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# ATHENS AIR QUALITY AND IMPORTANCE OF BIOGENIC EMISSIONS: A CASE STUDY

**P. POLYMENEAS**
**C. PILINIS***

*Department of Environment, University of the Aegean*
*University Hill, GR 81100 Mytilene, Greece*

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*to whom all correspondence should be addressed:
e-mail: xpil@aegean.gr

**ABSTRACT**
Two three dimensional Eulerian air quality models, PMCAMx and UAM-AERO are used to predict the concentrations of various pollutants in the Greater Athens Area (GAA) during the episode of June 22-26, 2003. The predicted levels of gases and aerosol pollutants are compared with measured concentrations from monitoring stations, in order to evaluate the performance of the models. Comparison of the results reveals that PMCAMx simulated the ozone diurnal pattern better than UAM-AERO, as the predicted by UAM-AERO maximum daytime concentrations were much higher than those of PMCAMx and way above the measured ones. PMCAMx also shows signs of superiority in terms of its ability to predict the $NO_2$ and NO concentrations. Two sets of simulations, one with both anthropogenic and biogenic emissions and one without biogenic emissions were performed. Comparison of the two simulations reveals the important role that biogenic emissions play in the formation of $O_3$, NOx and secondary aerosols. Additional simulations with PMCAMx show that the concentrations of all the pollutants depict a highly non-linear behaviour, when anthropogenic VOC emissions are reduced in the GAA.

**KEYWORDS**: Biogenic emissions; Ozone; Nitrogen dioxide; Nitrogen Monoxide; VOCs; Aerosols; Modeling; PMCAMx; UAM-AERO

## 1. INTRODUCTION

The problem of air pollution in Athens has been of great concern for the last 30 years, because of the imposed state and Commission of the European Communities limits on ambient $O_3$, $NO_2$ and NO concentrations. Numerous scientific papers studying the air pollution in Athens have been published in the international literature (Flassak and Moussiopoulos, 1988; Asimakopoulos *et al.*, 1992; Kallos *et al.*, 1993; Pilinis *et al.*, 1993; Ziomas, 1998; Sotiropoulou *et al.*, 2004; 2006). A number of critical factors have been confirmed which contribute to the formation of high levels of photochemical pollutants in the basin; the poor ventilation, the frequent appearance of the sea breeze, the intense solar irradiation combined with intense anthropogenic activity and finally the biogenic emissions (Bossioli *et al.*, 2007). The main anthropogenic sources of air pollution in the Greater Athens Area (GAA) can be classified into three main categories: a) industry (≈ 40% of the total Greek industrial activity), b) transportation (≈ 50% of the total automobile traffic) and c) heating. The main source of photochemical pollutants is considered to be transportation. The harbor of Piraeus, the biggest in the country and the international airport of Athens are also considered important sources of air pollution. Most of the industrial activities in the GAA and the country are concentrated in the Thriassion plain (Pilinis *et al.*, 1993).

In addition to anthropogenic emissions, biogenic emissions exist in this area. In most Mediterranean countries, the burden of biogenic emissions is comparable with that from anthropogenic sources (Simpson *et al.*, 1999). The role of anthropogenic and biogenic emissions on tropospheric ozone formation over Greece has also been simulated (Mimilidis *et al.*, 1994; Tsiligiridis and Pistikopoulos, 1995; Varinou *et al.*, 1999). The simulations revealed

that there was a significant increase of the calculated ozone concentrations over areas with significant precursor sources, when biogenic emissions are taken into account. Besides the fact that the biogenic emissions govern the air chemistry and SOA (secondary organic aerosol) formation in remote areas, biogenic sources have been documented to affect significantly air pollution in urban and near-urban environments. As an important amount of precursor pollutants is emitted by biogenic sources (Varinou *et al.*, 1999) their omission may result in insufficient control strategies. Air quality modelling, supported by monitoring activities at the regions under consideration, seems to be the only practical way today to cope with such a complex problem.

