in 2018). The strong seasonal variations in this factor suggest it originates from residential heating emissions. The study also examined Košetice, where no coal combustion factor was detected; however, biomass burning accounted for 15.5% of the total OA in winter 2019. Lhotka et al. (2025) conducted a PMF study with data also collected in 2019

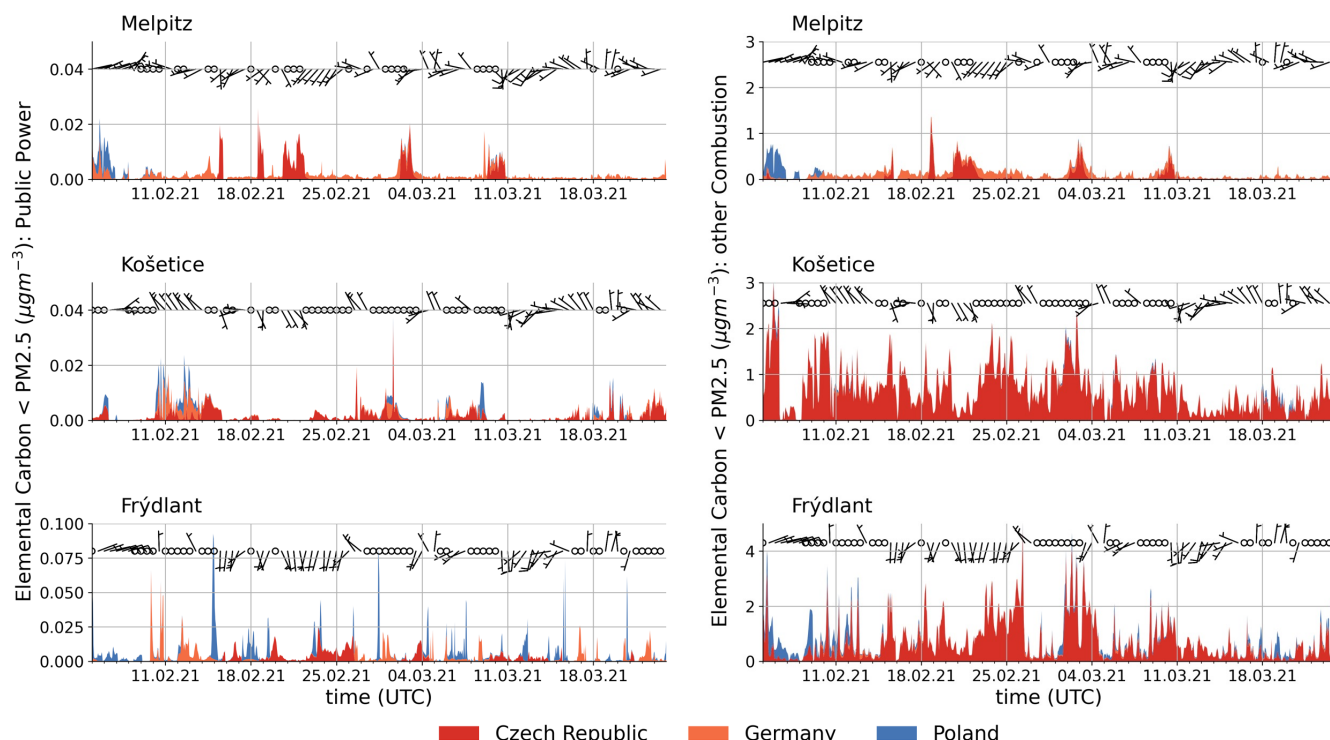


Figure 7. Elemental carbon < PM_{2.5} concentration, broken down by country of origin and source sector. Left: 'Public Power', right: 'other Combustion'. The wind barbs represent modelled 12-hour averages for each station.

in Košetice. They identified a coal combustion factor with the highest contribution of 5% to total OA in spring, while biomass combustion contributed most in winter (12% of total OA). Both factors showed similar diurnal cycles related to domestic heating, and a strong correlation between levoglucosan and the biomass combustion factor was observed in winter, indicating a high proportion of wood combustion. During a particularly cold period in January 2019, an increased contribution of coal was observed, probably due to its increased use in private households for heating, given its higher calorific value compared to wood. The results are also consistent with those of Horník et al. (2024), who performed a PMF study with samples collected during the TRACE campaign for water-soluble organic compounds using NMR. They found a high residential heating contribution with coal markers indicating additional coal combustion in early February in Košetice.

Pokorná et al. (2018) analysed changes in PM_{2.5} composition and sources from the 1990s to 2009/2010 in Košetice. During this period, the dominant sources shifted from lignite combustion by power plants and oil combustion to residential heating, mainly with coal and/or biomass. In the Czech Republic only 5% of total coal consumption in 2019 was used in the residential sector, as part of the 'other combustion' source sector (IEA, 2021). Hovorka et al. (2015) conducted a receptor modelling study in a residential area 64 km north-east of Prague in winter 2013, and estimated that wood burning contributed 49% to the mass

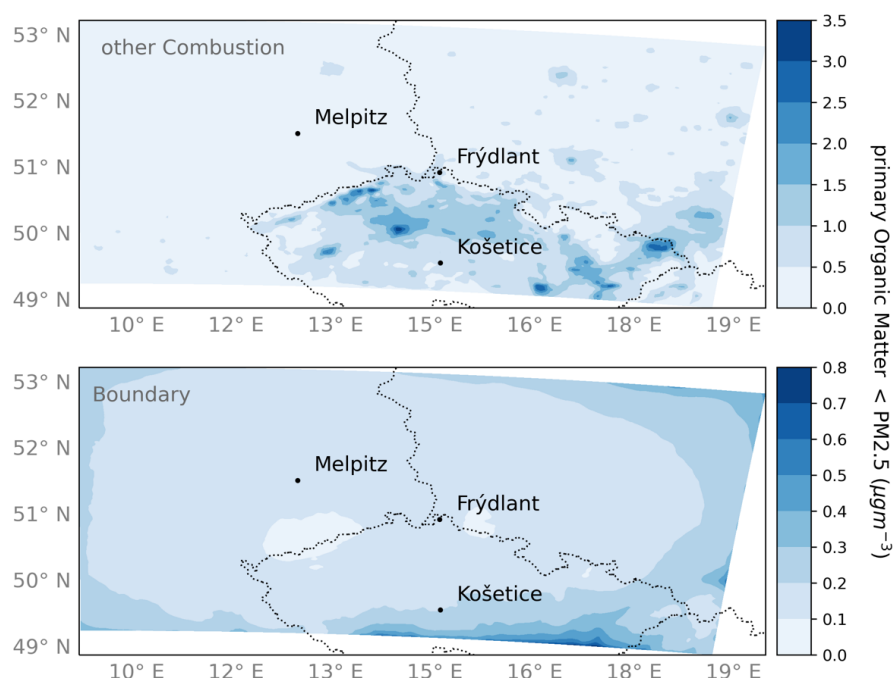


Figure 8. Spatial distribution of the average absolute contribution of emissions from source sectors 'other Combustion' (top) and 'Boundary' (bottom) to the concentrations of primary organic matter $< \text{PM}_{2.5}$.

of fine aerosol. They found high correlations between contributions from wood combustion and levoglucosan and suggested
385 that wood combustion in local boilers is common in suburban areas in the Czech Republic.

