

# Analysis of source regions and transport pathways of sub-micron aerosol components in Europe

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## 72        **Abstract**

73    It is important to study aerosols and their origins, as they pose various negative health and  
74    environmental impacts. In this study, we combined year-long datasets from 15 different countries  
75    with Trajectory Statistical Methods (TSMs) on such a comprehensive scale for the first time. We  
76    found possible source regions and seasonal variations of various particulate matter (PM)  
77    components, including total organic aerosol (OA), biomass burning OA (BBOA), oxygenated OA  
78    (OOA), ammonium (NH<sub>4</sub>), nitrate (NO<sub>3</sub>), and sulphate (SO<sub>4</sub>) in Europe. We found that for all of  
79    the studied components, eastern Europe was among the highest contributors. For NO<sub>3</sub>, other  
80    important source regions were northern France and the Benelux, while for SO<sub>4</sub>, there were  
81    significant contributions from the Mediterranean region. We also compared our measurement-  
82    based model with simulated concentrations of an atmospheric chemistry transport model (CAMx).  
83    We observed a satisfactory agreement in regions where we had sufficient coverage with air  
84    pollution monitoring stations. The main deviations for OA were found around the Po Valley, where  
85    CAMx consistently estimated higher concentrations, while the TSM analysis did not highlight it  
86    as a hot spot because long-term monitoring data sets in this region are lacking. CAMx also  
87    underestimated the concentrations around Poland, mainly from residential burning. Our results  
88    provide opportunities to refine European emission inventories and deliver valuable information on  
89    long-range transported air pollutants. It suggests that policies mitigating air pollution in Eastern  
90    Europe and the Benelux could help improve overall air quality in entire Europe more efficiently.

# 91 1 Introduction

92 Particulate matter (PM) remains a persistent environmental challenge, with adverse effects on  
93 human health, ecological systems, and global climate (Harrison and Yin, 2000). PM varies in size  
94 and composition and is emitted from a wide range of sources, encompassing both natural and  
95 human emissions. Among the primary sources of PM are transportation, biomass burning, dust  
96 resuspension, industrial processes, and agricultural activities. The impacts of PM on human health  
97 are profound and diverse. Inhalation of PM can lead to respiratory problems such as asthma,  
98 bronchitis, and other respiratory infections (Samoli et al., 2016). Fine particulate matter, known as  
99  $PM_{2.5}$  (particles with aerodynamic diameters of 2.5 micrometres or smaller), can penetrate deep  
100 into the lungs and even enter the bloodstream, increasing the risk of cardiovascular diseases,  
101 strokes, cancers, and other systemic health issues (Mitsakou et al., 2007).

102 Particulate matter consists of both organic and inorganic components. Examples of the latter are  
103 sulphate, nitrate, chloride, ammonium, and black carbon. The sources and formation processes of  
104 these inorganic components are relatively understood, while the other part of PM, organic aerosols  
105 (OA), consists of thousands of compounds; therefore, their formation processes and sources  
106 remain challenging to deconvolve. Previous measurements across the world suggested that OA  
107 can dominate the overall PM mass and consist of 20–90% of the total PM mass (Chen et al., 2022;  
108 Crippa et al., 2014; Jimenez et al., 2009). Recent studies have reported an overview of OA sources  
109 in Europe by employing advanced source apportionment techniques (Calvin et al., 2023; Chen et  
110 al., 2022).

111 While local sources undoubtedly play a significant role in aerosol pollution, it is crucial not to  
112 overlook the substantial impact of transboundary transport. PM can travel vast distances, extending  
113 far beyond its original emission point and affecting regions remote from its sources. To better  
114 grasp the complex interactions between aerosols and long-range transport, researchers have  
115 increasingly turned to trajectory statistical methods (TSMs) (Salvador et al., 2008). These  
116 analytical techniques leverage air mass backward trajectories, which trace the path of air masses  
117 from receptor locations back to their source regions, and PM concentration data collected at  
118 various receptors (Salvador et al., 2010). While TSMs do not account for factors like atmospheric  
119 dispersion, chemical conversion, or dry and wet deposition through the Hybrid Single-Particle  
120 Lagrangian Integrated Trajectory model (HYSPLIT) (NOAA, *accessed 1.11.25*), developed by  
121 NOAA's Air Resources Laboratory. However, they offer simplicity and effectiveness in  
122 application. TSMs can assist in pinpointing relevant source regions and distinguishing the airflow  
123 patterns associated with the transport of regional and large-scale air pollution. By examining the  
124 relationship between air mass trajectories and PM concentrations, TSMs enable the identification  
125 of potential emission hotspots, the characterisation of pollutant transport pathways, and the  
126 assessment of the efficacy of regional air quality management strategies. Utilising TSMs with a  
127 large number of trajectories can notably diminish trajectory uncertainty, which arises from  
128 interpolation and truncation processes, low temporal or spatial resolution of wind data, or  
129 inadequate selection of starting heights (Lupu and Maenhaut, 2002).

