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Contributions of local and regional sources to fine PM in the megacity of Paris

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Abstract

The Particulate Matter Source Apportionment Technology (PSAT) is used together with PMCAMx, a regional chemical transport model, to estimate how local emissions and pollutant transport affect primary and secondary particulate matter mass concentration levels in Paris. During the summer and the winter periods examined, only 13 % of the $PM_{2.5}$ is predicted to be due to local Paris emissions, with 36 % coming from mid range (50–500 km from the center of the Paris) sources and 51 % from long range transport (more than 500 km from Paris).

The local emissions contribution to predicted elemental carbon (EC) is significant, with almost 60 % of the EC originating from local sources during both summer and winter. Approximately 50 % of the predicted fresh primary organic aerosol (POA) originated from local sources and another 45 % from areas 100–500 km from the receptor region during summer. Regional sources dominated the secondary PM components. During summer more than 70 % of the predicted sulfate originated from SO_2 emitted more than 500 km away from the center of the Paris. Also more than 45 % of secondary organic aerosol (SOA) was due to the oxidation of VOC precursors that were emitted 100–500 km from the center of the Paris. The model predicts more contribution from long range secondary PM sources during winter because the timescale for its production is longer due to the slower photochemical activity.

PSAT results for contributions of local and regional sources were compared with observation-based estimates from field campaigns that took place during the MEGAPOLI project. PSAT predictions are in general consistent (within 20 %) with these estimates for OA and sulfate. The only exception is that PSAT predicts higher local EC contribution during the summer compared to that estimated from observations.

1 Introduction

Fine particles ($PM_{2.5}$) can cause a variety of problems to human health including premature death (Dockery et al., 1993; Kunzli et al., 2000; Klemm et al., 2000; Pope et al., 2002). $PM_{2.5}$ is also responsible for visibility reduction (Seinfeld and Pandis, 2006) and contributes to acid rain formation (Burtraw et al., 2007) and climate change by affecting the energy balance of our planet (Schwartz, 1996).

One of the main challenges when trying to improve air quality in large urban areas is the quantification of the contributions of local as well as long range pollutant transport. Identifying the areas affecting the receptor is also a crucial issue.

A number of approaches have been proposed and used in order to estimate the role of local and regional sources in different areas. Three-dimensional Chemical Transport Models (CTMs) can simulate all relevant atmospheric processes, so they have been used in several different approaches. The simplest method is the so called "zero-out analysis" or "brute-force method" (Park et al., 2003; Knipping et al., 2006; Chin et al., 2007; Koo et al., 2009) which requires running the corresponding CTM multiple times by zeroing out the emissions of a specific source area or type. This is a computationally expensive method because it requires multiple simulations, one for each source area of interest. Another approach is the direct decoupled method (DDM) (Dunker et al., 1981) which can provide local derivatives of the predicted pollutant levels to model parameters (such as emissions or process rate constants). DDM has been used to calculate the sensitivity of the concentration levels to changes in emissions. In general, DDM cannot be applied in large scale perturbations (Dunker et al., 2002). To overcome these problems Ying and Kleeman (2006) developed the source-oriented external mixture (SOEM) approach introducing additional species for each source and tracking them separately throughout the model. In SOEM each secondary PM species is traced back to its precursors. For example the concentration of particulate sulfate is traced back to SO_2 sources. Ying and Kleeman (2009) used SOEM to quantify transport distances in areas of California. SOEM is accurate but computationally demanding.

Wagstrom et al. (2008) developed a computational efficient apportionment algorithm, the Particulate Source Apportionment Technology (PSAT) and used it for the first time in the Eastern United States (Wagstrom and Pandis, 2011a) to estimate the contributions of emissions as a function of distance from the receptor for several urban and rural sites. PSAT is computationally efficient as it works in parallel with the main CTM without interfering with the CTM's calculations. Koo et al. (2009) used PSAT to predict concentrations from different source regions and compared them with the brute force method's results. They showed that DDM and PSAT can give similar results for pollutants that are related linearly with emissions but otherwise the results differ because of the nonlinearity of the chemistry of secondary species or other indirect effects.

In this study we will focus on the Megacity of Paris. Air quality of Paris has been investigated through different models such as CHIMERE (Zhang et al., 2013), PMCAMx (Fountoukis et al., 2013), POLYPHEMUS (Royer et al., 2011), and H^2O (Couvidat el al., 2013). Sciare et al. (2010) compared SOA predictions of the regional CTM, CHIMERE using a single step oxidation SOA scheme (Pun et al., 2006) against observation-based estimates from the "EC-tracer" method (Turpin and Huntzicker, 1995; Strader et al., 1999; Cabada et al., 2004) at an urban site. Fountoukis et al. (2013) applied a fine grid nest over the Megacity of Paris with high resolution (4 km × 4 km). The evaluation of the model against high time resolution (AMS) measurements of fine particulate matter from 3 sites was encouraging. Zhang et al. (2013) implemented the VBS approach into CHIMERE to simulate the behavior of organic aerosol and compared with measurements performed during the MEGAPOLI summer campaign.