The landscape surrounding Košetice is mainly agricultural with scattered woodland, the only direct sources of pollution are local roads and domestic heating (Zikova and Zdimal, 2016). It is plausible to assume high rates of wood burning, given the proximity of the timber factory. Levoglucosan, an aerosol tracer which is associated with biomass burning, measured during the TRACE campaign show highest mean concentrations in Košetice ($0.32 \mu\text{g m}^{-3}$) and lowest in Melpitz ($0.15 \mu\text{g m}^{-3}$).
390 Polycyclic aromatic hydrocarbons (PAH) are also good tracers of combustion processes, e.g. retene is a unique marker of wood combustion (Ramdahl, 1983). The average retene concentration in Košetice is 2.13 ng m^{-3} at average total PAH concentration of 24.43 ng m^{-3} . In Frýdlant the averaged total PAH concentration is comparable (24.73 ng m^{-3}), but retene concentrations are lower (1.01 ng m^{-3}). Melpitz shows similar retene concentrations as Frýdlant (1.16 ng m^{-3}) at lower total PAH levels (14.12 ng m^{-3}). The high relative and absolute levels of retene and levoglucosan in Košetice are a good indicator for a high
395 contribution of wood burning (Arora et al., in preparation). The results are also consistent with those of Horník et al. (2024), who reported high levels of levoglucosan in Košetice and Frýdlant. Overall, the results indicate a strong influence of wood burning for domestic heating during winter in the Košetice area. During particularly cold periods, residents appear to supplement wood with coal, leading to a greater local impact of coal emissions on air quality. The higher coal contributions observed



in Melpitz seem to be mainly driven by long-distance transport, whereas in Frýdlant, additional contributions from the nearby
400 power plant are evident.

3.4 Effects of COVID-19 containment measures

With containment measures still in place during the campaign, the daily lives of many of the region's citizens were disrupted. Different patterns of mobilization, the closure of businesses and changes in leisure habits are all factors that can affect air quality. The emission inventories used in this study do not take into account exceptional events affecting emissions, such as the
405 COVID-19 restrictions. Several studies have looked at the impact of these restrictions on air quality. Most of them focus on the year 2020, when the pandemic peaked.

Gkatzelis et al. (2021) reviewed over 200 papers to assess the impact of lockdowns on air quality around the world. They found significant reductions in NO₂ and CO levels, small reductions in PM_{2.5} and increases in O₃ concentrations. The effects varied by season and region, and the study highlighted the need for future research to include meteorological corrections for
410 accurate results. Only about a third of the studies reviewed included methods for meteorological correction or normalisation.

Matthias et al. (2021) conducted a modelling study for Central Europe, estimating emission reductions for January to June 2020. For secondary pollutants, they found that meteorological effects outweigh the effect by emission reductions from restrictions. Putaud et al. (2023) compared measurements at 28 sites across Europe for spring 2020 with CAMS ensemble forecasts and found a slight decrease in PM_{2.5} and PM₁₀ during the lockdown and a strong increase after the measures were lifted. The
415 study corrects the occurring bias between modelled and measured values by a time-dependent normalisation of the CAMS forecasts to the observations estimated from 2019 data. They concluded that the increased ozone levels due to reduced NO_x lead to altered oxidation capacities and therefore more SOA formation. The study also analysed data collected in Melpitz and Košetice before, during and after the lockdown in March 2020. In Melpitz, slightly higher PM_{2.5} concentrations than expected by CAMS were detected during the lockdown. In May 2020, after the lockdown, they were even twice as high as modelled.
420 In Košetice, the values before and during the lockdown were slightly below the expected values, while the concentrations afterwards were 30% higher. Forster et al. (2020) calculated emission trends based on Google mobility data for six sectors (land transport, residential, energy, industry, public and aviation) per country. These data show that in March 2021, BC emissions from the residential sector in Germany and the Czech Republic were increased by approximately 10%, while BC emissions in all sectors combined were decreased by about 20% compared to a baseline scenario. Mbengue et al. (2023) conducted an
425 extended study analysing the effects of the COVID-19 lockdowns at Košetice using normalisation techniques to account for meteorological effects. They found that during the winter of the second lockdown (December 2020 - February 2021), dispersion normalised concentrations of EC were reduced by 28%, while OC and SOC concentrations increased by 19% and 51%, respectively. They concluded that this was due to a greater influence of emissions from local domestic activities. Considering that our study sites are background stations with low traffic influence and high contribution of domestic heating emissions,
430 locally increased emissions due to the COVID-19 mitigation measures seem plausible, leading to higher PM_{2.5} and probably SOA concentrations than without these measures. These changes are not included in the emissions in the model and may be another source of underestimation in the model.



4 Anthropogenic secondary organic aerosol

Domestic heating, a major source of primary elemental carbon and organic matter, also emits gases that serve as precursors to anthropogenic SOA. Previous source apportionment studies have shown that residential heating is a significant contributor to SOA formation. Lhotka et al. (2025) identified a relationship between primary organic aerosol (POA) and oxidized organic aerosol (OOA) source factors associated with residential heating. The high contribution of highly oxidized OA in winter can be attributed to the local influence of biomass burning. In contrast, at Melpitz, coal combustion plays a more prominent role in oxidized OA formation, indicating the impact of long-range transport (Atabakhsh et al., 2023). An intensive tagging study by Bartík et al. (2024) utilized the PSAT module in CAMx, supplementing the CAMS emission inventory with more detailed residential emission data for the Czech Republic and additional intermediate - volatility organic compound (IVOC) emissions from wood combustion. Their findings indicate that VOC and IVOC emissions from the 'Other Combustion' sector represent the largest source of SOA in Central Europe during winter, contributing up to $0.4 \mu\text{g m}^{-3}$. Bergström et al. (2012) found an underestimation of winter organic aerosol in a modelling study focusing on several years in Europe. Their conclusion was that emissions from wood combustion are under-represented in current emission inventories. In order to investigate whether a potential underestimation of SOA precursors from domestic heating has contributed to the lower than expected concentrations of OM in our model, we have carried out a sensitivity study.

