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**Summertime impacts of eastern mediterranean megacity emissions**

U. Im and M. Kanakidou

# Summertime impacts of Eastern Mediterranean megacity emissions on air quality

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## Abstract

Megacities are large urban agglomerations with intensive anthropogenic emissions that have significant impacts on local and regional air quality. In the present mesoscale modeling study, the impacts of anthropogenic emissions from Istanbul and Athens on local and regional air quality in the Eastern Mediterranean are quantified and the responses to hypothetical decentralization scenarios applied to the extended areas of these densely populated regions are evaluated. This study focuses on summertime impacts on air quality. The results show that Athens emissions have larger regional (0.8 %) and downwind (2.7 % at Finokalia) impacts on O<sub>3</sub> than Istanbul emissions that contribute to surface O<sub>3</sub> by 0.6 % to the domain-mean and 2.1 % to the levels at Finokalia. On the opposite, regarding fine particle (PM<sub>2.5</sub>) levels, Istanbul emissions have larger contribution both inside the megacity itself (75 %) and regionally (2.4 %) compared to Athens emissions, which have a local contribution of 65 % and domain-wide contribution of 0.4 %. Biogenic emissions are found to limit the production of secondary inorganic aerosol species due to their impact on oxidant levels.

Hypothetical decentralization plans for these urban agglomerations, maintaining the total amount of their anthropogenic emissions constant but homogeneously distributing it over larger “new” extended areas, would result in higher O<sub>3</sub> mixing ratios inside the urban core (215 % and 26 % in Istanbul and Athens, respectively). On the opposite, PM<sub>2.5</sub> concentrations would decrease by 67 % and 60 % in Istanbul and Athens, respectively, whereas they would increase by 10 % and 11 % in the rural areas of Istanbul and Athens, respectively. Concerning the “new” extended areas, Athens would experience a reduction in O<sub>3</sub> mixing ratios by ~2 % whereas Istanbul would experience an increase by ~15 %. Overall decreases of PM<sub>2.5</sub> levels by 32 % and 9 % are calculated over the Istanbul and Athens “new” extended areas.

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## 1 Introduction

Over the past decades the increasing urbanization resulted in large urban agglomerations around the world, now hosting more than half of the world's population (UN, 2007). These regions show also high demands of energy, transportation, industry and other facilities in their extended areas. Large urban agglomerations with more than 10 million inhabitants are characterized as megacities and represent concentrated sources of anthropogenic emissions (Lawrence et al., 2007). Their impacts on the surrounding environment and the regional and global air quality have gained significant attention in the last decades. Simulations of the export of air pollution to downwind locations via long-range transport (LRT) have shown different transport patterns depending on the megacity location: in the tropics export is occurring mostly via the free troposphere, whereas at mid and high latitudes it occurs within the lowest troposphere (Lawrence et al., 2007). Butler and Lawrence (2009) simulated small impacts of megacities on the oxidizing capacity of the atmosphere and larger on reactive nitrogen species on global scale. They also pointed out the need of parameterization of the sub-grid effects of megacities. Butler et al. (2008) analyzed different emission inventories and found substantial differences in emission's geographical distribution within countries even if the country total emissions are the same. They also reported large differences in the contribution of various sectors to the total emissions from each city. Thus, the representation of the emissions in the model is an important issue for reliable evaluation of the impacts of megacities.

In addition to these global studies of the impacts of megacities on regional and global atmosphere, regional studies have been implemented for different megacities around the world (e.g. Gaffney et al., 1999; Li et al., 2011; and Shrivastava et al., 2011 for Mexico City; Hodnebrog et al., 2011 for European megacities and Cairo; Lin et al., 2010 for Beijing and Pearl River extended areas). Gaffney et al. (1999) based on measurements of peroxyacetyl nitrate (PAN) as proxy for photochemistry and anthropogenic impacts in Mexico City and on box model calculations, showed that megacities can be important pollution sources for the surrounding areas and the PAN produced in

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megacities can contribute to regional scale ozone ( $O_3$ ) and secondary aerosols during long-range transport (LRT). Tie et al. (2009) used the WRF-CHEM mesoscale chemistry and transport model in the frame of the MIRAGE-Mex project (Megacities Impact on Regional and Global Environments- Mexico City Case Study) and demonstrated that large amounts of CO are transported downwind Mexico City and contribute significantly to the OH reactivity in the city and along the outflow. During the extensive field campaign MILAGRO (Megacity Initiative: Local and Global Research Observations) Molina et al. (2010) attributed significant enhancements of background  $O_3$  to the Mexico City emissions and transformations during transport. They also indicated the key role played by the photochemically formed PAN in the LRT of reactive nitrogen species. They showed that aerosol originating from urban areas can double the concentrations over the rural areas, leading to significant human exposure and health impacts. Lin et al. (2010) identified significant differences between the WRF-CHEM and CMAQ regional models and the global MOZART model in simulating the pollution inflow and outflow over East Asia and in particular the vertical mixing of trace gases that transports pollution to the free-troposphere and from there to distant locations.

In the frame of the CityZen project (Megacity-Zoom for the Environment; <https://wiki.met.no/cityzen/start>), Hodnebrog et al. (2011) investigated the impact of model and adopted emission inventory resolution for the megacities of London, Ruhr area and Cairo on large scale ozone levels, using the WRF-CHEM model. They found that inaccuracies in  $O_3$  computations by high resolution global modeling are relatively small ( $\sim 12\%$ ) compared to more accurate mesoscale modeling and they suggested that the fine-scale impacts are more likely to be resolved with high resolution mesoscale simulations. Royer et al. (2011) by comparing ground-based and lidar observations of  $PM_{10}$  in the Paris area with two different chemistry transport model results in the frame of MEGAPOLI (Megacities: Emissions, Urban, Regional and Global Atmospheric Pollution and Climate Effects, and Integrated Tools for Assessment and Mitigation; <http://megapoli.dmi.dk/>) summer campaign, demonstrated the difficulties to accurately simulate background conditions, urban plume location and dispersion, and

chemistry during transport. Fires together with the local anthropogenic pollution induced by stagnant meteorological conditions have been identified by Konovalov et al. (2011) mesoscale modeling study with the CHIMERE model as the main reason of high CO (up to  $10 \text{ mg m}^{-3}$ ) and  $\text{PM}_{10}$  (up to  $700 \mu\text{g m}^{-3}$ ) concentrations in Moscow megacity during the 2010 Russian wildfires.

Air pollution is an important environmental problem in the Eastern Mediterranean where high  $\text{O}_3$  and particulate matter (PM) levels are observed that often exceed the limits. These elevated  $\text{O}_3$  and PM levels result from transported pollution mixed with anthropogenic and natural emissions in the region under warm and sunny conditions that affect the region most of the year and enhance photochemical production of secondary pollutants such as  $\text{O}_3$  and secondary aerosols (Kanakidou et al., 2011). Eastern Mediterranean is sensitive to climate change and expected to be exposed to higher levels of pollutants in the future (Im et al., 2011a, b). The region hosts important megacities, such as Istanbul, and large urban agglomerations, such as Athens, that significantly impact on regional air quality. Rappengluck et al. (2003), based on PAN observations at two sites on the island of Crete in frame of the PAUR-II campaign, and on air mass back-trajectory analyses, showed that Athens and Istanbul urban plumes affect remote sites in the Eastern Mediterranean. Koçak et al. (2011) based on observations of  $\text{PM}_{10}$  chemical composition from Istanbul and on forward air mass trajectory analysis showed that Istanbul emissions influence western Black Sea and Eastern Europe during winter and Aegean and Levantine Sea during summer. Kanakidou et al. (2011) illustrated the main transport pathways of air masses from Istanbul, Athens and Cairo based on forward air mass trajectory analyses (Kindap et al., 2009) for a 30-year period using reanalysis data from NCEP/NCAR. However, dedicated modeling studies on the evaluation of the impacts of the emissions from the Istanbul and Athens extended areas on the air quality in the Eastern Mediterranean are missing. Recent developments of emission inventories for Athens (Markakis et al., 2010a and b) and Istanbul (Im, 2009) provide a solid base for the regional scale modeling of air quality in the area (Im et al., 2011a, b).

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A limited number of mesoscale model investigations of air quality in the Eastern Mediterranean elaborate the regional impact of specific emission sectors (i.e. shipping; Poupkou et al., 2008) or of total anthropogenic emission reduction scenarios on urban air quality (Im et al., 2010). Among them, CityDelta, a European-wide intercomparison study of model with different spatial resolutions, ranging from 5 to 50 km, covered partly the Eastern Mediterranean (Cuvelier et al., 2007). Recently, Im et al. (2011a, b) mesoscale modeling studies have investigated the response of gaseous and particulate air pollutants to increases in air temperature during summertime, susceptible to occur during climate warming. These air quality studies had both regional focus on the Eastern Mediterranean and local focus on Istanbul and Athens extended areas as well as on the remote costal location of Finokalia on Crete island in the Eastern Mediterranean, acting as receptor site of air pollution.