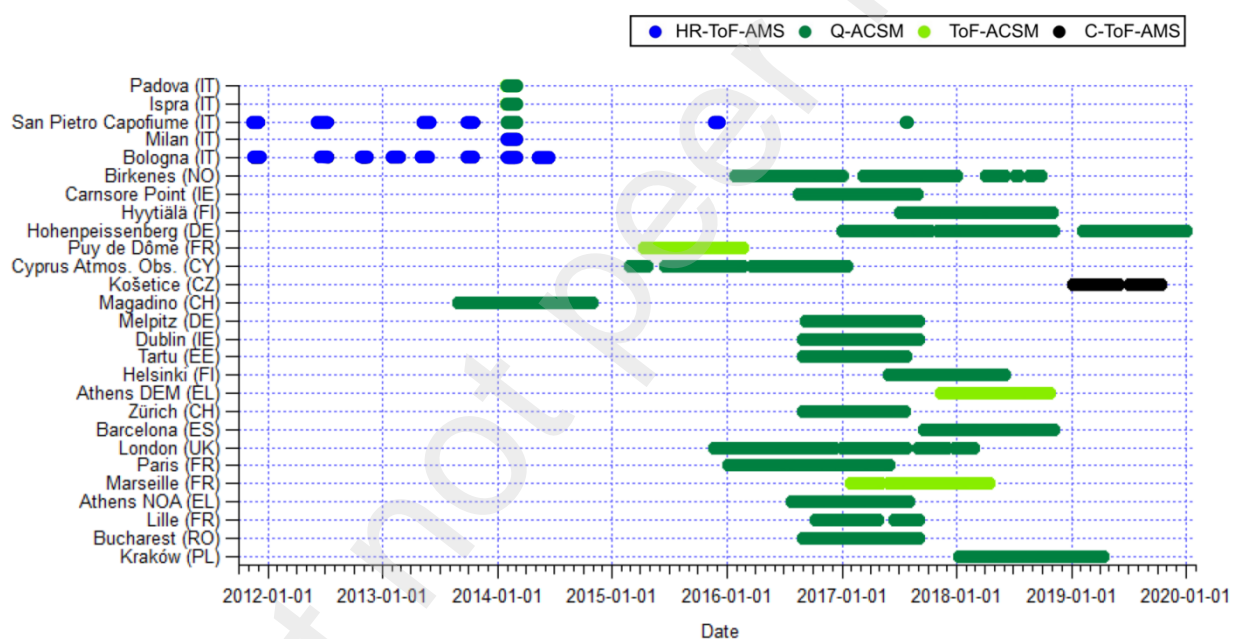
130 Previous studies have highlighted the potential of incorporating multiple receptors in TSMs to  
131 enhance accuracy (Hsu et al., 2003; Lee and Ashbaugh, 2007). In this study, we leverage  
132 comprehensive datasets utilised in harmonised OA source apportionment across Europe to identify

133 potential source hotspots in the region using TSMs. This has not been done previously on this scale  
134 and with this level of high-quality source apportionment data from high-time resolution  
135 measurements (30 min to 4 hours). By integrating all datasets, trajectories converge at various  
136 receptor sites, facilitating a more robust analysis (Stohl, 1996). This convergence, together with  
137 variations in concentrations at each site, amplify the redistribution process, thereby improving  
138 efficacy. The findings from TSM-based investigations are invaluable for policymakers,  
139 environmental agencies, and public health authorities aiming to devise targeted interventions to  
140 mitigate the impacts of transboundary aerosol pollution.

## 141 2 Methodology

142 In this study, we used the chemical composition data and the source apportionment results of 22  
143 air quality observational sites from 15 different countries over Europe, published in (Chen et al.,  
144 2022). In addition, 5 sites in Italy were included, some of which have been published previously  
145 in Paglione et al. (2020). An overview of the countries and the measurement periods can be found  
146 in Figure 1. Measurement devices include the Aerodyne Aerosol Mass Spectrometer (AMS) and  
147 the Aerosol Chemical Speciation Monitor (ACSM), which both deliver real-time mass  
148 concentrations of aerosol species (i.e., organic, nitrate, sulphate, ammonium, and chloride). In  
149 particular, data from 18 Q-ACSM (quadrupole ACSM (Ng et al., 2011)), 3 ToF-ACSM (time-of-  
150 flight ACSM (Fröhlich et al., 2013)), 1 C-ToF-AMS (compact time-of-flight AMS (Drewnick et  
151 al., 2005)), and 2 HR-ToF-AMS (High-Resolution ToF-AMS (Paglione et al., 2020)) were used  
152 in this study.

153 The measurements of the concentrations were averaged every 12 hours (at 12 a.m. and 12 p.m.).  
 154 These averaged values were used in TSMs analysis by combining them with trajectory endpoints  
 155 using an R script. The script can be found in the supplementary material. A statistical analysis  
 156 (Concentration Weighted Trajectory, CWTs) was done using the **openair** R package (Carslaw,  
 157 2019), an open-source tool for analysing air pollution data. The R script for the plotting can be  
 158 found in the supplementary material, and the results are shown in section 3.



159  
 160 **Fig. 1.** The contributing countries and the measurement periods considered in this experiment. 24  
 161 ACSM measurements and 3 AMS measurements were used in this study (Chen et al., 2022;  
 162 Paglione et al., 2020).