4.1 Sensitivity study

The parameterisation of SOA is influenced by two key variables: the precursor gases emitted and the rate at which SOA is formed from these precursors. Previous studies suggest, that phenol is a significant component of emissions from incomplete combustion processes like wood burning. Phenol is one of the key gaseous precursors responsible for the formation of SOA during biomass burning activities (Hatch et al., 2015; Bruns et al., 2016). Liu et al. (2024) highlight the critical role of nighttime NO_3 oxidation of anthropogenic VOCs from biomass combustion, a process they find is inadequately represented in current atmospheric models. Their results show that increasing both phenol emissions and the associated SOA yield leads to a twofold increase in SOA production via NO_3 oxidation across Europe during winter. In our model, phenol is included in the lumped species CSL (cresol and other aromatics). NMVOC emissions are delivered by the UBA and CAMS emission inventories (Schneider et al., 2016; Kuenen et al., 2022). The NMVOC emission flux is split into the different relevant model species based on emission profiles created by Theloke and Friedrich (2007) for 306 individual species including phenol. These profiles are based on a NMVOC source database from 1990 (Olivier et al., 1996) and do not include phenol emissions from domestic heating. Therefore, it is reasonable to assume that the VOCs emitted from domestic heating are not fully captured by the model. Natural fire emissions provided by GFAS do not include CSL, but toluene (TOL) and xylene (XYL) emissions. Given the low impact of natural fires in winter in Europe, it can be assumed that they do not contribute much to the formation of secondary particles.

The SORGAM module (see section 2.3) estimates SOA formation from aromatic precursors using data from Odum et al. (1997), who conducted smog chamber experiments to quantify SOA production from gasoline vapor. This method is therefore



primarily tailored to traffic emissions. However, for aromatic precursors emitted by domestic heating, an increase in SOA yield aligned with phenol SOA formation rates is more suitable. Due to the limited availability of chamber studies on phenol gas-phase SOA formation, we derived a new yield estimate based on four OH oxidation measurements from Yee et al. (2013). Given the importance of nocturnal oxidation, we also applied these modifications to the NO_3 reaction. A non-linear least squares fit for the α_i values was performed with fixed $K_{\text{om},i}$ coefficients ($K_{\text{om},1} = 0.2899$, $K_{\text{om},2} = 0.0103$). As α_2 yielded negative values, we decided to keep α_2 fixed and performed the fit solely for α_1 (see Fig. A5 in the Appendix). These adjustments result in an approximately threefold increase in SOA yield.

Further, an adjustment of the input emission was done. To get a good representation of phenol emissions from domestic heating processes, we decided to scale the emissions to the CO emissions of the emission sector 'other combustion'. Following wood combustion chamber studies from Bruns et al. (2016) on average all NMVOC emissions make up 0.22 times the CO emissions. According to Schauer et al. (2001), phenol and substituted organic compounds are approximately 10% of the overall NMVOC emissions from wood combustion. Accordingly, we set our 'other Combustion' sector CSL emissions to 0.022 times the sector's CO emissions.

We simulated three sensitivity runs to compare these adjustments. First with the adjusted SOA yield alone (S1), second with the new CSL emissions alone (S2), and third with both combined (S3). Table 3 gives an overview of the coefficients and emissions used in the different sensitivity runs and the original base run. All sensitivity runs were performed for our middle domain, TRACED0 (see Fig. 1), as it provides the best trade-off between spatial resolution and area coverage. The three sensitivity runs were not nested, but use the same initial and boundary conditions as the base run.

Table 3. Sensitivity simulation setup.

simulation	stoichiometric coefficient		CSL emissions
	α_1	α_2	'other Combustion'
base run	0.039	0.108	CAMS NMVOC split
S1	0.219	0.108	CAMS NMVOC split
S2	0.039	0.108	$0.022 \times \text{CO emissions}$
S3	0.219	0.108	$0.022 \times \text{CO emissions}$

4.2 Sensitivity study results

The changes in CSL emission flux and the corresponding mean OM concentration across the three sensitivity runs are presented in Table 4. The values for each station represent the model result from the 4×4 km grid cell in which the station is located. In scenario S1, the increased SOA yield for aromatic precursors has the most pronounced effect in urban areas, as it influences emissions from all source sectors, including industry and transport. The adjusted SOA yield applies to both daytime OH oxidation and nighttime NO_3 oxidation. Among the stations, Melpitz experiences the highest relative increase (39%) due to high aromatic precursor concentrations.

**Table 4.** Changes in CSL emission flux and mean organic matter $< \text{PM}_{2.5}$ for all sensitivity runs compared to the base run.

	CSL emission [$\mu\text{g m}^{-2} \text{s}^{-1}$]	OM mean [$\mu\text{g m}^{-3}$]		
	S2, S3	S1	S2	S3
domain	+ 0.00001 (0.08%)	+ 0.27 (18%)	+ 0.07 (4%)	+ 0.62 (39%)
Melpitz	- 0.0016 (- 60%)	+ 0.52 (39%)	+ 0.21 (16%)	+ 0.77 (58%)
Košetice	+ 0.0115 (202%)	+ 0.21 (13%)	+ 0.12 (7%)	+ 0.67 (40%)
Frýdlant	+ 0.0230 (188%)	+ 0.42 (23%)	+ 0.28 (15%)	+ 0.95 (53%)

In scenario S2, the emissions of aromatics from domestic heating are introduced as CSL emissions by the sector 'other Combustion'. Although total CSL emissions across the domain remain constant, their spatial distribution shifts: emissions decrease in Melpitz but increase significantly in Frýdlant and Košetice. The domain-wide mean OM concentration shows an overall modest increase of 4%, with the largest increases observed in the central Czech Republic and southern Poland, where domestic heating sources are abundant. Interestingly, despite a reduction in CSL emissions at Melpitz compared to the base run, OM concentrations at Melpitz increase similarly to those at Frýdlant (+16% at Melpitz and +15% at Frýdlant). This is attributed to increased CSL emissions in the surrounding areas and the transport of SOA and its precursors to the site. These findings align with previous studies: Poulain et al. (2011) linked winter OM at Melpitz to transported particles, while Spindler et al. (2012) reported that SOA concentrations peaked in winter air masses arriving from the east, highlighting the role of anthropogenic precursor-driven SOA formation during long-range transport. This is also consistent with the conclusions of Atabakhsh et al. (2023).