Beekmann et al. (2013) used measurements inside the center of the Paris and the surrounding areas to estimate what fraction of the particulate matter is local. The data were collected, during July 2009 and January to mid February 2010 as a part of the MEGAPOLI intensive field campaigns (Beekmann et al., 2013). PM_1 concentration in Paris during summer was on average between 5.3 and 7.5 $\mu g\, m^{-3}$, but during winter was between 15.2 $\mu g\, m^{-3}$ and 18.5 $\mu g\, m^{-3}$. The same authors also estimated, based on the measurements in the appropriate upwind site, that more than 70 % of $PM_{2.5}$

was transported into Paris from areas far away from it. During more polluted periods ($PM_{2.5} > 40\,\mu g\,m^{-3}$ at the urban site), the regional contribution was even larger (on average 80 %). 70 % of the OA was found to be transported from regional sources during both summer and winter. Freutel et al. (2013) found that the concentrations of secondary species such as oxygenated OA, nitrate and ammonium during the summer MEGAPOLI campaign were quite similar for the three measurement sites in Paris, indicating the dominance of regional transport. Significant levels of secondary species were transported to Paris when the corresponding air masses originated from continental Europe. The importance of the regional sources in Paris was also confirmed by Crippa et al. (2013) for the winter MEGAPOLI period.

The MEGAPOLI measurements provide a unique opportunity to compare the PSAT predictions with the estimated regional contributions. In addition, PSAT can quantify the average transport distance from regional sources to the Paris area, thus providing additional insights. The original version of PSAT has been based on PMCAMx-2002, a CTM that assumes that primary OA is non-volatile and inert and simulated SOA formation using the Odum et al. (1996) 2-product approach. Recent developments including the semivolatile POA and the continuous aging of OA (Robinson et al., 2007) have not been considered in previous source-receptor analyses by PSAT or other algorithms.

In this work we first extend PSAT so that it can work with the VBS approach (Donahue et al., 2006) for OA. We then apply the extended PSAT in PMCAMx-2008 in the European domain focusing on Paris for the first time. This paper describes the PSAT application to the megacity of Paris for two different seasons corresponding to the MEGAPOLI intensive campaigns.

2 Model description

2.1 PMCAMx-2008

The model used in this study is PMCAMx-2008 (Fountoukis et al., 2011), a regional three-dimensional CTM. PMCAMx-2008 is applied to Europe, covering a region of 5400 km × 5832 km, using 36 km × 36 km grid resolution and 10 vertical layers extending up to 6 km. The model is applied for two periods, 1–30 July 2009 and 10 January–9 February 2010. The gas-phase chemical mechanism used is based on the SAPRC99 mechanism, including 211 reactions of 56 gases and 18 radicals. The PMCAMx SAPRC99 version uses five lumped alkanes, two olefins, two aromatics, isoprene, a lumped monoterpene species, and a lumped sesquiterpene species. OA is treated in this work as in Murphy and Pandis (2010). Fountoukis et al. (2011) evaluated the PMCAMx-2008 performance for the same European domain against ground measurements taken at four measurement stations and airborne measurements during May 2008. The model reproduced more than 87 % of the hourly measurements of PM_1 OA and more than 94 % of the daily averaged OA concentrations within a factor of 2. Fountoukis et al. (2013) investigated the role of grid nesting for air quality simulations in Paris, by comparing the results of PMCAMx-2008 with 4 km × 4 km grid resolution to those of lower resolution (of 36 km × 36 km) for the summer and winter period of the MEGAPOLI campaign. Fountoukis et al. (2013) also evaluated the model against ground based measurements at the three MEGAPOLI sites of Paris. For the site located in the center of the Paris (LHVP) the average predicted sulfate during summer was within 0.2 $\mu g\,m^{-3}$ of the measured value and the predicted EC was within 0.5 $\mu g\,m^{-3}$. For the suburbs of Paris (SIRTA site) during summer, the model predictions were also in agreement with measurements of sulfate and OA components. EC was predicted to be lower than the measured concentration, by less than 0.2 $\mu g\,m^{-3}$.

2.2 PSAT description

The Particulate Source Apportionment Technology (PSAT) (Wagstrom et al., 2008) is an apportionment algorithm that can track different pollutant categories originating from different area sources or different source types. It is computationally efficient as it works in parallel with the main CTM without increasing the CTM's computational time. PMCAMx-2008 communicates with the apportionment algorithm after each step of each process (e.g., gas phase chemistry, horizontal and vertical advection, etc.) by sending the corresponding concentrations of each species calculated by the CTM's modules to the apportionment algorithm. After each process step of the main CTM, PSAT calculates the apportionment for each source and for each species. PSAT uses the apportionment of the upwind grid cell to apportion species after transport calculations instead of tracking the source-specific species separately. The apportionment of the newly formed secondary PM is calculated based on the apportionment of its precursor species. This means that the apportionment of each secondary PM species is calculated by the same equations as the apportionment of its gas phase precursor.

We have extended the original PSAT algorithm of Wagstrom et al. (2008) to describe the OA mass based on the VBS approach (Donahue et al., 2006; Stanier et al., 2008). Primary OA emissions are distributed into different classes of saturation concentrations (volatility bins). The corresponding gas-phase POA compounds can be oxidized by the hydroxyl radical leading to products of lower volatility. In the same way, SOA is considered to be semivolatile and can also be oxidized further in the gas phase forming lower volatility products. PSAT keeps track of the sources of all OA components (fresh primary, oxidized primary, fresh and aged anthopogenic and biogenic secondary) using the same approach used for all semivolatile species. All OA species are distributed into different volatility bins and are tracked by PSAT as separate species. The apportionment of all semivolatile secondary organic species is treated as a reversible process and the apportionment of the newly formed OA, regardless of its volatility, is based on the apportionment of its gas-phase VOC precursor.

2.3 Definition of source regions within and around the Paris metropolitan area

We have chosen nine computational cells to describe the Paris metropolitan area. We have also defined areas covering distances of approximately 50 km, 500 km, and greater than 500 km away from the center of the Paris metropolitan area (Fig. 1) for the calculation of the corresponding source contributions to PM levels.