163 We chose to focus on specific PM factors based on their universality, i.e., their presence in most  
 164 of the stations, as well as their relative importance, based on the results from Chen et al. (2022).  
 165 We studied two organic PM factors: biomass burning OA (BBOA) and oxygenated OA (OOA),  
 166 as well as total organics, i.e., the sum of all OA factors. BBOA originates mainly from residential  
 167 heating activities, therefore it is more regional and could be more prevalent in rural areas. OOA is

168 an important factor, as it makes up 70% of total OA. Rapid oxidation of primary emissions (e.g.,  
169 BBOA, HOA, COA, biogenic) can be a considerable pathway of OOA formation. Consequently,  
170 biogenic OOA is more prevalent in warmer seasons, while anthropogenic OOA has larger  
171 contributions in colder seasons. In addition, OOA is expected to be transported over long distances.  
172 Other organic components resolved by Chen et al. (2022), such as traffic OA or cooking OA, were  
173 not considered, as they only exist in specific urban environments. Even more site-specific factors,  
174 such as cigarette smoke and ship industry OA, were also not considered for the same reasoning.  
175 Additionally, we also considered three inorganic PM components: ammonium ( $\text{NH}_4$ ), nitrate  
176 ( $\text{NO}_3$ ), and sulphate ( $\text{SO}_4$ ).  $\text{NH}_4$  contributes on average 10% of overall sub-micron aerosol. In  
177 Europe, ammonium emissions are dominated by agriculture, with some contribution from traffic  
178 exhausts (Liu et al., 2024).  $\text{NO}_3$  has a similar overall PM contribution but shows a higher  
179 variability than  $\text{NH}_4$ , as it can make up to 30% of total aerosol.  $\text{NO}_3$  comes from the oxidation of  
180  $\text{NO}_x$ , which in Europe is mostly emitted from traffic and industry.  $\text{SO}_4$  is the highest inorganic  
181 sub-micron aerosol contributor, on average 16%. Sulphate originates from the oxidation of  $\text{SO}_2$ ,  
182 mainly emitted from heavy fuel oil and coal combustion (Bressi et al., 2021).

### 183 *2.1 Back-trajectory analysis*

184 The backward trajectories were modelled using HYSPLIT. It has been widely used in many  
185 previous studies to successfully investigate the impacts of atmospheric transport and dispersion all  
186 over the world (Chen et al., 2021; Escudero et al., 2011; Stein et al., 2015). For the input  
187 meteorological data, we used the GDAS (Global Data Assimilation System) global archive from  
188 2006–present with 1-degree resolution, provided by the National Weather Service's National

189 Centres for Environmental Prediction (NCEP) (GDAS, *accessed 1.11.2025*). We used each of the  
 190 27 measurement stations as end points and modelled one 72-hour back-trajectory every 12 hours  
 191 (at 12 a.m. and 12 p.m. UTC) for the periods where the ACSM/AMS data were available, with  
 192 consideration of the computational capacity. The height of the trajectories arriving at the end points  
 193 was set to 200 metres above ground, and points in the trajectories reaching over 1500 metres above  
 194 ground were filtered out.

## 195 2.2 Concentration weighted trajectory

196 The **openair** package provides two kinds of statistical methods to identify possible source  
 197 locations by using back-trajectories: the Potential Source Contribution Function (PSCF) and the  
 198 Concentration Weighted Trajectory (CWT). Both methods divide the map into grid cells (i,j) and  
 199 can be easily applicable to multiple receptors. The PSCF method takes the number of times a  
 200 source concentration was high when the trajectories passed through the cell (i,j), and divides it by  
 201 the number of times a trajectory passed through the cell (i,j). The CWT, on the other hand,  
 202 calculates the mean concentration species, i.e., their logarithm, for each grid cell as follows:

$$203 \quad \ln(\bar{C}_{ij}) = \frac{1}{\sum_{k=1}^N \tau_{ijk}} \sum_{k=1}^N \ln(c_k) \tau_{ijk}$$

204 where  $i$  and  $j$  are the indices of grid,  $k$  the index of trajectory,  $N$  the total number of trajectories  
 205 used in analysis,  $c_k$  the pollutant concentration measured upon arrival of trajectory  $k$ , and  $\tau_{ijk}$  the  
 206 residence time of trajectory  $k$  in grid cell (i,j). With this CWT method, air packets that stay longer  
 207 in a cell are weighted higher, which is not the case for PSCF. Additionally, the PSCF method

208 requires a weighting function for multi-site analysis, which remains a challenge. Furthermore, as  
209 previously mentioned, PSCF relies on an arbitrary threshold, causing the model to emphasize high-  
210 source contributions. This can result in an imbalanced representation of moderate and relatively  
211 high source contributions. For these reasons, we have chosen to use CWT for the purpose of this  
212 study. A high value of  $\bar{C}_{ij}$  means that air parcels passing over cell  $(i,j)$  would, on average, cause  
213 high concentrations at the receptor site (Carslaw, 2019; Wilks, 2019).