In the combined S3 run, average OM concentrations in Melpitz increase by 58%, the highest relative impact among all stations. This results from enhanced SOA transport and formation from aromatic precursors. Frýdlant shows the largest absolute OM increase, with an average increment of $0.95 \mu\text{g m}^{-3}$. Figure 9 compares diurnal OM cycles from the base and sensitivity runs with measurements. At Frýdlant and Košetice, the combined adjustments in S3 produce greater impacts on OM concentrations compared to the individual sensitivity runs. At Melpitz, however, the difference between S1 and S3 is less distinct, suggesting that higher baseline precursor concentrations already contribute significantly to SOA formation at this site. The correlation of the modeled SOA with AMS data improves in Košetice, with the correlation coefficient increasing from 0.18 to 0.29, while Melpitz and Frýdlant show no significant improvement. Although the model now better reflects SOA contributions at Košetice, overall OM concentrations remain underestimated.

The spatial pattern of the increase in OM concentrations in S3 compared to the base run is shown in Fig. 10. The strongest increases occur in the Czech Republic and southern Poland. In particular, the city of Prague and its outskirts show a high increase, which is consistent with the small residential houses in the suburbs and surrounding areas still using coal and wood combustion for heating (Schwarz et al., 2008; Makeš et al., 2021). Domestic heating emissions likely increased during the COVID-19 measures as more people stayed at home, contributing to higher $\text{PM}_{2.5}$ and possibly SOA concentrations (Mbengue et al., 2023). Additionally, the campaign coincided with an unusually cold period in early February, likely elevating heating activity further — an effect not fully captured by the model.

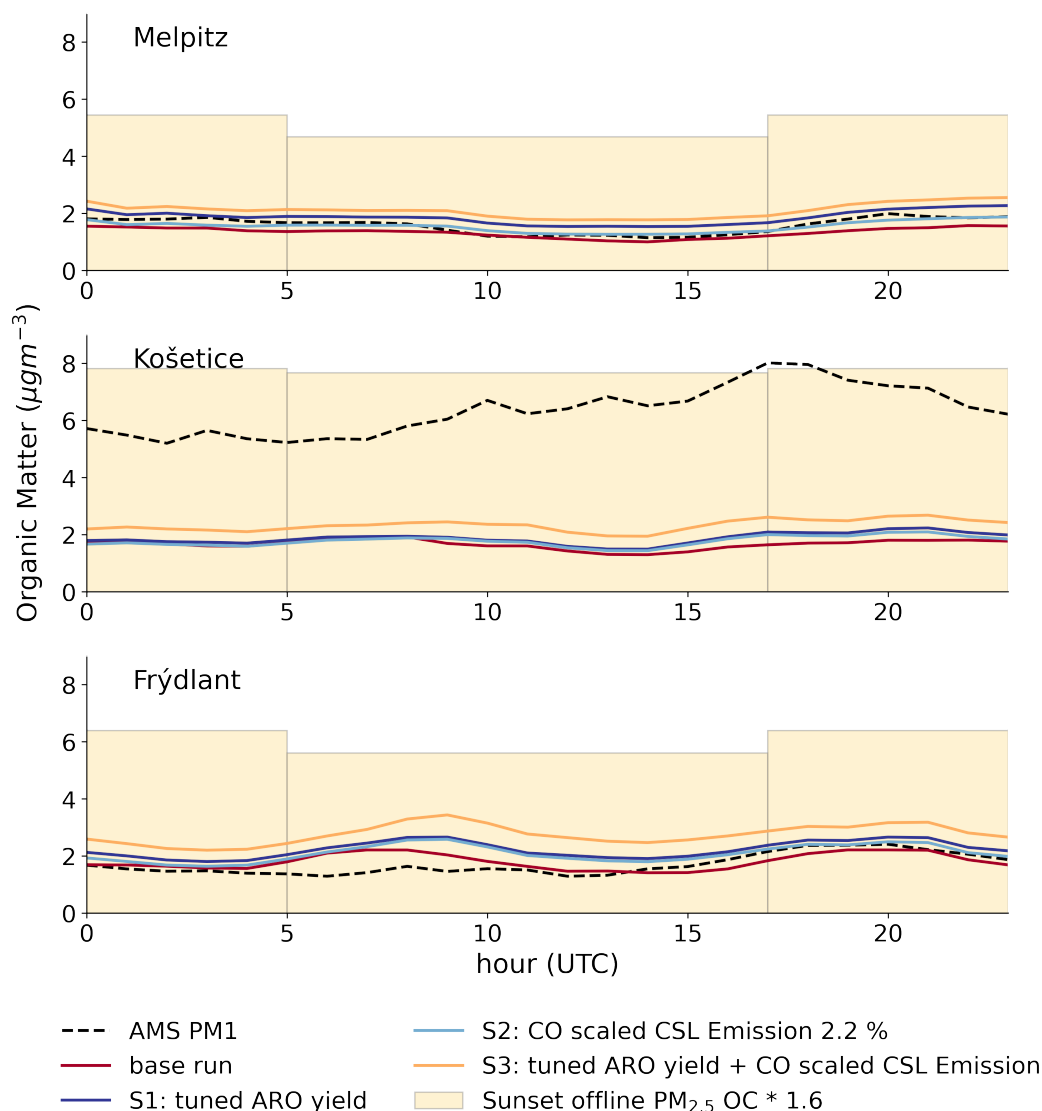


Figure 9. Hourly daily cycle over the entire measurement period for organic matter $< \text{PM}_{2.5}$ for all sensitivity runs compared to the base run. Bar graphs showing the daily averaged filter data for 12-hour sampling periods.

The simulation of the OM peak in early February, after the snow event, shows no noticeable improvement with the sensitivity runs, likely linked to the overall underestimation of emissions (see Fig. A7 in the appendix). Measurements reveal a distinct OM concentration peak on 3 March, particularly at Košetice and Frýdlant. The S3 run captures the peak reasonably well at Frýdlant but still underestimates OM at Košetice. HYSPLIT backward trajectories for Frýdlant on this date indicate a significant influence from air masses passing over the Czech Republic and Košetice (see Fig. A6 in the Appendix). At Košetice,

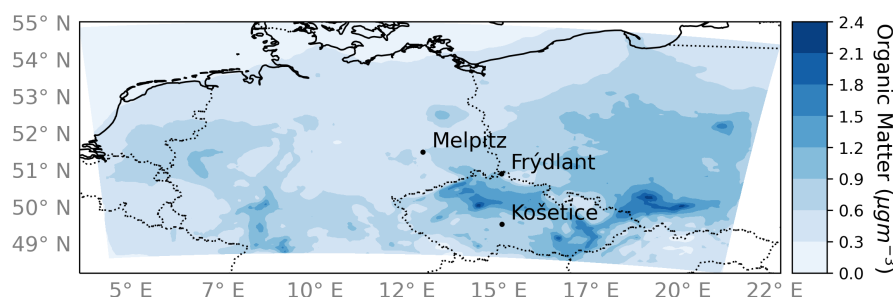


Figure 10. Difference between the averaged modeled organic matter $< \text{PM}_{2.5}$ in the S3 and base simulation.

wind direction shifts from east to west, as simulated by the model, while at Frýdlant, wind direction remains steady, allowing precursor accumulation and increased SOA formation.