PSAT also tracks the contributions to particle concentrations from initial conditions and boundary conditions. Concentrations resulting from emissions within Paris are considered as local. The sum of the concentrations resulting from emissions outside the Paris metropolitan area but within 500 km from the center of Paris is considered as contribution of mid-range transport and those resulting from emissions more than 500 km away from Paris (including the effects of boundary conditions) are considered as long range transport. The long range contribution therefore includes also the emissions outside of the modeling domain.

3 Quantification of source area contributions

3.1 Summer

We will focus first on mass concentration of three representative major PM species: elemental carbon, sulfate, and secondary organic aerosol. Elemental carbon is a non-reactive primary species, sulfate is secondary non-volatile, and SOA is a secondary semivolatile species.

Figure 2 depicts the predicted diurnal pattern of EC, fresh POA, sulfate and SOA and the corresponding source areas. The EC concentration peaks at 08:00 LT in the morning. The concentration of elemental carbon is on average 1 $\mu g\,m^{-3}$. During the hours of the day with the high concentrations the contribution of local transport is higher. The contribution of local sources for EC is on average 64 % while the contributions of mid and long range transport are 30 % and 6 %, respectively (Fig. 3). This behavior is characteristic of primary pollutants with significant urban sources (Wagstrom and Pandis, 2011b).

POA has almost the same behavior as EC with a morning peak at 07:00 LT in the morning, but with lower concentration level than EC (on average less than 0.5 $\mu g\,m^{-3}$).

The behavior of sulfate is quite different because it is a secondary species. Average contributions of local, mid range transport and long range transport to sulfate in Paris are 6 %, 23 % and 71 %, respectively (Fig. 3). This indicates that sulfate in Paris is mainly originating from areas more than 500 km away from the city. This is the result of the time required for its formation and the location of the major SO_2 sources.

Similarly to sulfate, less than 10 % of the SOA in Paris is predicted to be the result of local sources. SOA consists of all the secondary compounds formed from reactions of evaporated POA, intermediate volatility organic compounds (IVOCs), and VOCs. SOA also includes the contribution of the boundary conditions for OA. Paris has modest VOC emissions compared to its surroundings and also several hours are required for the formation of SOA from freshly emitted VOCs. At the same time VOC emissions originating from Paris can be transported to other areas away from the city and can be oxidized there, forming semivolatile oxidation products which can condense to form SOA. Mid and long range sources dominate contributing 45 % and 48 % of the SOA levels in the center of Paris, respectively.

Figure 3 summarizes the average predicted contribution from each source during clean and polluted days. Clean days are defined as the days with average concentrations of total $PM_{2.5}$ mass concentration in the lower quartile of the values predicted during this period. Polluted days are defined as the days with average total $PM_{2.5}$ concentrations in the upper quartile. During more polluted days 55 % of the EC is emitted by local sources with another 37 % coming from mid-range sources. On the other hand, during clean days these contributions of EC are on average 63 % and 32 %, respectively. The contributions of different sources for sulfate and SOA do not change very much during cleaner days compared to those of polluted days. It is noteworthy that PMCAMx/PSAT predicts that the relative contribution of sources in Paris to total $PM_{2.5}$

tends to be lower during polluted than during average days. This rather surprising result is consistent with the observation-based estimates of Beekmann et al. (2013).

3.1.1 Organic aerosol sources

A little more than 50 % of fresh POA in Paris is predicted to be the result of emissions from local sources according to PSAT, while another 45 % is due to mid range transport and less than 5 % comes from areas more than 500 km away (Fig. 4). The POA contributions are similar to those of the EC, but with a lower local contribution due to the evaporation of POA after emission.

Secondary organic aerosol precursors emitted far from Paris are oxidized to form SOA which is then transported to the megacity. Almost 50 % of anthropogenic SOA in Paris is predicted to be due to mid range transport and another 34 % due to long range transport. The behavior of biogenic SOA is quite similar with outside sources contributing on average 83 % (Fig. 4). Total organic aerosol behaves like SOA with local emissions contributing on average 11 %, mid-range transport 43 % and long range 46 % (Fig. 4). Long range transport dominates for total OA because the boundary conditions contribute on average 30 % during summer.

As the volatility of organics increases, the contribution of local sources is predicted to increase (Fig. 5). Organics originating from local sources are in general less oxidized with higher volatilities, but organics which originate from long distances are heavily oxidized and typically of lower volatility. For the least volatile OA components in PMCAMx (effective saturation of 10^{-1} µg m^{-3}), the contribution of local sources is predicted to be on average 10 % and the contribution of mid and long range transport is 62 % and 28 %, respectively. On the other hand for the more volatile OA components (effective saturation of 10^{4} µg m^{-3}), the contribution of local sources is 42 % and the contribution of mid range transport is on average 57 %. These high volatility compounds are mostly in the gas phase.

3.1.2 Effects of Paris on other areas

PSAT has the ability to estimate the impacts of a specific source area, Paris in this application, on other areas and also the impacts of the other specified areas on the receptor region. In order to quantify how far from Paris, the plume of each species can travel, we can use a variety of metrics (Wagstrom et al., 2011b). The Transport Distance (TD) is a useful metric of the average transport distance. TD has been defined by Wagstrom et al. (2011b) as the distance between the center of emissions and the center of the plume of the predicted concentrations at a given time. For the calculation of the TD of secondary species like sulfate and SOA the coordinates of the emission centers are calculated based on the emissions of their precursor gases, SO_2 and VOCs, respectively. Figure 6 shows the temporal variation of the predicted TD (from precursors emitted from the center of the Paris) during the simulated summer period, for EC, sulfate and SOA. The TD is quite variable because it depends strongly on meteorology. EC travels on average 400 km from Paris and this TD is predicted to be lower than that of sulfate (600 km) and SOA (800 km). Figure 7 shows the average impacts of local Paris emissions of EC, SO_2 and VOCs on PM in areas far from Paris. Even if the plumes extend hundreds of kilometers away from Paris, the absolute effects of this megacity are modest (less than 0.1 $\mu g\,m^{-3}$ for each $PM_{2.5}$ component) due to the relatively low emissions of the urban region.