### 214 2.3 Simulations with CAMx

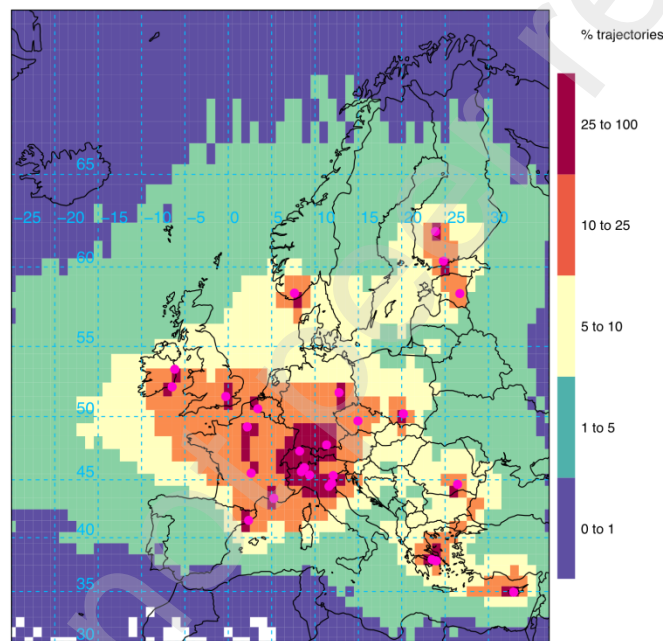
215 An atmospheric chemistry transport model (CAMx) (Yarwood et al., 2024) was deployed to  
216 simulate the distribution and transport of air pollutants for the year 2017. We chose that particular  
217 year because it had the largest overlap of the ACSM/AMS data (see Figure 1), making it more  
218 suitable for comparison with the TSM analysis. In addition, one year of modelling results should  
219 cover most of the relevant scenarios of meteorological conditions. The model domain covers  
220 nearly all Europe (Figure 2), with a horizontal resolution of 0.25 x 0.125 degrees (lon x lat) and  
221 14 vertical layers from the near surface to 7 km ASL. The high-resolution (0.1 x 0.1 degree)  
222 European emission inventory, Copernicus Atmosphere Monitoring Service-Regional Inventory-  
223 Version 4, developed by the Netherlands TNO (Kuenen et al., 2022), is deployed in the simulation.  
224 The gas phase chemistry is simulated using the Carbon Bond 6 Revision 2 mechanism  
225 (Hildebrandt Ruiz and Yarwood, 2013). The ISORROPIA model (Nenes et al., 1998) is used to  
226 simulate the thermodynamic and gas-particle partitioning of inorganic aerosols. Simulation of  
227 secondary organic aerosols is carried out using a 1.5-dimensional volatility basis set module (Koo  
228 et al., 2014). The CAMx model is driven by the hourly meteorology from the regional Weather

229 Research and Forecasting model (WRF, version 3.7), which is driven by and nudged to a 6-hourly  
230 reanalysis of global meteorology from the European Centre for Medium-range Weather Forecasts.  
231 The chemistry initial and boundary conditions for CAMx are provided by the MOZART4 global  
232 model (Horowitz et al., 2003). The CAMx model and the used configuration have been validated  
233 in many previous studies to well reproduce the air pollution over Europe (Daellenbach et al., 2023,  
234 2020; Jiang et al., 2019b, 2019a). More details of model configuration and validation are given in  
235 our previous research (Daellenbach et al., 2020; Jiang et al., 2019b, 2019a).

### 236 3 Results and Discussion

237 The frequencies of the trajectories, without the concentrations, are presented in Figure 2. Figure 3  
238 shows the yearly averaged results for the different species (BBOA, OOA, total organics,  $\text{NH}_4$ ,  $\text{NO}_3$ ,  
239  $\text{SO}_4$ ). For each species, the plot on the left-hand side displays the result of the TSM analysis, i.e.,  
240 the specific source areas. On the right-hand side, respectively, we have plotted the specific local  
241 concentrations as simulated by CAMx (Jiang et al., 2019b). Additionally, emission inventories of  
242 a few selected species can be found in Figure S1. We have decided to focus here on the comparison  
243 with the simulated concentrations in order to obtain a comprehensive picture of both the source  
244 regions and those most affected by air pollution. Note that the scales for the TSM and CAMx  
245 results are different in Figure 3, in order to visualise better some of the patterns. Figure S2 shows  
246 the side-by-side comparisons with the same scaling. Figure 4 contains the seasonal plots for June,  
247 July, August (JJA) and December, January, February (DJF) of BBOA,  $\text{NO}_3$ , and  $\text{SO}_4$ , all of which  
248 we expected strong seasonality.

249 Firstly, looking at the frequency plot (Figure 2), we notice that the highest percentage of  
250 trajectories are very close to the actual measurement stations, which is what we would expect. A  
251 significant percentage of trajectories are over continental Europe, and there are also some over the  
252 Atlantic. Overall, Figure 2 indicates a decent coverage of the air masses over the European domain  
253 of interest.