Overall, the sensitivity studies showed that scaling AVOC emissions from wood combustion to residential heating emissions improves the spatial distribution of SOA in the study area. Long-range transport of precursors and SOA is captured as well as the local influence on OM concentrations.

5 Conclusion, Limitations and Future Directions

The study investigates the sources of primary and secondary anthropogenic organic aerosol in Central Europe during winter. The chemical transport model COSMO-MUSCAT was used to analyse concentrations of particulate matter, in particular particles originating from combustion processes. The model results were compared with measurements made in Germany and the Czech Republic in terms of overall $\text{PM}_{2.5}$ concentration and concentrations of individual species. The model underestimated the total $\text{PM}_{2.5}$, especially during high concentration peaks. A pronounced underestimation occurred in early February, likely due to the prevailing meteorological conditions combined with changed heating behaviour. During this period, all tracers were underestimated, whereas after early February, the model accurately captured the behaviour of most tracers. However, the underestimation of $\text{PM}_{2.5}$ during concentration peaks remained evident. The discrepancies in modeled $\text{PM}_{2.5}$ concentrations do not appear to be due to deviations in EC, sulfate, or nitrate levels, but rather to the underestimation of OM. Although the present study reproduced total OM values well at two monitoring sites, measurements at Košetice are underestimated, partly due to an inadequate representation of SOA formation from residential heating (wood combustion), a major source of anthropogenic VOCs. These AVOCs contribute considerably to the formation of SOA, and it is likely that their insufficient representation in our model contributes to the overall underestimation of OM during winter. The effect is most pronounced in the central Czech Republic, where the basin-like topography allows air masses to linger, promoting the accumulation of emissions and extended SOA formation. We found a higher contribution of domestic heating in the eastern part of our study region, which is accompanied by high concentrations of OM, especially at the station in Košetice. Sensitivity tests with adjustment for SOA yields and AVOC emissions showed an average increase in OM concentrations of over 40% at the measurement sites. It is probable



that the model not only underestimates SOA precursors from already included domestic heating sources, but the underlying inventory may also miss additional unaccounted sources. The actual impact of these precursors on SOA formation may be even higher than in the simulation runs. In addition, the model may underestimate the contribution of SOA precursors other than phenol. There is a need for more detailed and up-to-date emission inventories that provide information on the types of fuels used, their spatial distribution and time profiles. Our findings highlight that regional domestic heating emissions contribute significantly to overall air pollution in the study area. Addressing these emissions is complex, as they are hard to quantify and regulations for private households are more challenging to implement. Consequently, obtaining more detailed information on these sources is vital for developing targeted and feasible measures. Besides updated time profiles representing seasonal, weekly and daily patterns of emissions, changing heating behaviour due to extreme meteorological conditions could be taken into account by implementing a temperature dependence of emissions. The Heating Degree Day (HDD) approach, introduced by Guevara et al. (2021), considers the influence of outdoor temperature on heating activity and its associated emissions. A recent study by Guion et al. (2024) enhanced this method by incorporating country-specific and species-specific parameters, demonstrating improved temporal correlations and more accurate detection of PM emission threshold exceedances compared to simulations using fixed parameters or monthly temporal factors. Implementing this approach in COSMO-MUSCAT could enhance the accuracy of our model results during winter. In addition, heating emissions may not only be underestimated in quantity, but the contribution of different fuel types to the domestic heating sector may also vary with temperature, as additional coal burning in households may occur during colder periods.

Comparing the non-reactive tagging approach in COSMO-MUSCAT to measurement-based, receptor-oriented source apportionments can further evaluate its capability and identify areas for improvement. This comparison can provide valuable insights into the performance of the model and guide future refinements. By addressing these gaps and incorporating the necessary updates, such as updated emission inventories, improved SOA yields and model evaluation through comparison with measurement data, the model could provide a more comprehensive and accurate representation of SOA formation processes, enabling better understanding for air quality management.

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570 **Appendix A**



Table A1. Time-averaged measured and modeled mass concentrations and the associated Root Mean Squared Error (RMSE) and Correlation Coefficient (R) for the whole campaign period. The modeled data were adjusted to match the measurement intervals before statistical analysis. Online refers to in situ measurements and offline to filter sampling. A factor of 1.6 was applied to the OC measured by the Sunset offline instrument.

		model	online	offline				
		mean [$\mu\text{g m}^{-3}$]	mean [$\mu\text{g m}^{-3}$]	RMSE [$\mu\text{g m}^{-3}$]	R	mean [$\mu\text{g m}^{-3}$]	RMSE [$\mu\text{g m}^{-3}$]	R
PM _{2.5}	Melpitz	6.80	-	-	-	12.43	14.26	0.25
	Košetice	7.61	-	-	-	17.24	13.85	0.61
	Frýdlant	8.17	-	-	-	15.07	10.92	0.34
OM (AMS PM ₁)/	Melpitz	1.34	1.59	1.17	0.60	5.06	4.95	0.24
OM (offline PM _{2.5})	Košetice	1.66	6.37	6.49	0.39	7.74	8.12	0.63
	Frýdlant	1.81	1.71	2.01	0.19	6.18	5.18	0.48
eBC (AE33 PM ₁₀)/	Melpitz	0.36	1.00	1.06	0.35	0.47	0.37	0.29
EC (offline PM _{2.5})	Košetice	1.00	0.76	0.66	0.50	0.41	0.70	0.61
	Frýdlant	1.06	1.11	0.93	0.45	0.44	0.88	0.47
sulfate (AMS PM ₁)	Melpitz	0.66	0.62	0.49	0.71	-	-	-
	Košetice	0.73	1.52	1.38	0.36	-	-	-
	Frýdlant	0.86	0.78	0.86	0.40	-	-	-
nitrate (AMS PM ₁)	Melpitz	2.13	1.58	1.66	0.62	-	-	-
	Košetice	1.95	2.65	2.77	0.16	-	-	-
	Frýdlant	2.03	1.63	2.15	0.46	-	-	-