In the present study that builds upon these earlier works, the impacts on air quality of the anthropogenic emissions from Istanbul megacity and from Athens extended area are separately quantified by additional mesoscale simulations. The study aims (1) to quantify the contribution of these large urban centers of anthropogenic emissions to the air quality in the urban center itself and in the surrounding locations in the Eastern Mediterranean as well as; (2) to evaluate the impact of a “hypothetical decentralization plan” for these intensively populated areas to the air quality in nearby locations.

## 2 Materials and methods

The air quality modeling system used for this study consists of the Weather Research and Forecasting model (WRF-ARW v3.1.1; Sharmock and Klemp, 2008) coupled with the US EPA Community Multiscale Air Quality (CMAQ) model, version 4.7 (Byun and Schere, 2006). The model of Emissions of Gases and Aerosols from Nature (MEGAN) is used for the calculation of biogenic emissions and the global Transport Model v.4 (TM4-ECPL; Myriokefalitakis et al., 2010 and references therein) provides the chemical initial and boundary conditions. The model system and the adopted emissions are

described in detail by Im et al. (2011a, b) and references therein. The domain over the Eastern Mediterranean (from 31.89° N, 17.34° E to 43.69° N, 36.46° E), the model horizontal (30 × 30 km) and vertical (30 layers) resolutions, the physical and chemical options, the initial and boundary conditions and the performed scenarios used in the models are described in detail by Im et al. (2011a) and references therein. The model performances to simulate air quality in the region for the studied period have been evaluated by comparison with available observations by Im et al. (2011a, b). Hereafter, information on adopted anthropogenic emissions for the simulations period, useful for the discussion of the results of the present study is outlined.

## 2.1 Anthropogenic emissions

The anthropogenic emissions for the model domain are compiled by merging emissions from Greece on 10 km and from Istanbul and Athens on 2 km resolution into the emission inventory of the French National Institute of Industrial Environment and Risks (INERIS: <https://wiki.met.no/cityzen/page2/emissions>) for Europe as detailed in Im et al. (2011a). The anthropogenic emissions from Istanbul and Athens, integrated over the 15-day simulation period, are presented in Table 1. The PM<sub>2.5</sub> anthropogenic emission speciations for Athens and Istanbul (Fig. 1) for the simulation period show significant fractions of unspecified anthropogenic PM<sub>2.5</sub> emissions (~40 %) in both cities. Elemental carbon (EC) presents a significant mass fraction of PM<sub>2.5</sub> anthropogenic emissions in Athens (25 %) whereas Istanbul emissions are characterized by organic carbon (OC) emissions (32 %).

During the simulation period, road transport is the main contributor to CO emissions both in Istanbul (93 %) and in Athens (67 %). Other sectors contributing to CO in Istanbul are residential combustion (2 %), industrial processes (3 %) and waste management (1 %). In Istanbul, road transport sector has also the largest contribution to nitrogen oxides (NO<sub>x</sub>) (83 %) and non-methane volatile organic compounds (NMVOC) (45 %) emissions (Im, 2009). Sulfur dioxide (SO<sub>2</sub>) is largely emitted by energy production (38 %), industrial combustion (25 %) and cargo shipping (25 %). PM<sub>10</sub> emissions

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are due to industrial combustion (48%), road transport (20%) and industrial processes (17%). In Athens, 51% of  $\text{NO}_x$  and 38% of NMVOC emissions originate from the road transport and 50% of NMVOC emissions are generated by non-road traffic (Markakis et al., 2010a, b). In addition,  $\text{SO}_2$  is emitted by industrial combustion (44%) and shipping (37%). Finally,  $\text{PM}_{10}$  is emitted from on-road traffic (31%), from off-road traffic (24%), from maritime (18%) and from industrial combustion (16%).

Table 1 also reports the biogenic NMVOC and soil-NO emissions over the extended areas of Istanbul and Athens, integrated for the simulation period. The calculated biogenic NMVOC emissions in Athens are 2.4 times higher than those in Istanbul. However, the anthropogenic emissions are 2.7 and 7.3 times larger than the biogenic NMVOC emissions in Athens and Istanbul, respectively, therefore, only small changes in the NMVOC/ $\text{NO}_x$  molar emission ratios are calculated when biogenic emissions are taken into account (Table 1).

## 2.2 Scenarios

The impact of anthropogenic emissions from Istanbul and Athens on the air quality of the region during summer has been investigated by a set of eight simulations. Brute force analysis has been applied in order to zero-out the anthropogenic emissions in Istanbul and Athens, individually and together, by perturbing the emission files. The impact of a “hypothetical decentralization plan” for these intensively populated areas to nearby locations has been evaluated with additional simulations. All simulations have been conducted for a 26-days period from 20 June to 15 July 2004. The first 11 days (20–30 June) have been considered as spin-up period and not evaluated in the analyses. The studied scenarios are as following:

- a. Base case scenario (S0): The base case simulation uses anthropogenic and natural emissions as described in detail by Im et al. (2011a, b) and in Sect. 2.1. The evaluation of the simulation by comparison with observations has been presented in Im et al. (2011a, b).

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- b. Scenario S1: As S0 but masking all biogenic emissions in the model domain (NoBiog).
- c. Scenario S2: As S0 but masking all anthropogenic emissions from the extended Istanbul area (NoIst).
- 5 d. Scenario S3: As S0 but masking all anthropogenic emissions from the extended Athens area (NoAth).
- e. Scenario S4: As S0 but masking all anthropogenic emissions from both the extended areas of Istanbul and Athens (NoIstAth).
- f. Scenario S5: As S0 but masking all anthropogenic emissions in the model domain (NoAnth).
- 10 g. Scenario S6: This scenario investigates the impact of a “hypothetical decentralization plan”. For this, it is assumed that the extended areas of the two cities are further extended over the surrounding regions. These “new” extended areas of the cities have been arbitrary defined (Fig. 2). The land surface area in the original extended areas of Istanbul and Athens, as predefined in Markakis et al. (2010a, b) and Im (2009) are calculated to be  $\sim 3000$  and  $7500 \text{ km}^2$ , respectively. The “new” extended areas of Istanbul and Athens on the other hand, are increased by a factor of 4.9 ( $\sim 15\,000 \text{ km}^2$ ) and by a factor of 2.6 ( $\sim 20\,000 \text{ km}^2$ ), respectively. A larger increase in Istanbul extended area is assumed due to the significantly larger population in Istanbul ( $\sim 13$  million) than in Athens ( $\sim 4$  million). The anthropogenic land emissions from the extended areas of Istanbul and Athens together with those from their surroundings regions are homogeneously re-distributed to the areas covering the “new” extended areas of the cities (H-Anth). Figure 2 depicts the  $\text{NO}_x$  emissions, integrated over the 15-day simulation period before (Fig. 2a) and after the decentralization is applied (Fig. 2b). The biogenic emissions for S6 are kept unchanged from S0 simulation.
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h. Scenario S7: As S6 but modifying the biogenic emissions (H-AnthBiog) in the “new” extended areas by setting them to those from a grid cell of each city that could be characterized as suburban.

### 3 Results

#### 3.1 Air quality levels

The surface mixing ratios of  $O_3$ , PAN and  $PM_{2.5}$  calculated for the base case simulation (S0) and averaged over the 15-day simulation period, are shown in Fig. 3a–c. Table 2 further provides the surface concentrations of the major gases and aerosols as well as the molar NMVOC/ $NO_x$  and CO/ $NO_x$  over the urban, the rural locations and the entire extended areas of Istanbul (IST) and Athens (ATH), and at the remote site of Finokalia (FKL), on Crete, Greece, averaged over the 15-day simulation period.

The lowest  $O_3$  mixing ratios are calculated for the urban sites of Istanbul (~19 ppb) and Athens (~52 ppb) due to significant  $NO_x$  emissions depleting  $O_3$  locally (Table 2). The large gradient between the urban and rural sites of Istanbul (~19 to 65 ppb) points out the importance of emissions hot spots in an extended area. The higher  $O_3$  levels in Athens than in Istanbul are due to the elevated background of  $O_3$  in the Athens extended area resulting from regional influences and long range transport. The increased background levels in Athens and over the Mediterranean Sea are also supported by the CO/ $NO_x$  molar ratios (Table 2). This ratio increase from Istanbul to Athens and then to Finokalia showing the significant impact of local emissions in Istanbul compared to Athens and to Finokalia (Im et al., 2011a; Kanakidou et al., 2011). The highest  $O_3$  mixing ratios are calculated over the Mediterranean Sea and particularly west and south of Crete (Fig. 3a), where they are supported by  $NO_x$  emissions from shipping. The impact of shipping routes is also seen in the spatial distribution of PAN (Fig. 3b) as discussed by Im et al. (2011a). Surface  $O_3$  at the Finokalia station is calculated to reach ~70 ppb. Another striking point is the difference in the distribution of the NMVOC/ $NO_x$  molar

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ratios in the two extended areas. While the NMVOC/NO<sub>x</sub> ratios increase from urban to rural sites in Istanbul (~2.5 to ~13), Athens experiences the opposite spatial pattern change (~192 to ~77), particularly due to lower NO<sub>x</sub> and similar NMVOC emissions compared to Istanbul (Table 1).