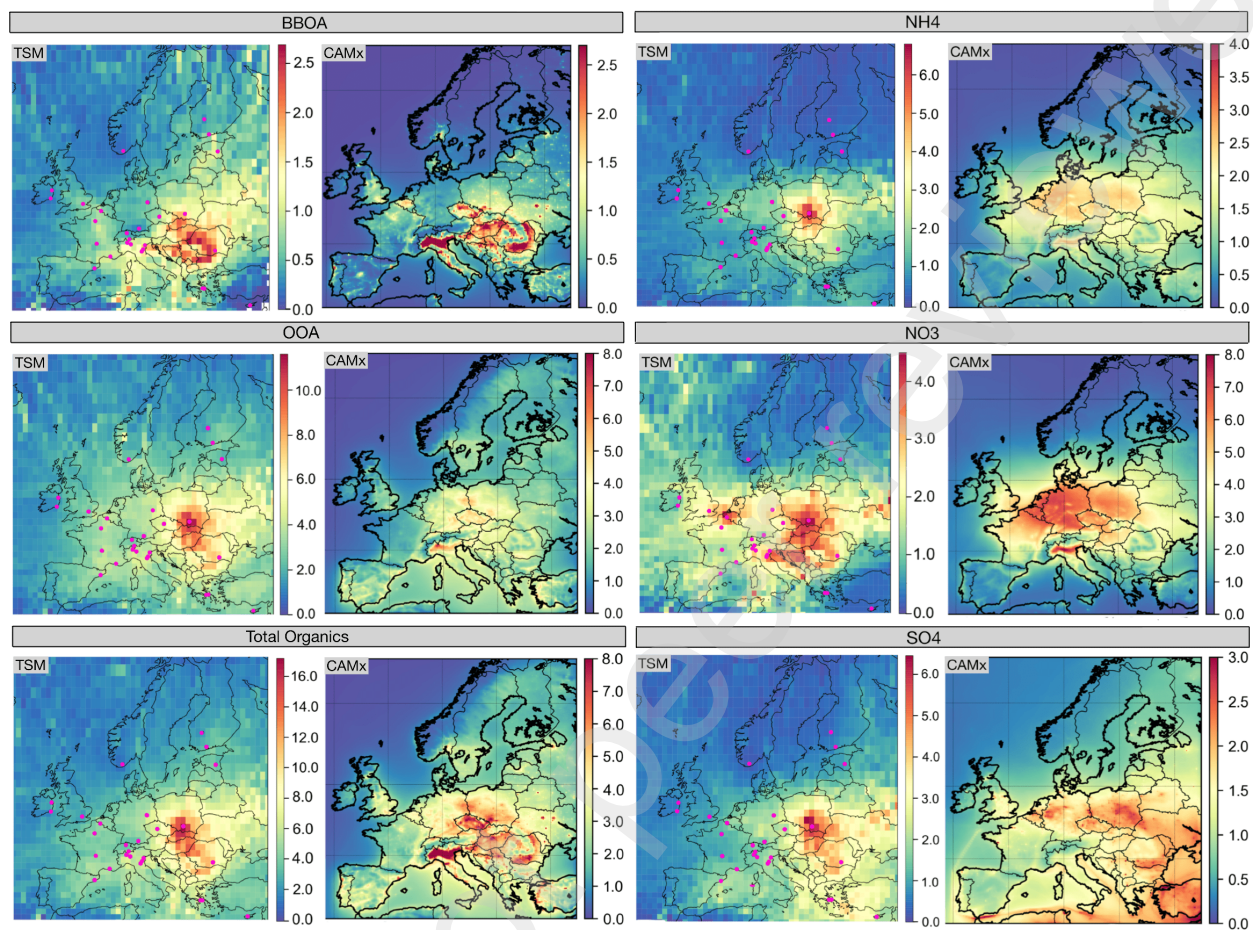


254

255 **Fig. 2.** Frequency plot of the trajectories without the concentrations. The colours indicate the  
256 percentages of the total amount of trajectories passing through each grid cell. The pink dots  
257 indicate the locations of the measurement sites.