3.2 Winter

The relative source contributions during winter are quite similar to those during summer, despite the differences in absolute concentration levels. Winter is generally characterized by higher emissions of EC and fresh POA and lower emissions of biogenic SOA precursors compared to summer (Table 1). During winter the average predicted concentration of EC in Paris was 2 $\mu g\,m^{-3}$, while during summer was 1 $\mu g\,m^{-3}$. Also the difference in concentrations between the two periods for fresh POA is significant. The predicted POA during winter is 1.5 $\mu g\,m^{-3}$ and during summer 0.2 $\mu g\,m^{-3}$. The predicted diurnal patterns of the concentrations and the corresponding source areas of EC, sulfate, fresh POA and SOA are shown in Fig. 8. The diurnal average profiles of EC and fresh POA are quite similar, with a morning peak and reveal that during the hours with the maximum concentration levels, the contribution of local emissions is higher. During winter the concentrations of secondary species such as sulfate and SOA are lower compared to summer because of the slower photochemistry. The diurnal profile of sulfate is almost flat showing again that local emissions do not play an important role.

EC during winter is characterized by a little lower local Paris contributions and a little higher transport from mid range sources compared to summer (Fig. 9). The contribution of long range transport for sulfate is lower during winter due to lower regional sulfate levels. During winter the contributions of long range transport are lower, because PM components cannot travel long distances as they are removed by rain.

4 Comparison of PSAT results and observation-based estimates

Beekmann et al. (2013) performed a detailed analysis of measurement data collected inside and outside Paris, in order to determine the role of local and regional sources. They concluded that on an annual basis, 70 % of total $PM_{2.5}$ is transported into Paris from areas far away from it. PSAT estimated (as the average of two periods) that 80 % is transported. Both the analysis of measurements and the PSAT predictions suggest that more than 80 % of the sulfate is transported to Paris from other areas (Fig. 10). The two approaches are in excellent agreement during the winter, but PSAT predicts a lower contribution of local emissions during the summer. The discrepancy is of the order of $0.2\,\mu g\,m^{-3}$ and could be the result of underestimating the effect of local sources, as PMCAMx underpredicts the average sulfate in the center of the city by the same amount (Fountoukis et al., 2013) while it is not biased outside Paris. Also the local predicted contributions of OA are lower than those estimated by observations for both summer and winter. This underestimation of local sources (almost $0.2\,\mu g\,m^{-3}$ for summer and winter) could be explained by the behavior of fresh POA. Both the measurement-based studies of Beekmann et al. (2013) and Freutel et al. (2013) found that almost 62 % of the OA in the center of Paris was HOA and cooking OA and 38 % was oxygenated OA. The discrepancy between the measurements and the model can be explained by the cooking emissions which have not been included in the PMCAMx inventory used in this study. Cooking was estimated by Beekmann et al. (2013) to be a significant local source in Paris. On the other hand the local predicted contributions of EC are higher than the measured especially during summer but the absolute discrepancies are of the order of 0.2 $\mu g\,m^{-3}$ for both summer and winter. Generally the predictions of the model are in a good agreement with the results estimated based on measurements and this agreement is encouraging for both approaches.

5 Comparison with other source-receptor analysis methods

We also used PMCAMx-2008 (Fountoukis et al., 2011) to predict the contributions of each selected source area to PM in Paris by zeroing out all the emissions (particles and gases) from different areas around Paris. These areas were selected to be the same with those defined in PSAT simulations. Two additional simulations were performed: one zeroing out the emissions of Paris (local emissions) and one, zeroing emissions originating from areas that are outside of Paris but less than 500 km away from it. An additional simulation taking into account only the boundary conditions was performed in order to calculate the impacts of the boundary conditions on the concentration levels considering them as long range transport. The results of these were combined with those of the base case simulation. Figure 11 compares the results of the two methods (brute force and PSAT) during summer. There is good agreement between the two methods for both the primary and secondary components. This agreement of the contributions of secondary species is unexpectedly good given the nonlinearity of the system. However, PSAT requires 5 times less CPU time than the brute force method for these calculations and is more accurate.

6 Conclusions

In this study we implemented the PM source apportionment algorithm PSAT together with the volatility basis set framework for OA, in the megacity of Paris. This new version of PSAT is used to quantify the contributions from selected source areas to Paris fine PM mass concentrations for two periods, a summer and a winter month. The contribution of local sources (within 50 km from the center of the Paris) was found to be around 60 % for EC during both periods. On the other hand the concentrations of secondary PM components like sulfate and SOA were dominated by long range and mid range transport, respectively. Long range transport was responsible for 71 % of the sulfate during summer and 51 % during winter. For SOA during summer the contributions of mid and long range transport were quite the same (45 % and 48 %, respectively) while during winter long range transport dominates (81 %). During more polluted days the contribution of regional sources was predicted to increase slightly for all PM species.