5 The highest aerosol concentrations are calculated over the emission hot spot areas. Istanbul and Athens areas also stand out with high PM<sub>2.5</sub> concentrations (27 and 23 μg m<sup>-3</sup>, respectively) compared to their surroundings (Im et al., 2011b; Kanakidou et al., 2011) as depicted in Fig. 3c. In Istanbul and Athens, PM<sub>2.5</sub> decrease by a factor of 3–4 from the urban center to the rural suburbs. Significant PM<sub>2.5</sub> levels are also  
10 computed over the Mediterranean Sea. The differences between the urban and rural sites are most pronounced for the OC (2–7 times) and EC (6–7 times) concentrations, since large fraction of OC is emitted as primary anthropogenic OC.

### 3.2 Impact of regional biogenic and anthropogenic emissions

The impact of biogenic emissions on air quality due to the chemical interactions with pollutant emissions like NO<sub>x</sub> is investigated by S1, where the biogenic emissions in the  
15 whole model domain are omitted. The differences from the base case simulation (S0) are depicted in Fig. 3d–f. O<sub>3</sub> mixing ratios are reduced throughout the domain (Fig. 3d). The 15-day mean differences are not very large, being 0.56 ppb in Istanbul, 0.46 ppb in Athens and 1.10 ppb at Finokalia. On the other hand, maximum hourly impacts  
20 of 8.85 ppb in Istanbul, 2.92 ppb in Athens and 3.39 ppb at Finokalia are calculated. These results for Istanbul are comparable with the 10 ppb and up to 25 ppb increases in summertime O<sub>3</sub> due to biogenic emissions calculated by Poupkou (2006) and Im et al. (2011c), respectively. It should be noted that the differences in the spatial resolutions of the models used for these studies affect the magnitude of the calculated impacts  
25 (Cohan et al., 2006; Hodnebrog et al., 2011). The omission of biogenic emissions impacts on the OC concentrations via reduction of secondary organic aerosol (SOA) formation (Fig. 4a). The major differences are generally computed over the source areas and downwind. The 15-day mean and hourly maximum absolute changes in

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OC minimize at Finokalia ( $0.05$  and  $0.19 \mu\text{g m}^{-3}$ , respectively), whereas the relative change is highest there (19%). On the opposite, removal of biogenic emissions leads to an increase in secondary inorganic aerosol components and particularly  $\text{nss-SO}_4^{2-}$  and therefore, to an overall increase in  $\text{PM}_{2.5}$  concentrations by 1.9% (Table 4 and Fig. 4b). This result points to the role of biogenic NMVOCs in limiting the production of secondary inorganic species (e.g.  $\text{nss-SO}_4^{2-}$  and  $\text{NO}_3^-$ ) by reducing the oxidant levels in the region.

The contribution of all regional anthropogenic emissions (S5) on the domain-mean gaseous air pollutant levels (Fig. 3g–i) is 19% for  $\text{O}_3$ , 9% for CO, 66% for PAN and 56% for  $\text{HNO}_3$  (Table 4). This anthropogenic contribution (NoAnth) is significantly higher than the biogenic contribution (NoBiog), pointing to the importance of anthropogenic emissions for air quality in the area.

### 3.3 Impacts of megacity emissions

#### 3.3.1 Gases

The impacts of Athens and Istanbul emissions on the spatial distributions of 15-day mean surface  $\text{O}_3$ , PAN and  $\text{PM}_{2.5}$  concentrations are depicted in Fig. 5, where simulations S2 (NoIst), S3 (NoAth) and S4 (NoIstAth) are compared with the base case simulation (S0). This figure clearly shows the large spatial extent of the impact of Istanbul emissions downwind, mostly over Marmara Sea and the Northern Aegean. Although Istanbul emissions have a larger impact on the city itself, Athens has larger  $\text{O}_3$  and PAN contribution on downwind areas and clear effect on the southern Aegean and Eastern Mediterranean (Fig. 5). This result agrees with the previous studies by Poupkou et al. (2008) and Kanakidou et al. (2011). In both cities, omission of anthropogenic emissions leads to an increase of  $\text{O}_3$  in the urban core and a reduction in the suburbs downwind locations. In the urban core, the reduction in  $\text{NO}_x$  levels slows down the NO reaction with  $\text{O}_3$  to form  $\text{NO}_2$  that further reacts with OH radicals producing nitric acid that removes  $\text{NO}_x$  from the system terminating the NMVOC oxidation

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chain without forming O<sub>3</sub> (Finlayson-Pitts and Pitts, 2000). Therefore, in the absence of anthropogenic emissions, NMVOC oxidation in the urban core is enhanced, produces peroxy radicals and finally forms O<sub>3</sub>. For PAN, reductions are calculated for the urban core of Athens and the downwind location Finokalia, whereas an increase is calculated for the Istanbul urban area. The spatial extent of the changes in O<sub>3</sub> mixing ratios follows that of the changes in PAN (Fig. 5). The relative changes in the major gas and aerosol component concentrations in the cities for each scenario with respect to the base case simulation are given in Table 3 and the domain-averaged changes are for scenarios S1 to S5 are presented in Table 4.

Omitting anthropogenic emissions of Istanbul results in a very large increase of O<sub>3</sub> mixing ratios in the urban center by 42 ppb (229 %), whereas the impact on the whole megacity is lower (14 ppb: 28 %). Im et al. (2011a) indicated that Istanbul urban center acts as a net chemical sink for O<sub>3</sub> due to relatively high NO<sub>x</sub> emissions compared to NMVOC emissions as shown in Table 1. Removal of the anthropogenic NO<sub>x</sub> emissions, increases the NMVOC/NO<sub>x</sub> ratio and thus the efficiency of the remaining NO<sub>x</sub> molecules to produce O<sub>3</sub> (Lin et al., 1988). Therefore, O<sub>3</sub> mixing ratios are calculated to increase. Although local impact is dominant, Istanbul plume also affects the O<sub>3</sub> mixing ratios downwind. Istanbul emissions contribute to O<sub>3</sub> levels at Finokalia by 1.5 ppb (2.1 %) that is comparable with the impact of Athens emissions (Table 3). Additionally, Istanbul emissions contribute by 1.3 % to the urban O<sub>3</sub> mixing ratios in Athens and by 0.8 % to the Athens extended area (Table 3).

Omission of Athens emissions leads to an increase in O<sub>3</sub> by up to 13 ppb (~25 %) in the urban core of the city itself (Table 3). The smaller effect of local emissions in Athens compared to Istanbul is due to the higher background levels of pollution in Athens than in Istanbul (Im et al., 2011a; Kanakidou et al., 2011) as discussed earlier. On the other hand, O<sub>3</sub> mixing ratios over the Athens extended area are reduced by 0.7 ppb (~1 %; Table 3). At Finokalia, which is downwind of Athens, there is a decrease of 1.9 ppb (~3 %) in O<sub>3</sub> levels (Table 3). The mean O<sub>3</sub> mixing ratio over the Istanbul extended area also decreases by 0.1 %, showing that Athens plume has minor

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contribution to air quality in Istanbul, due to the dominant northerly winds throughout the simulation period. Anthropogenic emissions from Athens contribute by 0.5 ppb ( $\sim 1\%$ ) to the domain mean surface  $O_3$  mixing ratios (Table 4).

Anthropogenic emissions of both urban agglomerations contribute by 0.9 ppb (1.4%) to the domain mean surface  $O_3$  mixing ratios (Table 4) whereas the percent impact on the downwind FKL is larger (5%) but smaller in absolute amount (0.02 ppb). On the other hand, the city centers are mainly affected by their own emissions (Table 3). However, Athens air pollution further benefits from reduction in Istanbul anthropogenic emissions since these emissions contribute to the elevated background air pollution levels in Athens. Overall, while the impacts over Istanbul remain almost similar to impacts from S2, where only Istanbul emissions are omitted, the impacts over Athens (S4) are relatively larger than the impacts induced by eliminating the Athens emissions only (S3).