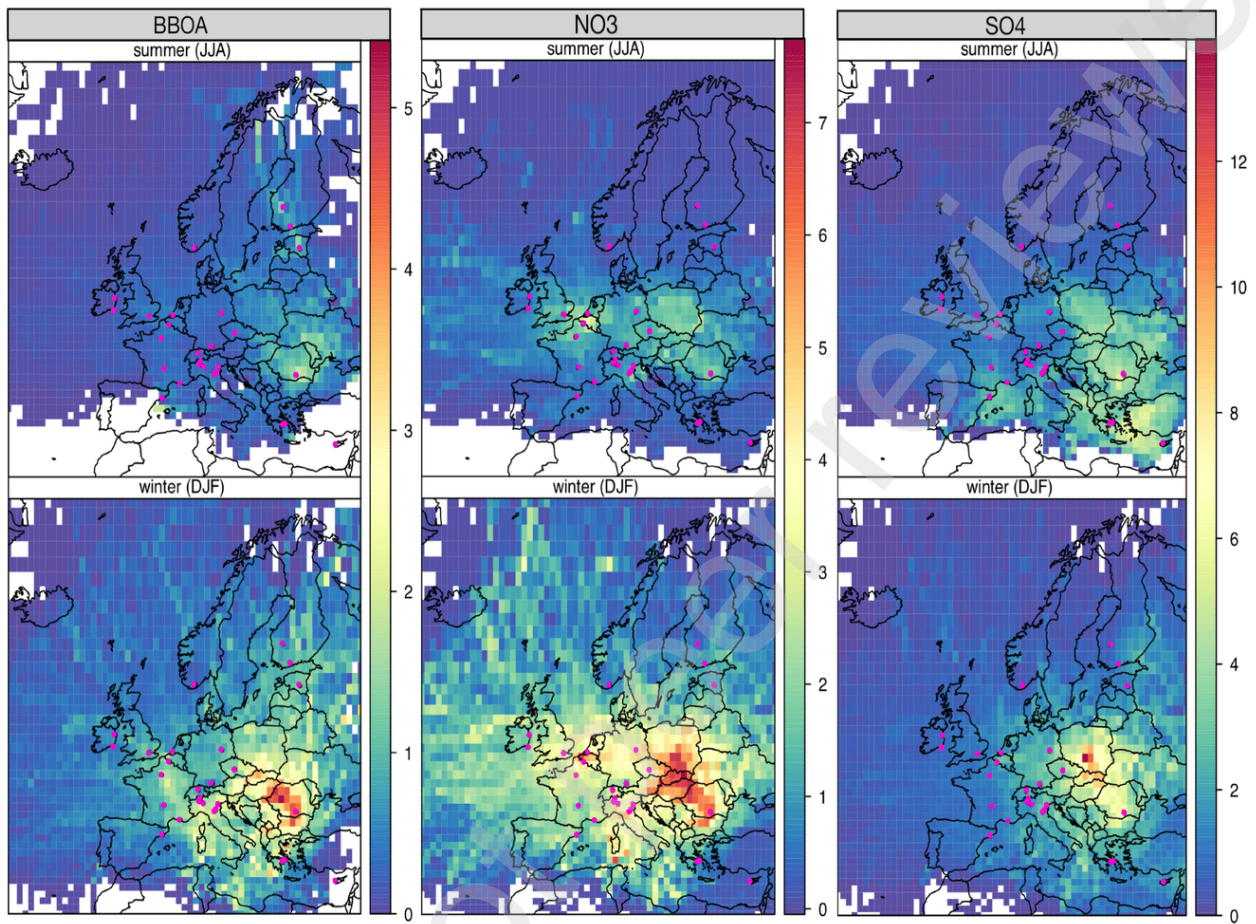
258 It is important to note that in our analysis, all of the available ACSM/AMS data were used with  
259 the CWT method in a single run, not accounting for different campaign durations and times. As  
260 can be seen in Figure 1, some sites have much shorter campaigns and fewer data points in overall  
261 comparison. Even though the CWT method weighs according to concentrations, not the number  
262 of measurement points, this could still potentially lead to an underestimation of shorter campaigns.

263 To study this limitation, we have conducted different analyses, which are included in the  
264 supplementary. Firstly, Figure S3 shows the year-round TSM analysis for only the stations in the  
265 Po Valley, which had shorter campaigns compared to other regions. Figure S4 shows the results  
266 of the TSM analysis for only the month of January with the most overlapping measurements in the  
267 Po Valley, averaged over all years. Figure S5 again shows only the January average, excluding the  
268 stations in Krakow and Bucharest. Comparing Figure S4 with the original DJF plots for BBOA,  
269  $\text{NO}_3$ , and  $\text{SO}_4$ , we see that the overall picture does not change significantly. Even excluding  
270 Krakow and Bucharest from the analysis (Figure S5) only changes the scaling, not the source  
271 regions highlighted. Comparing measurements from different times and of different durations  
272 definitely poses a challenge, and we acknowledge this limitation, which is generally difficult to  
273 quantify, in our analysis. But seeing that our results are sufficiently robust under different  
274 circumstances gives us confidence in our approach and its ability to identify aerosol source regions,  
275 while also allowing for a straightforward integration of datasets in the future. Nonetheless, we  
276 highlight the need for long-term measurement datasets and a wide-spread network of stations.



277

278 **Fig. 3.** Comparison of year-round TSM and CAMx results for BBOA, OOA, total OA, NH<sub>4</sub>, NO<sub>3</sub>,  
279 and SO<sub>4</sub>. Color scale is in  $\mu\text{g}\cdot\text{m}^{-3}$ .



280

281 **Fig. 4.** Seasonal plots showing TSM results for BBOA, NO<sub>3</sub>, and SO<sub>4</sub>, with summer results on top  
 282 and winter results on the bottom. Color scale is in  $\mu\text{g}\cdot\text{m}^{-3}$ .

### 283 3.1 BBOA

284 The results of the TSM analysis show that most of the contributions are from continental Europe.

285 This is what we would expect since BBOA is only emitted over land. Most notably, the highest  
 286 contributions come from eastern Europe, stretching from southern Poland to northern Greece.

287 There is hardly anything from Western Europe.

288 In agreement with the TSM predictions, high BBOA concentrations were predicted in Eastern

289 Europe by CAMx, in particular in Hungary, Romania, and the Balkans. This would suggest that

290 most of the BBOA emitted in those regions stays local as it's highly reactive in the atmosphere  
291 (Hennigan et al., 2010) and oxidises quickly. We can also observe concentration hotspots around  
292 major cities. The most significant difference between CAMx and TSM predictions can be observed  
293 in the Po Valley, where CAMx estimates a high BBOA concentration while not being elevated as  
294 a source region by the TSM analysis. When including the PMF results for Po Valley in the analysis  
295 we find a marginal increase in concentrations in those regions, compared to without (Figure S3).  
296 However, this region is still not at the same level of concentration as Eastern Europe. Potentially,  
297 the emission inventory for BBOA in the CAMx model is overestimated. Or, oxidation processes  
298 lead to the formation of OOA from BBOA, as described in Paglione et al. (2020). As we include  
299 multiple different sites, we could conclude that, relatively, the Po Valley is a less important source  
300 region of BBOA than Eastern Europe.