PSAT-predicted local contributions of EC are higher than those estimated by the observation-based method especially during summer (with local predicted being 64 % and observation based 33 %) either indicating that the local emission sources of EC in Paris have been overestimated or that the observation-based approach is biased low. On the other hand the comparison between the predicted contributions and those estimated based on measurements for sulfate is encouraging with predicted transported contribution being 94 % during summer and 83 % during winter and the measured 81 % and 84 %, respectively. Also OA mass was found to be transported into Paris from regional sources, on average 89 % during summer and 62 % during winter and these results are relatively consistent with the observation-based estimates values (80 % and 55 %, respectively). The dominance of the contribution of regional sources for secondary species like sulfate and SOA for the center of the Paris is the result of the time required to convert precursors to PM species. The contribution of long range transport was found to increase as the particles were further more oxidized. PSAT results were found to be in good agreement with those of different “annihilation” scenarios, but PSAT is more computationally efficient.

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Title Page
Abstract | Introduction
Conclusions | References
Tables | Figures
◀| |▶
◀ ▶
Back | Close
Full Screen / Esc
Printer-friendly Version
Interactive Discussion

Perrussel, O.: Formation of organic aerosol in the Paris region during the MEGAPOLI summer campaign: evaluation of the volatility-basis-set approach within the CHIMERE model, Atmos. Chem. Phys., 13, 5767–5790, doi:10.5194/acp-13-5767-2013, 2013.

Table 1. Emission densities for each source area during the simulated summer and winter periods.

	EC (kg km^{-2})		Fresh POA (kg km^{-2})		Anthropogenic SOA precursors (kg km^{-2})		Biogenic SOA precursors (kg km^{-2})		SO_2 (kg km^{-2})	
	summer	winter	summer	winter	summer	winter	summer	winter	summer	winter
Local	9.7	12.6	5.6	13.4	298.9	366.8	136.9	22.8	11.2	47.2
Mid range	5.7	7.9	6.9	10.2	133.7	123.8	309.3	24.3	26.16	36.1
Long range	1.2	0.3	1.5	0.1	45.9	16.7	136.5	3.9	8.9	8.9

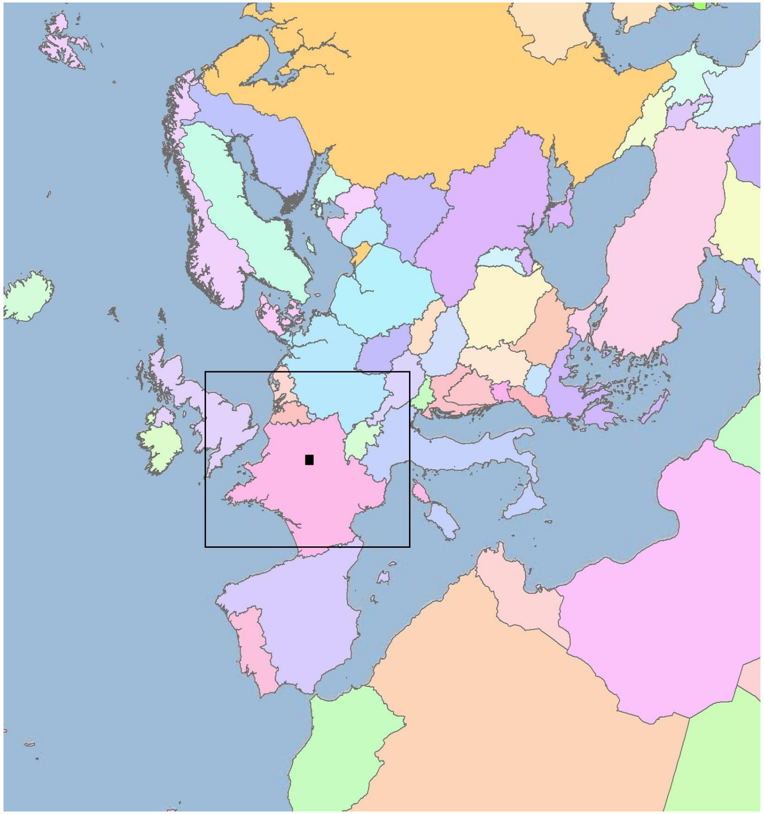

Fig. 1. Definition of different source areas around Paris indicating local Paris emissions (black filed area), 500 km from the center of Paris (area inside the black box not including Paris) and > 500 km (area outside the black box).

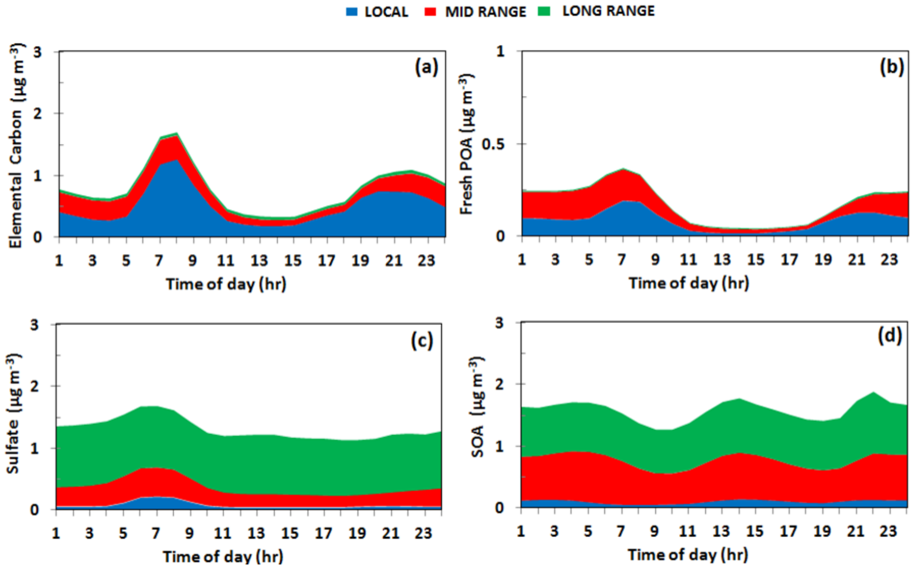

Fig. 2. Average predicted diurnal profiles of $PM_{2.5}$ components and their sources for the summer period for: **(a)** elemental carbon; **(b)** fresh POA; **(c)** sulfate and **(d)** SOA.