The mixing ratios of CO and  $HNO_3$  decrease between 49 and 77% in the urban cores of both cities when the anthropogenic emissions of both cities are masked (Table 3). CO decreases, averaged over the extended areas, are similar ( $\sim 30\%$ ), whereas  $HNO_3$  decreases are larger in Istanbul ( $\sim 53\%$ ) compared to Athens ( $\sim 21\%$ ). An opposite pattern is calculated for PAN; mixing ratios decrease by 6% in Istanbul and by 21% in Athens extended areas. When omitting the anthropogenic emissions of Istanbul,  $NO_x$  is drastically reduced whereas significant NMVOC from vegetation remain. Therefore, the NMVOC/ $NO_x$  molar ratios in the megacity increase by a factor of 8 in the urban core and a factor of 2.8 in the whole extended area. Thus, the chemical environment changes from NMVOC-limited to higher- $O_3$ -production and  $NO_x$ -limited conditions (Dodge, 1977). On the other hand, the already high NMVOC/ $NO_x$  ratios in Athens (77 in rural to 192 in urban; Table 2) are decreasing (by 54% in the urban core and 85% in the whole extended area of the city) when local emissions are masked (Table 3). This is due to the very high anthropogenic NMVOC emissions in Athens compared to the  $NO_x$  emissions. Indeed, the NMVOC/ $NO_x$  molar ratio in the anthropogenic emissions for Athens ( $\sim 73.2$ ) is much higher than that for Istanbul ( $\sim 1.4$ ) (Table 1).

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### 3.3.2 Particulate matter

Concerning aerosol mass concentrations, anthropogenic emissions from Istanbul have a larger influence on the air quality in the region than those from Athens, both in terms of the area influenced and the magnitude of the impact as seen in Fig. 5c and f and Tables 3 and 4. Figure 5f shows that the outflow from Athens particularly extends southward over the south Aegean Sea and the Eastern Mediterranean Sea with major transport pathway between Peloponnesus and Crete followed by significant eastward flow to the South of Crete. On the other hand, Istanbul outflow impacts both southward and northward, however, with main export pathway towards the southwest over the entire Aegean Sea and the Eastern Mediterranean (Fig. 5c). Anthropogenic emissions of Istanbul are responsible for  $19.8 \mu\text{g m}^{-3}$  ( $\sim 75\%$ ) of the calculated  $\text{PM}_{2.5}$  levels in the city's urban core, only  $0.8 \mu\text{g m}^{-3}$  (10%) in the rural suburbs and  $6.2 \mu\text{g m}^{-3}$  (48%) in the whole Istanbul extended area. Similarly, masking the Athens anthropogenic emissions results in a  $2.2 \mu\text{g m}^{-3}$  (22%) reduction in  $\text{PM}_{2.5}$  in the Athens extended area and  $14.9 \mu\text{g m}^{-3}$  (65%) reduction in the urban core of the city, whereas it has no effect on the Istanbul air quality. On the opposite, Istanbul anthropogenic emissions contribute by  $0.2 \mu\text{g m}^{-3}$  (2%) to the  $\text{PM}_{2.5}$  levels in the Athens extended area. Istanbul outflow is responsible for  $0.5 \mu\text{g m}^{-3}$  (5%) whereas Athens emissions contribute by less than  $0.1 \mu\text{g m}^{-3}$  (0.7%) to the  $\text{PM}_{2.5}$  at Finokalia. Domain-wide, Istanbul anthropogenic emissions are responsible for  $0.2 \mu\text{g m}^{-3}$  (2.4%) of the  $\text{PM}_{2.5}$  concentrations whereas Athens emissions have significantly smaller impact ( $0.03 \mu\text{g m}^{-3}$  that is 0.4%). These results clearly show that Istanbul anthropogenic emissions have larger impact on the aerosol levels in the area and downwind, compared to Athens emissions. Furthermore, the removal of all anthropogenic emissions in the model domain (S5) leads to a 28% reduction in the domain-mean  $\text{PM}_{2.5}$  concentrations (Table 4).

The changes in the chemical composition of the aerosols are also summarized in Tables 3 and 4. OC concentrations decrease by  $0.1 \mu\text{g m}^{-3}$  (16%) in Athens and by  $1.7 \mu\text{g m}^{-3}$  (67%) in Istanbul extended areas when each city emissions are masked

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separately. OC concentrations at Finokalia decrease by less than  $0.1 \mu\text{g m}^{-3}$  (7%) when Istanbul emissions are masked, which is 10 times higher than the impact of the Athens emissions ( $0.01 \mu\text{g m}^{-3}$ :  $\sim 1\%$ ). Elemental carbon (EC) concentrations also significantly decrease by  $0.15 \mu\text{g m}^{-3}$  (42%) and  $0.66 \mu\text{g m}^{-3}$  (75%) in the extended areas of Athens and Istanbul, respectively. Athens emissions contribute by  $0.01 \mu\text{g m}^{-3}$  (3%) whereas Istanbul emissions by  $0.03 \mu\text{g m}^{-3}$  (9%) to the EC levels at Finokalia. Domain-wide, 22% of  $\text{nss-SO}_4^{2-}$ , 69% of  $\text{NO}_3^-$ , 32% of OC and 48% of EC concentrations are due to all anthropogenic emissions (S5) in the domain as shown in Table 4.

### 3.4 Impact of “decentralization plan” on air quality

A hypothetical “decentralization” mitigation scenario for the extended areas of Istanbul and Athens has been investigated by a set of two simulations S6 and S7, described in Sect. 2.2. For this (S6: H-Anth), the anthropogenic emissions from the extended areas of these cities (Fig. 2a) are redistributed homogeneously to larger “new” extended areas, as presented in Fig. 2b. In the second simulation (S7: H-AnthBiog), in addition to reallocation of anthropogenic emissions (done in S6), biogenic emissions in the “new” extended areas have been set to those in a sub-urban grid cell of the city (as in the base case scenario S0). The calculated impacts of both mitigation scenarios on the urban, rural and the whole extended areas are summarized in Table 5. The changes in the spatial distributions of  $\text{O}_3$ , PAN and  $\text{PM}_{2.5}$  surface levels in Istanbul, Athens and the model domain are depicted in Fig. 6.

Mixing ratios of  $\text{O}_3$  increase over both cities as expected, particularly due to the reductions in  $\text{NO}_x$  emissions (Fig. 6a, d, g). However the magnitudes of the impacts differ significantly between the two cities, due to the large difference in the  $\text{NO}_x$  emissions that result in very different NMVOC/ $\text{NO}_x$  ratios in the emissions. Thus, NMVOC/ $\text{NO}_x$  molar ratios increase by  $\sim 150\%$  over Istanbul and decrease by  $\sim 51\%$  over Athens. Therefore, in Istanbul urban  $\text{O}_3$  increases by more than a factor of 2, whereas in Athens the increase is much smaller ( $\sim 26\%$ ).

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The impacts on the rural levels are opposite in sign to those in the urban areas (Table 5). Due to the reallocation of the anthropogenic emissions from the urban core to rural areas, almost all species concentrations increase in these rural areas. The only exception is  $O_3$ , which decreases by 2 % in Istanbul and 1 % in Athens, due to increase in  $NO_x$  emissions at levels high enough to titrate  $O_3$  (Fig. 6a, d, g). Overall, the responses are larger over the Istanbul extended area compared to the Athens extended area.  $O_3$  mixing ratios over the Istanbul extended area increase by 15 % while over the Athens extended area,  $O_3$  decreases by 2 %. Around Istanbul  $O_3$  is increasing also over the Marmara Sea (to the South West of the city) downwind Istanbul, then decreases in the outflow over the Aegean and East Mediterranean seas. Similar patterns but less extended are computed for the  $O_3$  outflow from Athens. PAN mixing ratios follow the  $O_3$  changes with higher increases in Istanbul (38 %, Fig. 6b) than in Athens (1 %, Fig. 6e); and decrease downwind (Fig. 6h). On the other hand, urban CO mixing ratios decrease to less than half in Istanbul and by 72 % in Athens and by 22 % and 17 % in their extended areas, respectively.

Urban  $PM_{2.5}$  and major aerosol component concentrations decrease in both cities as seen in Table 5 and Fig. 6c, f, i. Due to higher  $PM_{2.5}$  emissions in Istanbul, the reductions in  $PM_{2.5}$  levels are larger there (67 %) than in Athens (59 %). Decreases have been also computed for all individual urban aerosol components in Istanbul (18 % for  $nss-SO_4^{2-}$ , 93 % for  $NO_3^-$ , 82 % for OC and 84 % for EC) and in Athens (17 % for  $nss-SO_4^{2-}$ , 50 % for  $NO_3^-$ , 46 % for OC and 75 % for EC). Rural  $PM_{2.5}$  concentrations increase by 10 % over Istanbul and 12 % over Athens (Fig. 6c, f, i). An overall reduction of aerosol concentrations is achieved by mitigating the particulate emissions. Over the Istanbul extended area,  $PM_{2.5}$  concentrations decrease by 32 %,  $nss-SO_4^{2-}$  by 4 %,  $NO_3^-$  by 70 %, OC by 51 % and EC by 56 %. Over the Athens extended area,  $PM_{2.5}$  is reduced by 9 %,  $nss-SO_4^{2-}$  by 2 %,  $NO_3^-$  by 35 %, OC by 7 % and EC by 21 %.