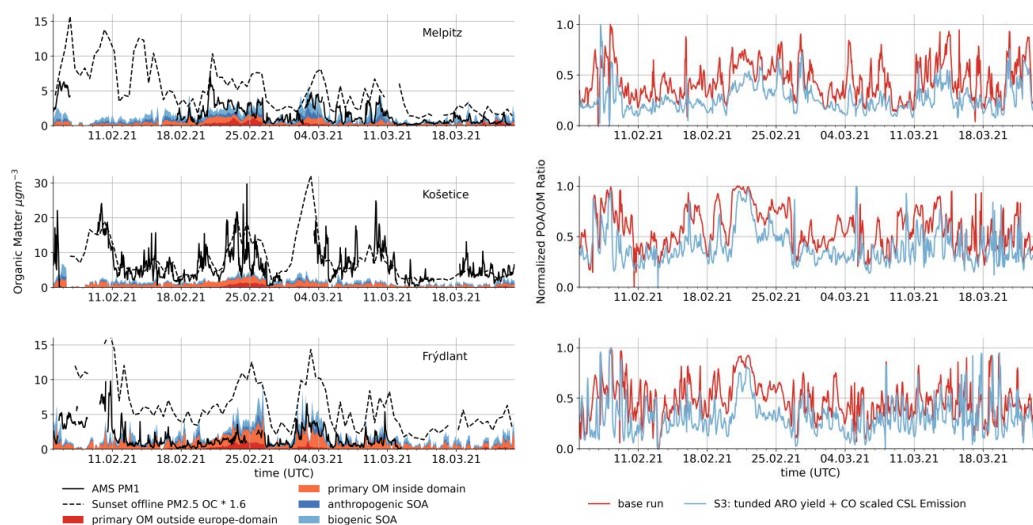


Figure A1. Left: Time series for organic matter mass concentration for the three sites. Comparison of measured AMS data with the modelled primary OM concentration, split into the share with origin outside the N0 domain, origin inside the study domains and secondary organic aerosol. Right: normalized POA to total OM ratio

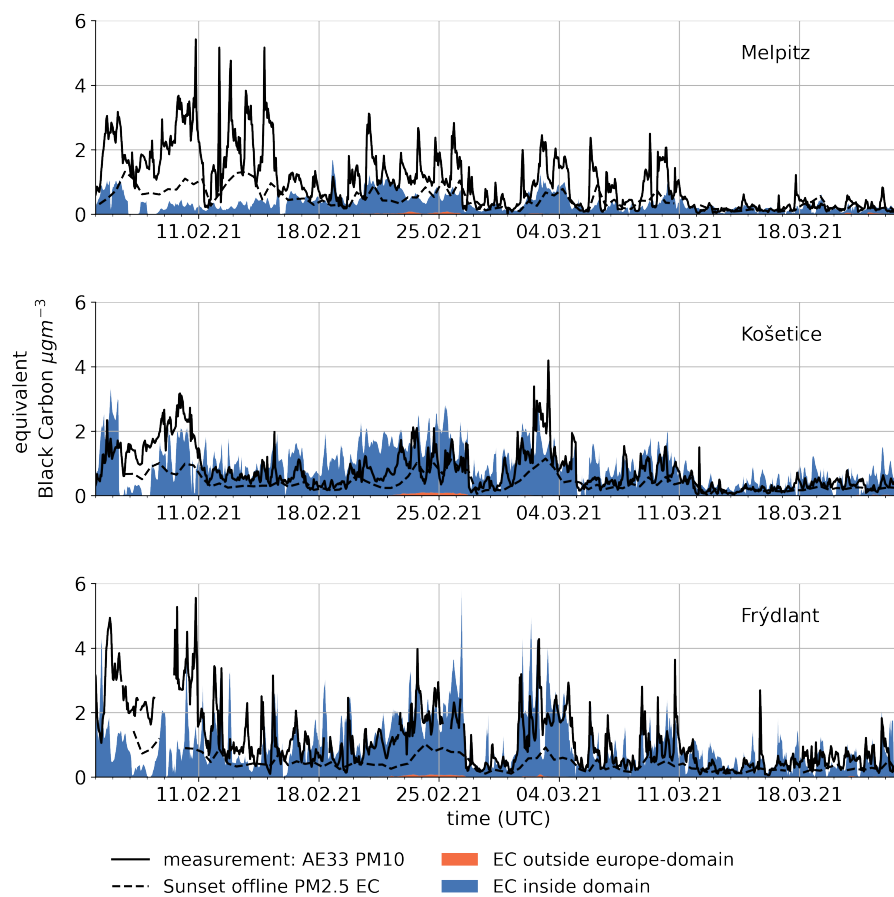


Figure A2. Time series for elemental carbon concentration for the three sites. Comparison of Aethalometer and Sunset Filter data and modeled data.

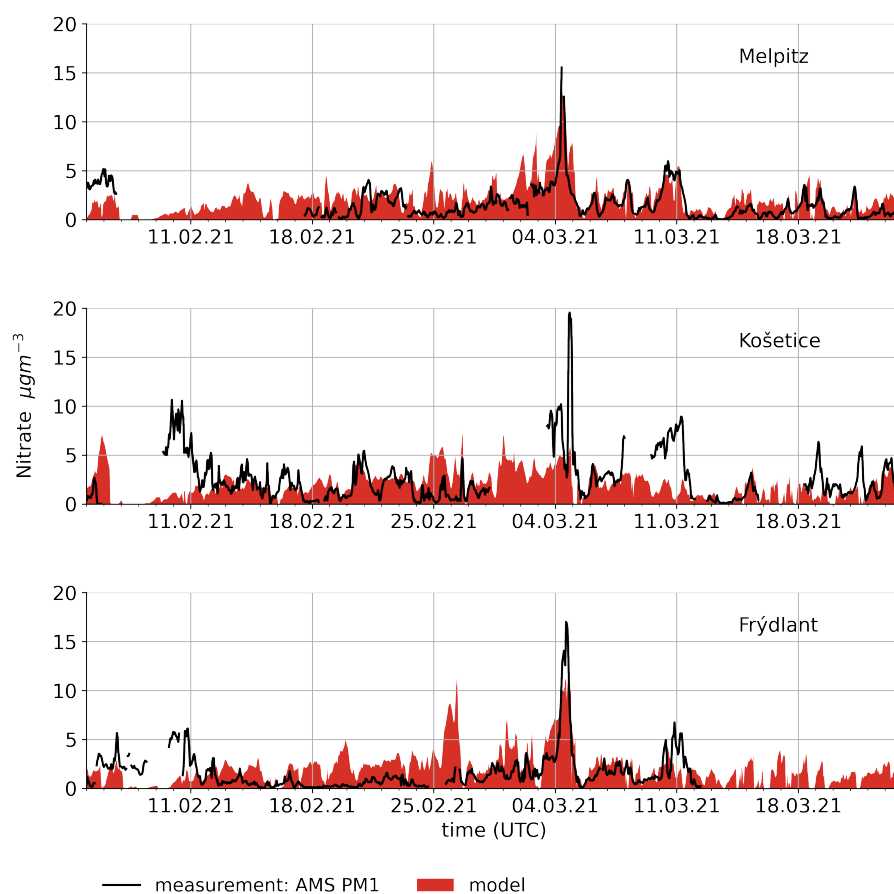


Figure A3. Time series for nitrate mass concentration for the three sites. Comparison of measured and modeled data.