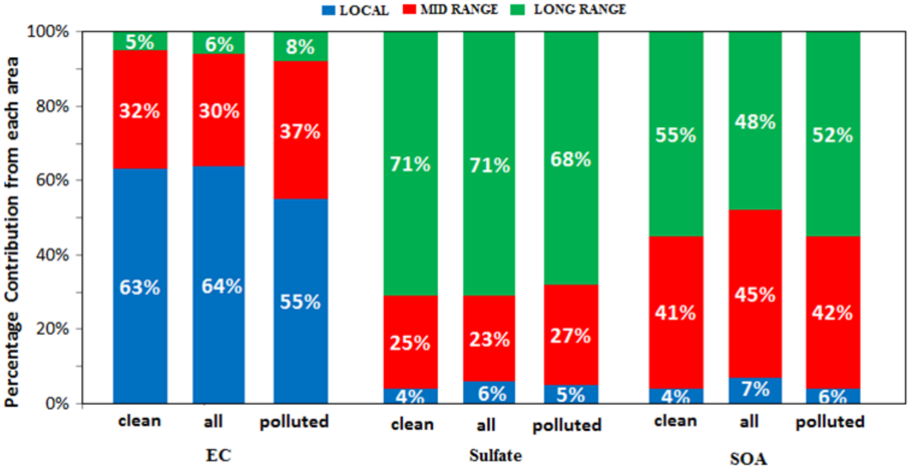

Fig. 3. Percent contributions from each area during high, all, and low particulate matter concentration days for the simulated summer period.

Title Page
Abstract
Introduction
Conclusions
References
Tables
Figures
◀▌
▶▌
◀
▶
Back
Close
Full Screen / Esc
Printer-friendly Version
Interactive Discussion

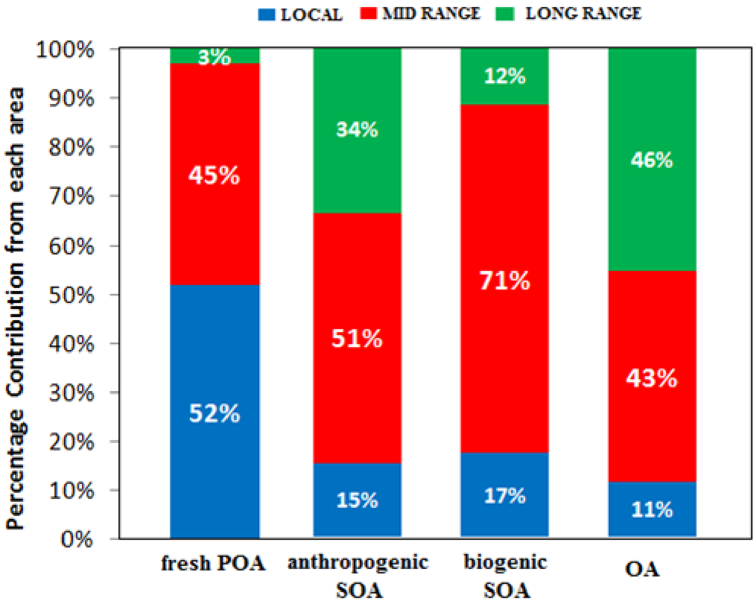

Fig. 4. Percent contributions from each transport category (local, mid-range and long-range) for different types of organic aerosol during the summer. Long range of OA also includes the transported OA into the domain.

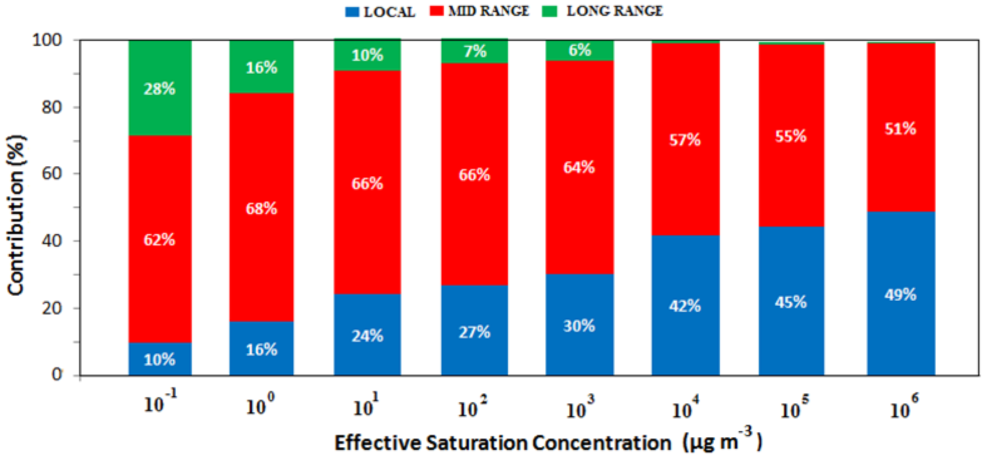

Fig. 5. Predicted source area contributions to organics (organic aerosol plus gas–phase organic mass) in Paris as a function of their volatility. Compounds with effective saturation concentrations above 10 μg m^{-3} are mainly in the gas phase, while the rest are partitioned between the gas and particulate phases.