The second mitigation scenario (S7), considering changes in biogenic emissions in the new extended areas, shows very similar impacts with those calculated from scenario S6 (Table 5). However, there are small differences on the secondary species

response to the re-allocation of the biogenic emissions. A striking difference is seen in the response of rural O<sub>3</sub> mixing ratios in Istanbul. While the re-distribution of the anthropogenic emissions leads to a decrease in the rural O<sub>3</sub> mixing ratios by ~2%, the impact of biogenic emissions leads to an increase of ~2%. This is due to the adoption of overall higher biogenic emissions in the “new” extended area than in the base simulation S0 that enhance O<sub>3</sub> formation in the area. The concentrations of species that are not related to biogenic emissions, EC for instance, do not change between the two simulations.

## 4 Conclusions

In the present study, WRF/CMAQ modeling system, coupled with the MEGAN biogenic emissions model, has been employed to investigate the impacts of Eastern Mediterranean megacity emissions on the local and regional summertime air quality. A number of emission scenarios have been applied over the extended areas of Istanbul and Athens to quantify the responses of urban and rural air quality to extreme abatement strategies. Athens emissions have regional (0.8 % for O<sub>3</sub>) and downwind (2.7 % of O<sub>3</sub> at Finokalia) impacts on the gaseous pollutants larger than those of Istanbul emissions (0.6 % domain-wide and 2.1 % at Finokalia). On the other hand, considering the local effects, Istanbul emissions have a larger contribution to the urban gaseous air pollutants (229 % for O<sub>3</sub>) in the city itself than those of Athens (25 % for O<sub>3</sub>). Istanbul emissions also have a larger contribution to the aerosol levels both locally (75 % for PM<sub>2.5</sub>) and regionally (2.4 % for PM<sub>2.5</sub>) compared to Athens, which has a local contribution of 65 % and domain-wide contribution of 0.4 %.

The impact of mitigation by a “hypothetical” de-centralization plan represented in the model through homogeneous redistribution of anthropogenic emissions results in higher O<sub>3</sub> mixing ratios at the emission hot spots (215 % and 26 % in Istanbul and Athens, respectively). On the opposite, significant reductions are calculated for pollutants dominated by primary emissions like CO (55 % and 72 % for Istanbul and Athens)

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and PM<sub>2.5</sub> (67 % and 60 % in Istanbul and Athens, respectively). However, the concentrations of most gases and aerosols at the rural sites of these megacities increase; 5 % and 16 % for CO, and 10 % and 12 % for PM<sub>2.5</sub> in Istanbul and Athens, respectively.

The results point out improvements for most gaseous and aerosol species in the urban areas of the cities, which could decrease the exceedences and the human exposure. Although the re-allocation of the anthropogenic emissions in the hot spots towards the rural areas increases the concentrations in these areas, the mean pollutant levels in the extended areas also decrease for most air pollutants. The studied mitigation scenario leads to overall decreases in pollutant surface levels over the Istanbul and Athens “new” extended areas, as follows: PM<sub>2.5</sub> by 32 % and 9 %, EC by 56 % and 21 %, CO mixing ratios by 23 % and 17 %, HNO<sub>3</sub> mixing ratios by 26 % and 7 %. On the opposite, O<sub>3</sub> and PAN mixing ratios increase overall the extended domain in Istanbul by 15 % and 6 %, respectively, whereas decreases by 2 % and 4 %, respectively, are calculated for the Athens extended area that experiences high levels of regional background O<sub>3</sub>.

Due to the non-linear responses of air pollution levels to the emission changes, higher spatial resolution and longer simulations that also include sector-based evaluations are needed to increase the accuracy of the results presented in this study and to evaluate the seasonal responses to these abatement strategies. Carefully designed decentralization plans have to be accompanied by emission abatement strategies and require thorough evaluation by dedicated high resolution mesoscale modeling.

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**Table 1.** Anthropogenic and biogenic emissions of Athens and Istanbul extended areas integrated over the 15-day simulation period. Anth denotes when only the anthropogenic emissions are accounted; AnthBiog denotes all emissions; Biog denotes when only the biogenic and soil emissions are accounted. NMVOC/NO<sub>x</sub> and CO/NO<sub>x</sub> are molar ratios.

| Pollutants (tons)     | Istanbul           |          |          | Athens             |          |          |
|-----------------------|--------------------|----------|----------|--------------------|----------|----------|
|                       | Anthropogenic land | shipping | Biogenic | Anthropogenic land | shipping | Biogenic |
| CO                    | 14 515             | 53       | –        | 21 497             | 252      | –        |
| NO <sub>x</sub>       | 9140               | 1036     | 5*       | 3450               | 1024     | 18*      |
| SO <sub>2</sub>       | 3084               | 1159     | –        | 1867               | 1083     | –        |
| NH <sub>3</sub>       | 91                 | –        | –        | 88                 | –        | –        |
| NMVOC                 | 6472               | 65       | 903      | 5715               | 141      | 2166     |
| PM <sub>10</sub>      | 1741               | 155      | –        | 352                | 32       | –        |
| PM <sub>2.5</sub>     | 979                | 146      | –        | 275                | 25       | –        |
|                       | Anth               | AnthBiog | Biog     | Anth               | AnthBiog | Biog     |
| NMVOC/NO <sub>x</sub> | 1.4                | 1.7      | 79       | 73.2               | 76.0     | 54       |
| CO/NO <sub>x</sub>    | 1.4                | 1.4      |          | 4.8                | 4.8      |          |

\* soil emissions.



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**Table 2.** Surface concentrations of main gaseous and particulate pollutants and ratios of non methane volatile organic compounds (NMVOC) to nitrogen oxides (NO<sub>x</sub>) and CO/NO<sub>x</sub> simulated for the base case and averaged over the 15-day period.

| Compounds   | IST   |       |          | ATH   |       |          | FKL   |
|---|-------|-------|----------|-------|-------|----------|-------|
|   | Urban | Rural | Extended | Urban | Rural | Extended | Rural |
| O <sub>3</sub> (ppb)                                    | 18.5  | 65.2  | 49.8     | 51.7  | 60.4  | 64.2     | 69.9  |
| CO (ppb)  | 288.4 | 129.3 | 170.0    | 503.3 | 117.2 | 164.2    | 122.5 |
| PAN (ppb)   | 0.4   | 0.5   | 0.5      | 0.4   | 0.3   | 0.4      | 0.6   |
| HNO <sub>3</sub> (ppb)                                  | 2.5   | 1.4   | 1.9      | 1.4   | 0.8   | 1.0      | 0.7   |
| NMVOC/NO <sub>x</sub>                                   | 2.5   | 12.9  | 5.7      | 191.9 | 77.0  | 102.6    | 39.5  |
| CO/NO <sub>x</sub>                                      | 2.8   | 30.4  | 5.8      | 13.7  | 97.8  | 32.2     | 111.1 |
| PM <sub>2.5</sub> (μg m <sup>-3</sup> )                 | 26.5  | 7.5   | 12.9     | 22.9  | 8.1   | 9.8      | 8.6   |
| nss-SO <sub>4</sub> <sup>2-</sup> (μg m <sup>-3</sup> ) | 6.3   | 4.5   | 5.3      | 6.8   | 5.2   | 5.6      | 5.7   |
| NO <sub>3</sub> <sup>-</sup> (μg m <sup>-3</sup> )      | 0.34  | 0.04  | 0.11     | 0.04  | 0.02  | 0.02     | 0.06  |
| OC (μg m <sup>-3</sup> )                                | 6.9   | 1.1   | 2.5      | 1.6   | 0.8   | 0.9      | 0.9   |
| EC (μg m <sup>-3</sup> )                                | 2.6   | 0.3   | 0.9      | 1.2   | 0.3   | 0.4      | 0.3   |

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**Table 3.** Differences (%) in the 15-day mean surface concentrations of air pollutants and chemical indicators in Istanbul and in Athens (urban core, rural suburbs and extended area) due to omission of anthropogenic emissions over the Istanbul and/or Athens extended areas.