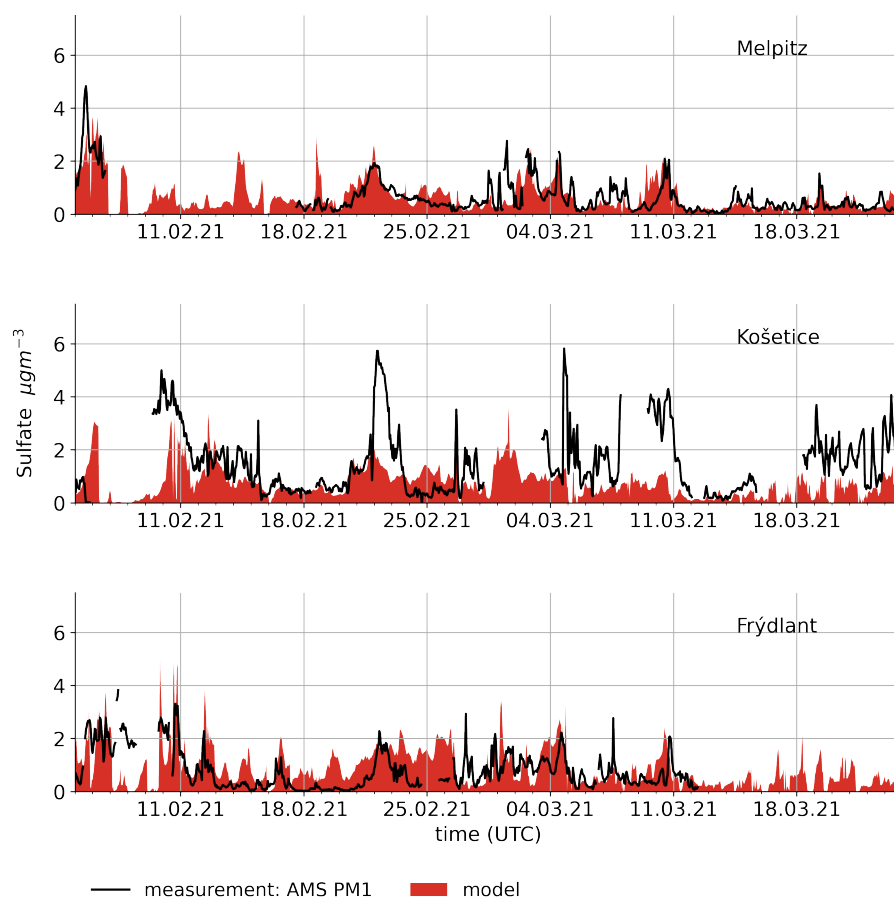


Figure A4. Time series for sulfate mass concentration for the three sites. Comparison of measured and modeled data.

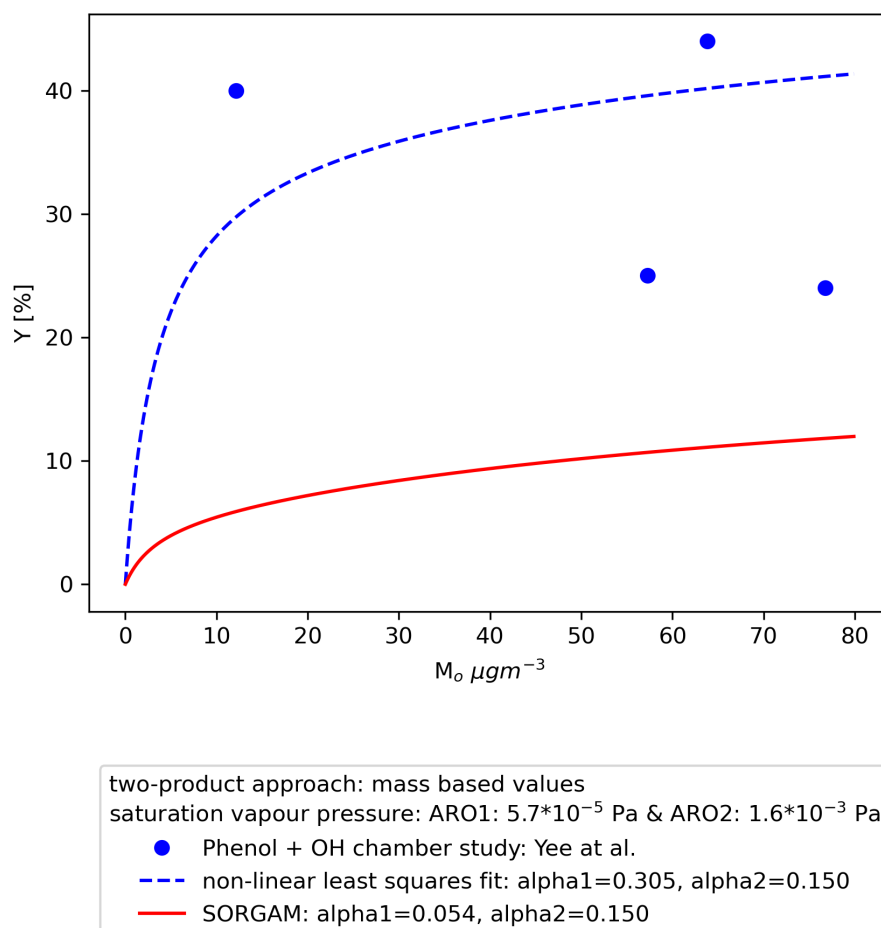


Figure A5. Non-linear least squares fit for two-product aromatic class.