301 Figure 4 shows high BBOA concentrations in winter, indicating the importance of domestic  
302 heating sources. We can also see a high contribution from Finland in the summer.

### 303 3.2 OOA

304 Our analysis indicates that OOA mainly originates from continental Europe, with the highest  
305 concentrations observed in eastern Europe (from southern Poland to Romania). Although Chen et  
306 al. (2022) reported exceptionally high concentrations measured by the Krakow station, excluding  
307 this station from our analysis did not significantly alter the results, as illustrated in Figure S6.  
308 Therefore, we can confidently conclude that these regions indeed serve as hotspots for OOA.  
309 CAMx also indicates high concentrations in eastern Europe, particularly in the Czech Republic,  
310 indicating a more local effect of OOA sources. This can also be seen in Hungary and southern

311 Romania, which are both elevated by the CAMx and TSM approaches. While the CAMx predicts  
312 high concentrations in the Po Valley, the TSM observed comparatively low contributions from  
313 this region. Once again, this could suggest that the Po Valley does not contribute much to the  
314 overall air pollution burden in Europe. Despite surrounding stations, the TSM did not predict  
315 significant contributions from Turkey, even though CAMx predicts high concentrations. This  
316 could possibly be due to limited trajectories passing through this region, as depicted in Figure 2.

### 317 3.3 Total organics

318 A distinct east-west contrast in OA concentrations is evident in both CAMx and TSM approaches,  
319 which indicates that OA is not transported over the entire continent. OA contributions primarily  
320 originate from continental Europe, notably southern Poland, extending to northern Bulgaria. The  
321 TSM highlights concentrations exceeding  $16 \mu\text{g}\cdot\text{m}^{-3}$  over Poland, compared to simulations  
322 reaching only up to  $4 \mu\text{g}\cdot\text{m}^{-3}$ . This disparity can be attributed to exceptionally high concentrations  
323 around Krakow (Tobler et al., 2021), which might disproportionately affect the TSM analysis.  
324 However, performing the TSM analysis without the Krakow station (Figure S6) still highlights  
325 eastern Europe as the main source region. CAMx also predicts high OA concentrations in that  
326 region, in addition to the Benelux, parts of Germany, the Po Valley, and Turkey. This could either  
327 suggest a tendency for CAMx to underestimate actual OA levels in southern Poland, or indicate  
328 transport processes. Yet, multi-site observations from this region would be needed to confirm this  
329 tendency. CAMx also depicts localised high OA concentrations around major cities, beyond the  
330 resolution of the TSM analysis.

### 331 3.4 $NH_4$

332 Our TSM analysis shows the highest  $NH_4$  contributions from the region in and around Krakow.  
333 Overall, eastern Europe is rather elevated, as are some regions in western Europe, such as the  
334 Benelux and Germany. The highest CAMx concentrations are over Belgium, Germany, Czechia,  
335 Po Valley, and Poland, with some slightly elevated concentrations over the southeast of the UK,  
336 northern France, Slovakia, Balkan regions, Ukraine, the south of Romania, and Turkey. Overall,  
337 both approaches show a consistent spatial distribution except for Po Valley, indicating that the  
338 emission inventory for CAMx is in agreement with the measurements except for Po Valley for the  
339 same reason as mentioned above. However, TSM model in general showed pronounced  
340 contributions of PM components in southern Poland, which leads to higher  $NH_4$  concentrations  
341 than the CAMx model (Figure S2).

### 342 3.5 $NO_3$

343 In the TSM analysis, we observe significant contributions from northern France, the Benelux  
344 region, and eastern Europe, which align with the expected patterns given the influence of high  $NO_x$   
345 and  $NH_3$  emissions from urban areas, particularly those with heavy traffic. Surprisingly, we also  
346 detected relatively elevated contributions from the Atlantic, which prompted further investigation.  
347 Upon conducting our analysis without data from select stations in the UK, Ireland, Paris, and Lille,  
348 we found that this oceanic influence vanished, as illustrated in Figure S7. This suggests that these  
349 unexpected findings are likely artefacts of the TSM process. Additionally, trajectory analysis (see  
350 Figure 2) reveals a notable proportion of trajectories passing over the Atlantic, corroborating our

351 assessment. As expected,  $\text{NO}_3$  exhibits a significant seasonal trend, with higher concentrations in  
352 winter due to enhanced partitioning at lower temperatures (Figure 4).

353 We observe that CAMx estimated concentrations of  $\text{NO}_3$  are considerably higher than the TSM  
354 model across the UK, northern France, Germany, Czechia, and Po Valley with some agreements  
355 in the Benelux region, eastern Europe and southern Europe (Figure S2). The discrepancies of  $\text{NO}_3$   
356 from the CAMx model in the aforementioned regions are potentially caused by scarcities of  
357 measurements for TSM analysis and overestimations of the CAMx model.