|  | IST       |           |              | ATH       |           |              | FKL (%) |
|--|-----------|-----------|--------------|-----------|-----------|--------------|---------|
|  | Urban (%) | Rural (%) | Extended (%) | Urban (%) | Rural (%) | Extended (%) |         |
| <b>O<sub>3</sub></b>                   |           |           |              |           |           |              |         |
| Nolst                                  | 229.2     | 2.5       | 28.4         | -1.3      | -0.3      | -0.8         | -2.1    |
| NoAth                                  | -0.1      | -0.1      | -0.1         | 24.7      | -0.3      | -1.1         | -2.7    |
| Nolst&Ath                              | 229.1     | 2.5       | 28.3         | 23.2      | -0.9      | -1.9         | -4.8    |
| <b>CO</b>                              |           |           |              |           |           |              |         |
| Nolst                                  | -59.4     | -4.8      | -29.5        | -0.2      | -0.4      | -0.3         | -1.8    |
| NoAth                                  | 0         | -0.1      | 0            | -76.6     | -5.5      | -30.7        | -4.0    |
| Nolst&Ath                              | -59.5     | -4.9      | -29.6        | -76.8     | -5.9      | -31.0        | -5.8    |
| <b>PAN</b>                             |           |           |              |           |           |              |         |
| Nolst                                  | 5.0       | -3.6      | -6.4         | -5.2      | -3.1      | -3.2         | -8.9    |
| NoAth                                  | -0.3      | -0.3      | -0.3         | -15.0     | -10.4     | -20.9        | -9.7    |
| Nolst&Ath                              | 4.7       | -3.8      | -6.7         | -20.4     | -13.4     | -24.3        | -18.6   |
| <b>HNO<sub>3</sub></b>                 |           |           |              |           |           |              |         |
| Nolst                                  | -67.6     | -26.6     | -53.2        | -7.9      | -5.5      | -6.5         | -12.1   |
| NoAth                                  | 0         | -0.1      | 0            | -41.8     | -7.3      | -21.2        | -1.5    |
| Nolst&Ath                              | -67.5     | -26.6     | -53.1        | -49.4     | -12.7     | -27.6        | -13.6   |
| <b>NMVOC/NO<sub>x</sub></b>            |           |           |              |           |           |              |         |
| Nolst                                  | 802.5     | 57.0      | 277.9        | -38.2     | 0.5       | 0.1          | -0.7    |
| NoAth                                  | -0.1      | -0.1      | -0.3         | -53.9     | -16.8     | -84.9        | -44.4   |
| Nolst&Ath                              | 795.0     | 54.7      | 273.9        | -65.6     | -15.8     | -40.5        | -45.2   |
| <b>CO/NO<sub>x</sub></b>               |           |           |              |           |           |              |         |
| Nolst                                  | 3688.7    | 316.8     | 1832.2       | 687.1     | -0.1      | -0.3         | 0.2     |
| NoAth                                  | 0         | -0.2      | -0.1         | 1497.6    | 133.9     | 518.9        | -4.7    |
| Nolst&Ath                              | 3680.7    | 316.1     | 1827.7       | 774.2     | 134.9     | 524.9        | -4.5    |
| <b>PM<sub>2.5</sub></b>                |           |           |              |           |           |              |         |
| Nolst                                  | -74.8     | -9.9      | -47.9        | -1.3      | -1.9      | -2.0         | -5.5    |
| NoAth                                  | 0         | 0         | 0            | -65.1     | -8.4      | -22.4        | -0.7    |
| Nolst&Ath                              | -74.8     | -10.0     | -48.0        | -65.2     | -10.1     | -24.4        | -6.2    |
| <b>nss-SO<sub>4</sub><sup>2-</sup></b> |           |           |              |           |           |              |         |
| Nolst                                  | -28.8     | -3.2      | -17.3        | -2.9      | -1.5      | -2.4         | -5.0    |
| NoAth                                  | 0         | 0         | 0            | -19.9     | -0.6      | -5.1         | -0.7    |
| Nolst&Ath                              | -28.8     | -3.2      | -17.3        | -22.9     | -2.7      | -7.5         | -5.6    |
| <b>NO<sub>3</sub><sup>-</sup></b>      |           |           |              |           |           |              |         |
| Nolst                                  | -94.8     | -16.0     | -79.2        | -9.7      | -4.1      | -7.0         | -22.4   |
| NoAth                                  | -0.2      | 0         | -0.1         | -54.7     | -53.2     | -34.9        | -1.4    |
| Nolst&Ath                              | -94.8     | -16.0     | -79.2        | -62.2     | -59.2     | -43.0        | -23.7   |
| <b>OC</b>                              |           |           |              |           |           |              |         |
| Nolst                                  | -88.4     | -17.8     | -67.3        | -1.9      | -1.8      | -2.1         | -6.6    |
| NoAth                                  | 0         | 0         | 0            | -50.4     | -5.2      | -15.6        | -1.4    |
| Nolst&Ath                              | -88.4     | -17.9     | -67.3        | -52.2     | -7.0      | -17.8        | -8.1    |
| <b>EC</b>                              |           |           |              |           |           |              |         |
| Nolst                                  | -91.5     | -24.1     | -74.5        | -1.2      | -2.6      | -2.6         | -9.4    |
| NoAth                                  | 0         | 0         | 0            | -81.1     | -24.8     | -42.1        | -3.0    |
| Nolst&Ath                              | -91.5     | -24.2     | -74.6        | -82.3     | -27.4     | -44.7        | -12.4   |

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**Table 4.** Domain-mean surface concentrations from the base case simulation (S0), NoBiog (S1), Nolst (S2), NoAth (S3), NolstAth (S4), NoAnth (S5), decentralization with anthropogenic emissions, H-Anth (S6), and decentralization with anthropogenic and biogenic emissions, H-AnthBiog (S7) scenarios, averaged over the 15-day simulation period. O<sub>3</sub>, CO, PAN, HNO<sub>3</sub> are given in ppb, NMVOC/NO<sub>x</sub> and CO/NO<sub>x</sub> are molar ratios, PM<sub>2.5</sub>, nss-SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup>, OC, EC are given in μg m<sup>-3</sup>. The parenthesis present the percent differences of scenarios S1–S5 relative to the base case scenario.

| Compounds                         | Base  | Nolst        | NoAth        | NolstAth     | NoBiog       | NoAnth        | H-Anth       | H-AnthBiog   |
|-----------------------------------|-------|--------------|--------------|--------------|--------------|---------------|--------------|--------------|
| O <sub>3</sub>                    | 67.5  | 67.1 (-0.6)  | 67.0 (-0.8)  | 66.5 (-1.4)  | 66.5 (-1.5)  | 54.8 (-18.8)  | 67.6 (0.2)   | 67.7 (0.3)   |
| CO                                | 121.6 | 120.8 (-0.7) | 119.8 (-1.5) | 119.0 (-2.2) | 118.9 (-2.3) | 111.3 (-8.5)  | 120.8 (-0.7) | 120.9 (-0.6) |
| PAN                               | 0.53  | 0.52 (-2.4)  | 0.51 (-3.6)  | 0.50 (-6.1)  | 0.5 (-10.3)  | 0.2 (-66.4)   | 0.53 (0.8)   | 0.54 (1.9)   |
| HNO <sub>3</sub>                  | 1.1   | 1.0 (-6.1)   | 1.1 (-0.1)   | 1.0 (-6.0)   | 1.1 (5.6)    | 0.5 (-56.3)   | 1.1 (-2.9)   | 1.0 (-3.3)   |
| NMVOC/NO <sub>x</sub>             | 30.8  | 30.9 (0.5)   | 25.0 (-18.8) | 25.1 (-18.4) | 25.8 (-16.3) | 64.9 (111.0)  | 29.0 (-5.7)  | 29.1 (-5.3)  |
| CO/NO <sub>x</sub>                | 76.3  | 82.7 (8.3)   | 76.7 (0.5)   | 83.3 (9.1)   | 75.3 (-1.3)  | 650.7 (752.5) | 79.4 (4.0)   | 79.6 (4.3)   |
| PM <sub>2.5</sub>                 | 8.2   | 8.0 (-2.4)   | 8.2 (-0.4)   | 8.0 (-2.8)   | 8.4 (1.9)    | 5.9 (-27.9)   | 8.2 (-0.7)   | 8.2 (-0.8)   |
| nss-SO <sub>4</sub> <sup>2-</sup> | 5.2   | 5.1 (-2.1)   | 5.2 (-0.2)   | 5.1 (-2.2)   | 5.4 (2.7)    | 4.1 (-22.0)   | 5.2 (-0.4)   | 5.2 (-0.5)   |
| NO <sub>3</sub> <sup>-</sup>      | 0.1   | 0.1 (-4.8)   | 0.1 (-0.2)   | 0.1 (-4.9)   | 0.1 (4.3)    | 0.0 (-68.9)   | 0.1 (-2.5)   | 0.1 (-2.6)   |
| OC                                | 1.0   | 1.0 (-2.7)   | 1.0 (-0.4)   | 1.0 (-3.1)   | 0.9 (-5.3)   | 0.7 (-32.1)   | 1.0 (-1.2)   | 1.0 (1.0)    |
| EC                                | 0.3   | 0.3 (-4.1)   | 0.3 (-1.3)   | 0.3 (-5.4)   | 0.3 (0.0)    | 0.2 (-47.6)   | 0.3 (-1.8)   | 0.3 (-1.8)   |