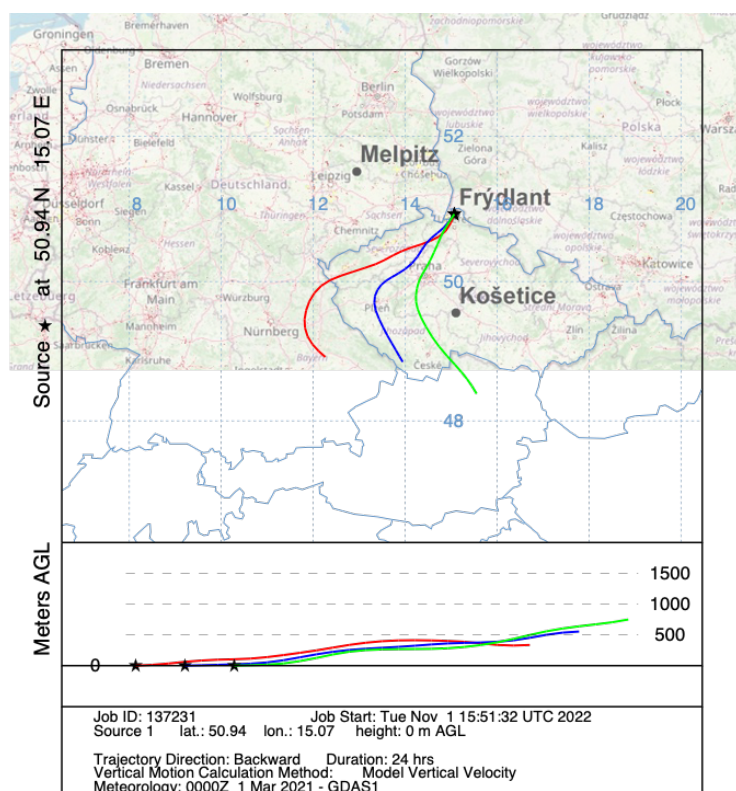


Figure A6. Backward trajectory ending 03 March 2021 at 12, 15 and 18 UTC in Frýdlant, created with NOAA HYSPLIT Trajectory Model (Rolph et al., 2017; Stein et al., 2015)

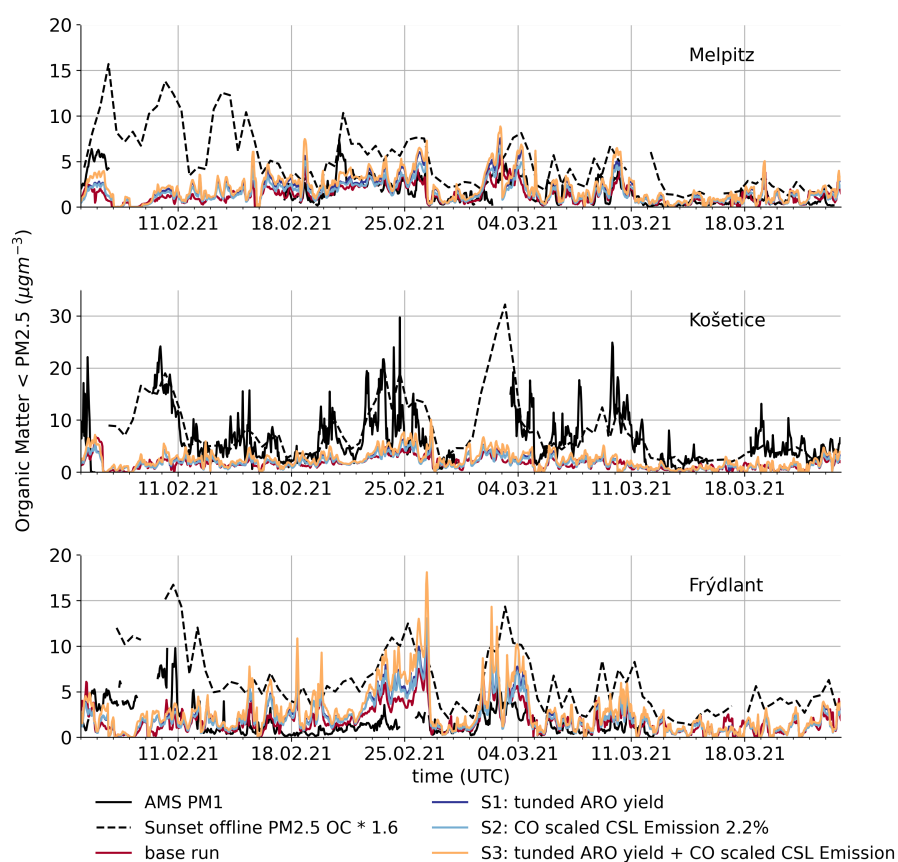


Figure A7. Time series for organic matter mass concentration for all sensitivity runs compared to the base run and measurements.



Table A1. Relative contributions of different source sectors and source regions to the total of ECfine, ECcoarse, OMfine and OMcoarse and absolute mean over all sectors. Contributions from outside the N0 domain are not included.

		ECfine [%]			ECcoarse [%]			OMfine [%]			OMcoarse [%]		
		Melpitz	Košetice	Frýdlant	Melpitz	Košetice	Frýdlant	Melpitz	Košetice	Frýdlant	Melpitz	Košetice	Frýdlant
sector	Public Power	0.6	0.3	0.9	0.9	2.9	5.3	0.5	0.3	0.7	0.1	0.2	0.8
	Industry	2.0	0.5	0.8	14.0	7.5	7.2	2.4	0.5	1.1	7.3	2.1	5.8
	other Combustion	34.6	72.9	76.3	5.5	18.8	30.9	30.9	69.6	72.6	0.0	0.1	0.4
	Traffic: gasoline	0.9	0.1	0.1	0.0	0.0	0.0	1.6	0.2	0.4	0.0	0.0	0.0
	Traffic: diesel	10.1	3.0	4.6	0.5	0.1	0.1	3.6	1.3	2.1	0.0	0.0	0.0
	Fugitives	1.2	0.3	0.5	27.1	17.4	25.5	0.4	0.1	0.2	2.4	1.6	2.8
	Traffic: non-exhaust	0.5	0.1	0.1	7.9	7.3	6.6	1.8	0.4	0.6	8.8	7.8	14.1
	off road	14.4	3.1	2.2	24.9	8.8	5.5	15.4	4.8	3.6	0.0	0.0	0.0
	Livestock	0.0	0.0	0.0	0.0	0.0	0.0	4.1	1.2	1.1	40.2	37.0	34.5
	Agriculture	0.0	0.2	0.1	0.1	5.0	1.1	0.3	0.7	0.2	16.2	18.0	12.4
	other	2.1	0.3	0.6	1.2	0.4	0.5	5.1	1.0	2.0	0.0	0.0	0.0
country	Boundary	33.2	19.1	13.6	17.5	30.7	16.8	33.5	19.8	15.2	20.2	28.4	26.0
	Czech Republic	6.9	76.8	63.0	7.0	49.8	34.8	6.1	75.8	63.3	2.0	55.8	32.6
	Germany	55.5	2.5	7.8	71.9	13.2	24.0	56.6	3.0	9.0	70.2	8.9	22.1
	Poland	4.2	1.4	15.2	3.3	5.1	23.8	3.5	1.0	12.0	3.0	2.1	16.0
absolute mean [$\mu\text{g m}^{-3}$]		0.3496	0.9838	1.0504	0.0775	0.0422	0.0611	0.3432	0.8624	0.8649	0.1094	0.0659	0.0579



Competing interests. The authors declare that they have no conflict of interest.

Author contributions. Conceptualization: HW

Data curation: HW

Formal analysis: HW

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