### 358 3.6 $\text{SO}_4$

359 In winter, the TSM model predicts the highest concentrations over eastern Europe, particularly in  
360 Poland, likely due to industrial emissions and residential coal burning for heating (Figures 3 and  
361 4). Significant contributions are also observed from the Mediterranean region, attributed to high  
362 ship emissions and enhanced photochemistry during the summer in this region (Figures 3 and 4).  
363 There is a match of hotspots between the TSM and CAMx simulations over Poland, the Balkans,  
364 the Mediterranean, and Turkey. However, the TSM model showed much higher concentrations  
365 than CAMx in eastern Europe due to the potential underestimation of the CAMx model from  
366 industrial and coal combustion sources. In addition, the CAMx model might slightly underestimate  
367 ship exhaust near the Aegean Sea and southwest of Turkey's coastline, resulting in relatively higher  
368 concentrations in these regions for the TSM model. Nevertheless,  $\text{SO}_4$  could still be  
369 underestimated in the Balkans where coal power plants is a quite common source due to both the  
370 scarcity of measurements in this region and limitations of the emission inventories. The TSM

371 results for  $\text{SO}_4$  in the UK show less pronounced contributions compared to the high concentrations  
372 predicted by CAMx, indicating a potential overestimation of  $\text{SO}_2$  emissions in these regions.

## 373 4 Conclusion

374 In this study, we investigated various PM components, both organic (BBOA, OOA, and total OA)  
375 and inorganic ( $\text{NH}_4$ ,  $\text{NO}_3$ ,  $\text{SO}_4$ ), and their long-range transport in Europe. Our aim was to identify  
376 their geographical origins by combining source apportionment results and measurements from 27  
377 air quality observational sites with TSMs. Using numerous trajectories and receptors reduced  
378 uncertainties, providing a comprehensive analysis of source origins. We then compared these  
379 results to estimations of concentrations by the CAMx air quality model in Europe. However, we  
380 acknowledge the caveat of this analysis to combine several receptors with different time periods,  
381 and the fact that back trajectory analysis does not reproduce all atmospheric physicochemical  
382 processes.

383 Starting with the organic components, most contributions come from continental Europe. BBOA  
384 hotspots are around southern Poland to northern Greece. The main source regions as predicted by  
385 our analysis mostly align well with regions of high concentrations as simulated by CAMx, except  
386 for the Po Valley. Seasonally, concentrations are low in summer and high in winter, likely due to  
387 heating. For OOA, the highest contributions are also from eastern Europe, in particular southern  
388 Poland to Romania. The CAMx also highlights this region, but predicts overall lower absolute  
389 concentrations, which could indicate transport processes. Excluding the Poland station did not  
390 change results, suggesting this region is a major contributor, possibly also underestimated in the

391 CAMx model. The TSM analysis also shows discrepancies with the CAMx results in the Po Valley,  
392 potentially indicating an overestimation in the CAMx model, or transport processes. For total  
393 organics, the highest contributions are from southern Poland to Bulgaria, with a clear east-west  
394 contrast well captured by our TSM analysis. Overall, eastern Europe seems to be the most  
395 important contributor.

396 For inorganic components, NH<sub>4</sub> shows high contributions from Poland, and the TSM results  
397 generally match predictions of regions with high concentrations from CAMx except for Po Valley.

398 The NO<sub>3</sub> contribution is highest from northern France, Benelux, and eastern Europe due to NO<sub>x</sub>  
399 emissions from traffic and agricultural emissions in Benelux. The CAMx results show higher  
400 concentrations than the TSM analysis in northern Europe, and Po Valley with good consistences  
401 in eastern Europe in general. TSM analysis showed lower NO<sub>3</sub> concentrations in summer, likely  
402 due to higher temperatures, photochemistry keeping NO<sub>3</sub> in the gas phase, and higher vertical  
403 dilution. SO<sub>4</sub> concentrations are high in Poland, likely from industrial and heating emissions, and  
404 over the Mediterranean, due to photochemistry and ship exhausts. This matches CAMx model  
405 highlights, though we do not find elevated concentrations over Benelux and Germany. Seasonally,  
406 SO<sub>4</sub> is higher in winter over Poland and in summer over the Mediterranean, supporting heating-  
407 related emissions and the photochemical transformation of SO<sub>2</sub> to SO<sub>4</sub>, respectively.

408 In conclusion, we have developed a novel approach to take advantage of available large datasets  
409 to have a comprehensive picture of the geographical origins of different PM components, such as  
410 BBOA, OOA, total OA, NH<sub>4</sub>, NO<sub>3</sub>, and SO<sub>4</sub> across Europe. It demonstrated great consistency with  
411 the concentrations simulated by the CAMx model. With the help of more data available in the  
412 future, the results of this analysis could be used to validate and improve the emission inventories

413 of air quality chemical transport models. By understanding the intricate interplay between local  
414 emissions and long-range transport, stakeholders can implement informed strategies to improve  
415 air quality on a larger scale, protect human health, and safeguard ecosystems in both source and  
416 receptor regions.

## 417 Data availability

418 The code to execute the TSM analysis can be found in the supporting material. Raw data of the  
419 TSM data is publicly available through Zenodo: <https://zenodo.org/records/6672710>. The raw data  
420 of CAMx results are available through a request to the corresponding authors.

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