**Table 5.** Changes (%) in surface concentrations of the main pollutants and chemistry indicators compared to the base case simulations when assuming “hypothetical decentralization plan” for Istanbul and Athens (S5; H-Anth) that is also accompanied by changes in biogenic emissions (S6; H-AnthBiog).

|  | IST       |           |              | ATH       |           |              |
|--|-----------|-----------|--------------|-----------|-----------|--------------|
|  | Urban (%) | Rural (%) | Extended (%) | Urban (%) | Rural (%) | Extended (%) |
| <b>O<sub>3</sub></b>                   |           |           |              |           |           |              |
| H-Anth                                 | 215.1     | -2.2      | 14.9         | 25.6      | -0.9      | -1.8         |
| H-AnthBiog                             | 221.6     | 1.9       | 17.4         | 26        | -0.7      | -1.5         |
| <b>CO</b>                              |           |           |              |           |           |              |
| H-Anth                                 | -54.5     | 4.7       | -22.9        | -72.4     | 16.1      | -17.2        |
| H-AnthBiog                             | -54.3     | 5.1       | -22.6        | -72.4     | 16.3      | -17.1        |
| <b>PAN</b>                             |           |           |              |           |           |              |
| H-Anth                                 | 38.4      | 6.0       | 10.6         | 1.2       | 3.3       | -4.1         |
| H-AnthBiog                             | 52.8      | 24.0      | 22.4         | 4.8       | 5.9       | -1.5         |
| <b>HNO<sub>3</sub></b>                 |           |           |              |           |           |              |
| H-Anth                                 | -41.2     | 10.8      | -25.8        | -33.9     | 12.8      | -6.6         |
| H-AnthBiog                             | -44.2     | 6.5       | -28.9        | -34.6     | 12.8      | -7.1         |
| <b>NMVOC/NO<sub>x</sub></b>            |           |           |              |           |           |              |
| H-Anth                                 | 145.5     | -55.7     | 14.9         | -50.9     | 42.2      | 5.9          |
| H-AnthBiog                             | 186.1     | -44.5     | 30.8         | -50.8     | 42.6      | 5.7          |
| <b>CO/NO<sub>x</sub></b>               |           |           |              |           |           |              |
| H-Anth                                 | 580.5     | -33.3     | 223.2        | 236.1     | -53.6     | 52.4         |
| H-AnthBiog                             | 590.7     | -30.5     | 229.1        | 236.6     | -53.6     | 51.8         |
| <b>PM<sub>2.5</sub></b>                |           |           |              |           |           |              |
| H-Anth                                 | -66.7     | 10.1      | -32.1        | -59.3     | 11.7      | -9.1         |
| H-AnthBiog                             | -66.6     | 11.9      | -31.8        | -59.3     | 11.6      | -9.1         |
| <b>nss-SO<sub>4</sub><sup>2-</sup></b> |           |           |              |           |           |              |
| H-Anth                                 | -17.8     | 5.4       | -4.4         | -16.9     | 4.5       | -2.0         |
| H-AnthBiog                             | -18.2     | 5.1       | -4.9         | -17.1     | 4.3       | -2.2         |
| <b>NO<sub>3</sub><sup>-</sup></b>      |           |           |              |           |           |              |
| H-Anth                                 | -92.6     | 10.6      | -69.7        | -48.9     | -41.3     | -34.9        |
| H-AnthBiog                             | -92.7     | 8.6       | -70.2        | -49.4     | -41.7     | -35.5        |
| <b>OC</b>                              |           |           |              |           |           |              |
| H-Anth                                 | -81.9     | 13.2      | -51.3        | -45.5     | 5.9       | -6.5         |
| H-AnthBiog                             | -81.4     | 22.6      | -49.7        | -45.1     | 5.8       | -5.9         |
| <b>EC</b>                              |           |           |              |           |           |              |
| H-Anth                                 | -84.3     | 17.7      | -55.9        | -75.2     | 2.4       | -21.0        |
| H-AnthBiog                             | -84.3     | 17.6      | -55.9        | -75.2     | 2.4       | -21.0        |

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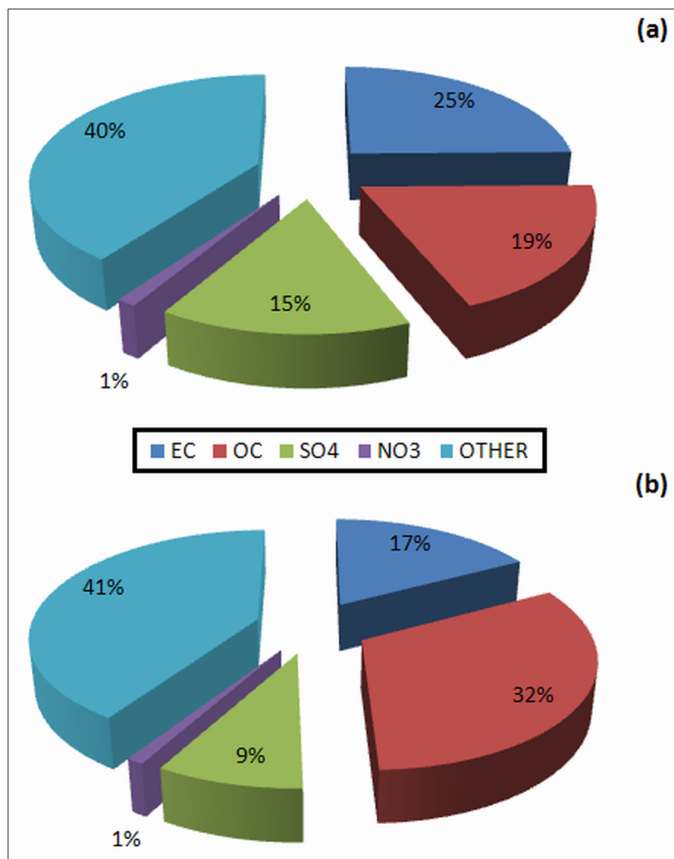
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**Fig. 1.** Speciation of PM<sub>2.5</sub> mass emissions for (a) Athens and (b) Istanbul anthropogenic emissions.

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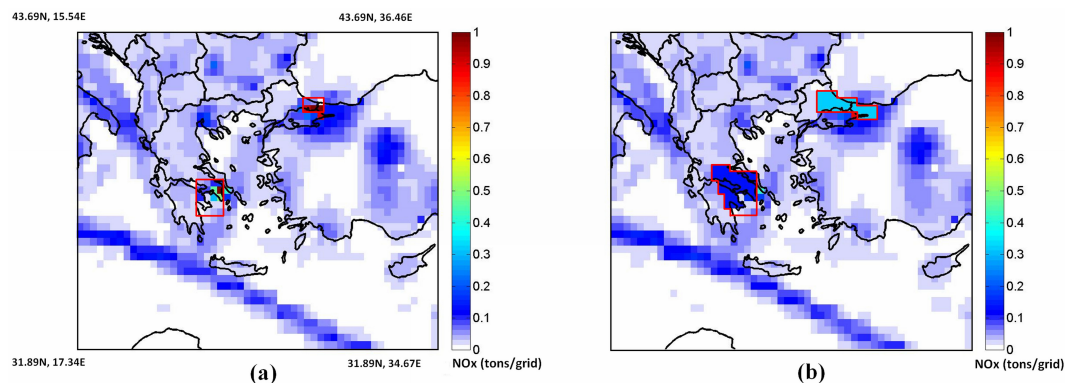
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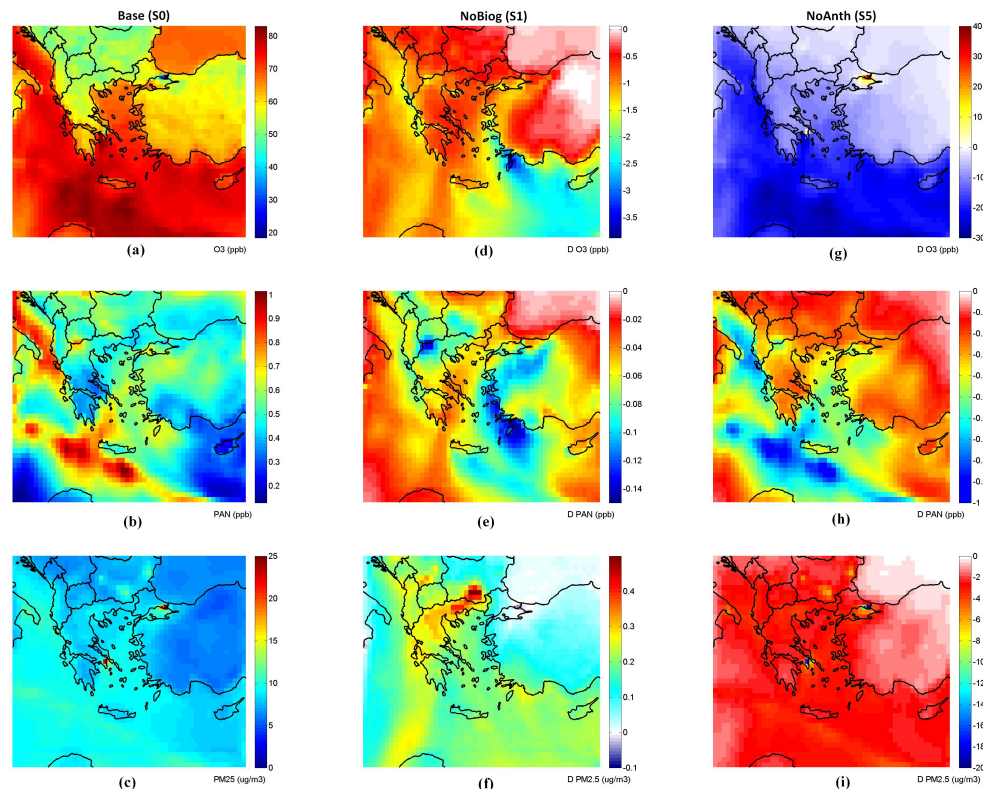


**Fig. 2.** Anthropogenic NO<sub>x</sub> emissions before (a) and after decentralization (b) integrated over the 15-day simulation period. The red boxes show the original (a) and new (b) extended areas of Istanbul and Athens.