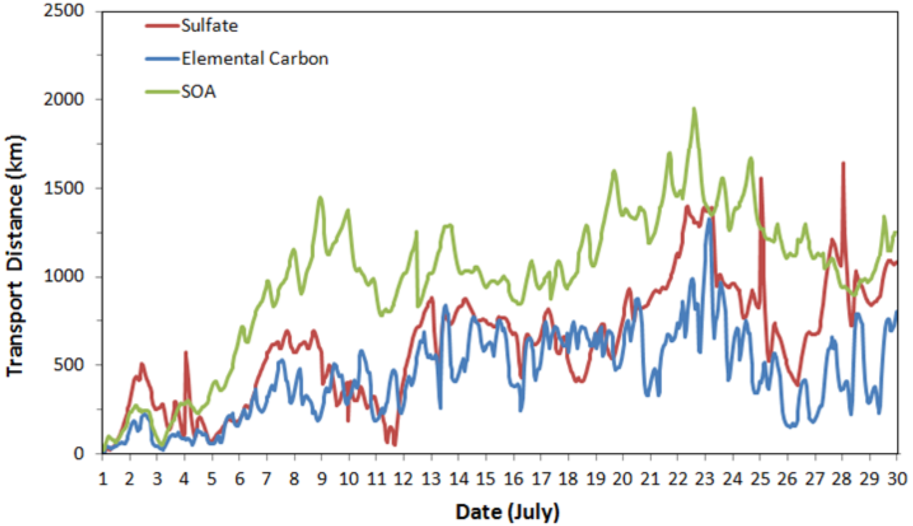

Fig. 6. Temporal variation of the transport distance for EC, sulfate and SOA for Paris emissions for summer period.

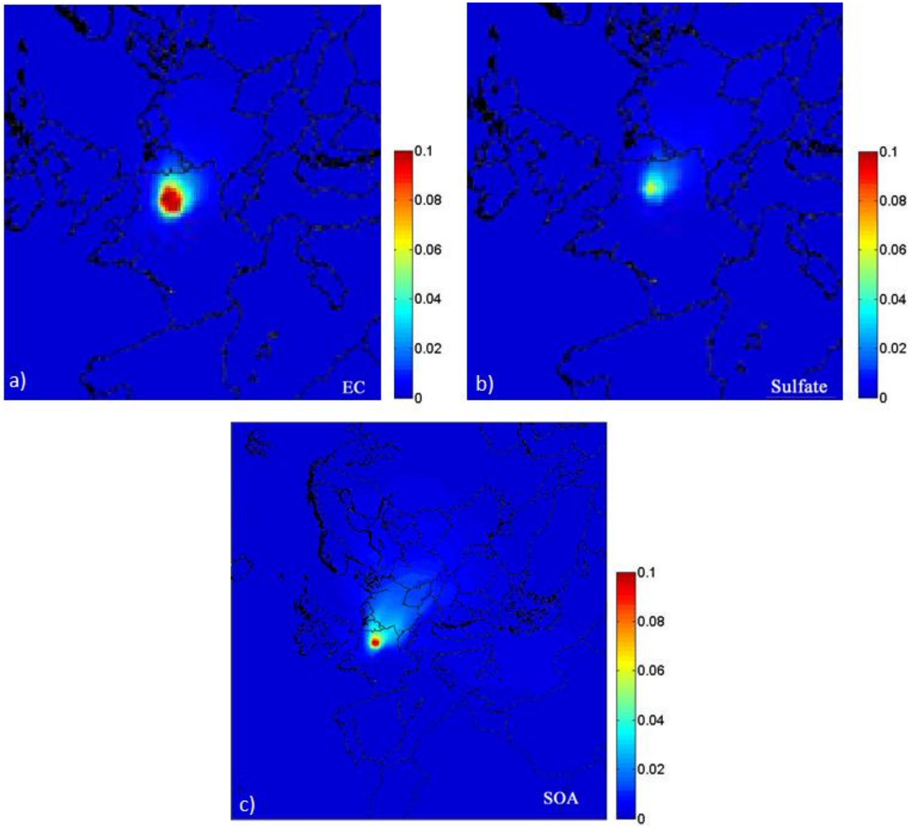

Fig. 7. Predicted average ground concentrations (in µg m^{-3}) of $PM_{2.5}$, EC, sulfate and SOA for local Paris emissions **(a–c)**, respectively.

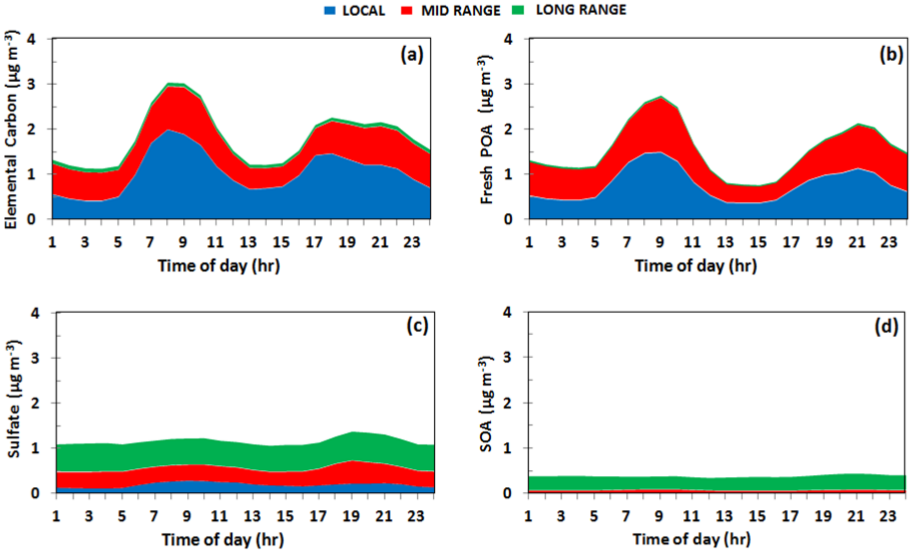

Fig. 8. Average diurnal profiles of $PM_{2.5}$ components and corresponding source areas for the winter period simulated for: **(a)** elemental carbon, **(b)** fresh POA, **(c)** sulfate, **(d)** SOA.