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**Fig. 3.** Surface O<sub>3</sub> (top panels), PAN (mid-panels) and PM<sub>2.5</sub> (bottom panels) concentrations calculated from the base case scenario (S0), left column, and the differences (%) of scenarios NoBiog (S1), middle column, and NoAnth (S5), right column, relative to the base case scenario, averaged over the 15-day simulation period.

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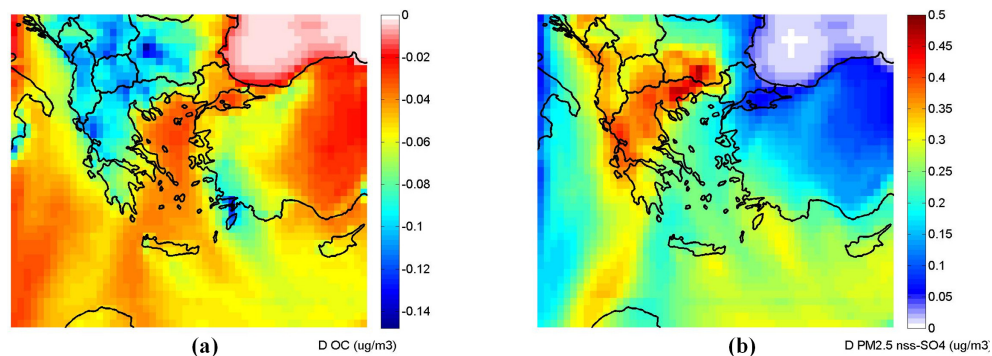
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**Fig. 4.** Impact of biogenic emissions on **(a)** OC and **(b)**  $\text{nss-SO}_4^{2-}$  surface concentrations, calculated as the difference of scenario S1 (NoBiog) from the base case scenario (S0), averaged over the 15-day simulation period.

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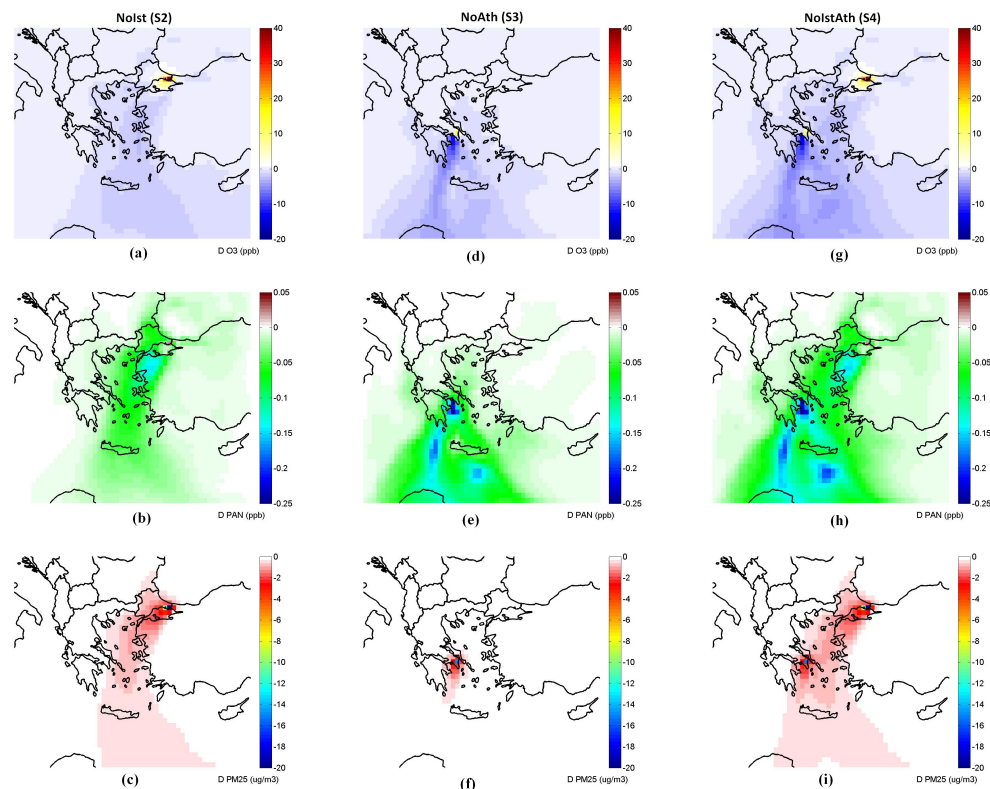
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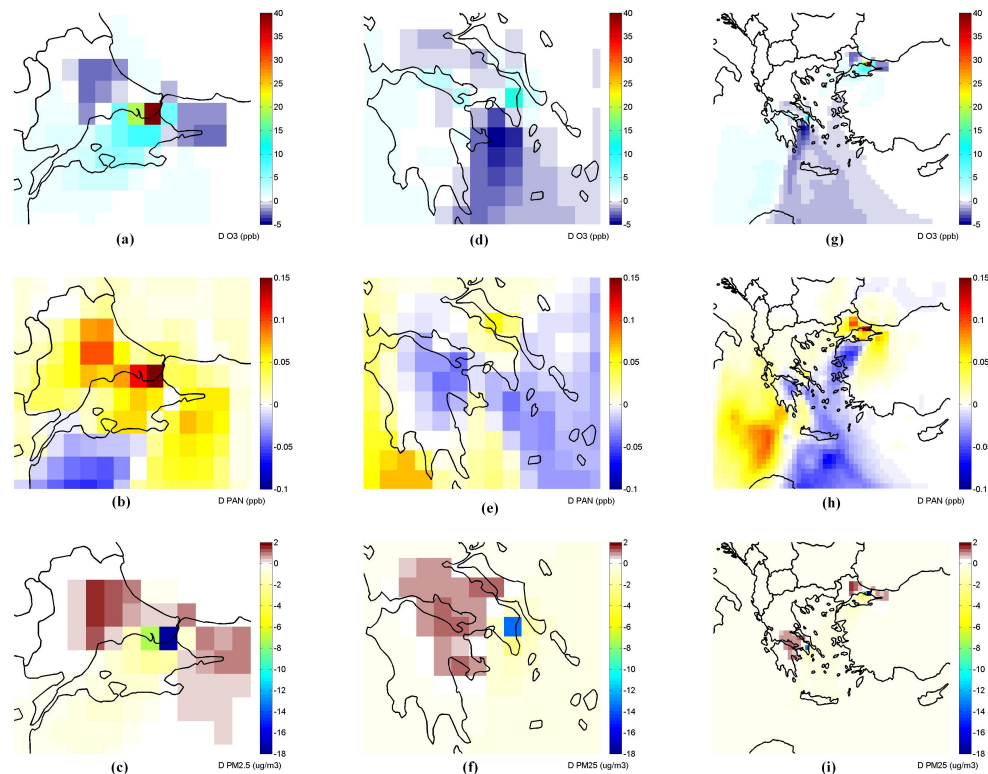


**Fig. 5.** Changes in O<sub>3</sub> (top panels), PAN (mid-panels) and PM<sub>2.5</sub> (bottom panels) concentrations for scenarios NoIst (S2), left column, NoAth (S3), middle column, and NoIstAth (S4), right column, relative to the base case scenario (S0), averaged over the 15-day simulation period.

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**Fig. 6.** Surface concentration changes due to the hypothetical decentralization of anthropogenic emissions (H-Anth; S6) on O<sub>3</sub> (top panels), PAN (mid-panels) and PM<sub>2.5</sub> (bottom panels) concentrations in Istanbul, left column, Athens, middle column, and the simulation domain, right column, averaged over the 15-day simulation.

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