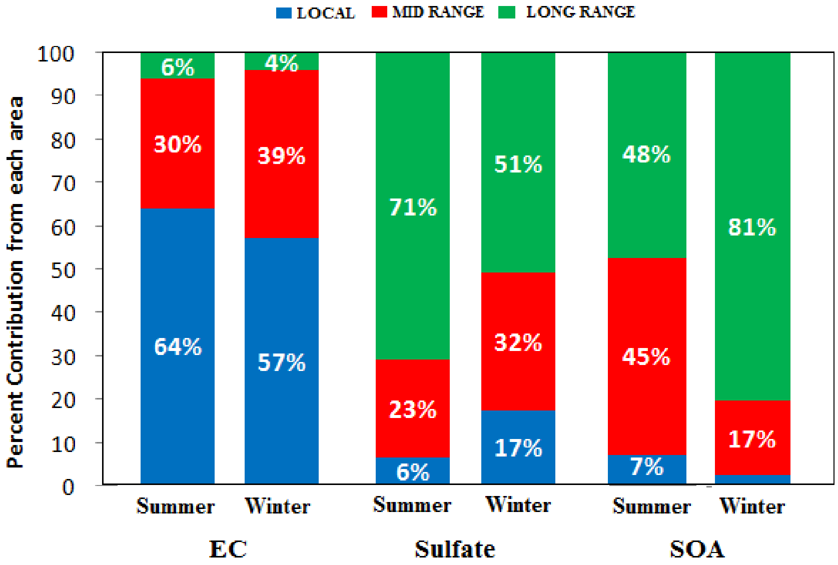

Fig. 9. Comparison of the contributions of each source area for the summer and winter periods.

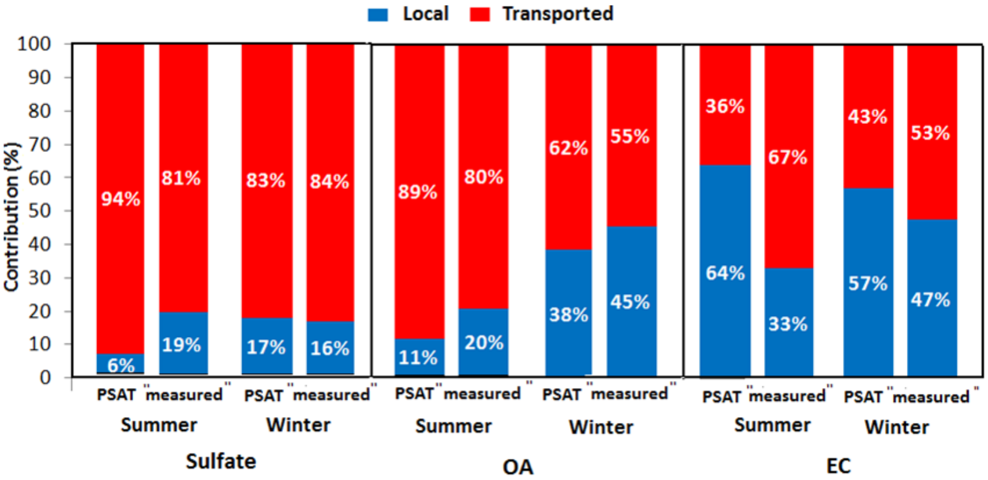

Fig. 10. Comparison of predicted and measured contributions of local and regional sources for EC, sulfate and OA for the summer and winter periods.

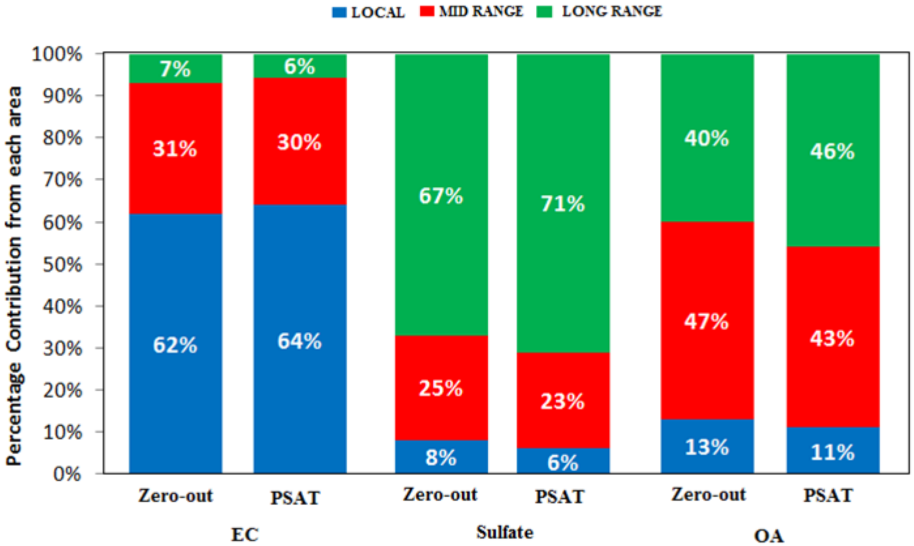

Fig. 11. Predicted contributions of emissions from different source areas to EC, sulfate and OA concentrations during the summer in Paris for the zero-out approach and PSAT.