The aim of this particular article is to examine the contribution of biogenic emissions in the formation of $O_3$, NOx and aerosols in the Greater Athens Area (GAA), using two three dimensional air quality models. The models were applied during the four-day episode in the Greater Athens Area that started at 0000 (LST) of June 22, 2003 until 0000 (LST) of June 26, 2003 (Sotiropoulou *et al.*, 2006). Finally, we examine the effects of control measures to the concentrations of air-pollutants in downtown area of Athens and Piraeus. The results of the models that concern the levels of gaseous and particulate pollutants reveal the importance of biogenic emissions in the pollution levels of the region and the necessity that they be included in the application of air quality models, as well as in the enforcement measures and control strategies.

## 2. DESCRIPTION OF THE AREA

The residential area of Athens is extended in the Attica peninsula and includes the Athens basin, Piraeus, the Thriassion and Mesogea plains. The total structured area is about 350 $km^2$, where more than 4 millions residents live. The cities of Athens, Piraeus and their suburbs are located in an elongated basin surrounded by 6 high mountains (Kitheronas, Pateras, Parnitha, Penteli, Hymettus and Egaleo) from three sides and open to the sea from the fourth. The basin is surrounded by mountains with elevation up to 1400 m while to the south is defined by the coast. These mountains are the physical barriers between the Athens Basin and the Thriassion and Mesogea plains. Between those mountains there are three big physical gaps. Moreover, in the Athens Basin there are some hills, with elevation up to 150 m above the bottom of the basin. The geographic expansion of the area not followed by the parallel development of the necessary infrastructures (road network, parks etc.) causes, for the last 40 years, increasing problems of pollution all over the basin.

## 3. DESCRIPTION OF THE MODELS

### 3.1 THE PM COMPREHENSIVE AIR QUALITY MODEL WITH EXTENSIONS

The PM Comprehensive Air Quality Model with Extensions (PMCAMx) is an extended version of the three-dimensional photochemical model CAMx. CAMx is an Eulerian photochemical grid model that allows the assessment of gaseous and particulate air-pollution over many scales ranging from urban to super regional. PMCAMx simulates the emission, dispersion, chemical reactions and removal of pollutants in the lower troposphere by solving the pollutant continuity equation for each chemical species on a system of nested three-dimensional grids. It is designed to unify all of the technical features required of “state-of-the-science” air quality models into a single system that is computationally efficient, easy to use, and publicly available. The model code has a highly modular and well documented structure which eases the insertion of new or alternate algorithms and features. The input (gridded surface emissions, fractional landuse distribution, height/pressure, horizontal wind components, temperatures, vertical diffusivities, water vapor, clouds and precipitation) and output file formats are based on the Urban Airshed Model and are compatible with many existing pre- and post-processing tools (ENVIRON, 2005).

PMCAMx uses the framework of CAMx and detailed aerosol modules to simulate inorganic aerosol growth, aqueous-phase chemistry, secondary organic aerosol formation, nucleation, and coagulation. The simulation order of the processes of PMCAMx is: emissions, horizontal advection, vertical advection, vertical dispersion, horizontal dispersion, wet deposition, gas-phase chemistry, aerosol processes (nucleation, coagulation, inorganic aerosol condensation and evaporation), SOA growth, and aqueous-phase chemistry. The PMCAMx gas-phase

chemistry was modified to simulate the formation of secondary aerosol precursors. The changes were as follows: i) tracks production of four condensable gasses (CG1 – CG4) formed in the oxidation of VOCs, that are secondary organic aerosol (SOA) precursors, ii) adds a new olefin species, called OLE2, to represent biogenic olefins so that high SOA-yield biogenic olefins (monoterpenes) can be distinguished from low SOA-yield anthropogenic olefins, iii) tracks ammonia as a precursor to ammonium aerosols, iv) tracks $SO_2$ and gaseous sulfuric acid as precursors to sulfate aerosols, v) tracks HCl as a precursor to and vaporization product of sea salt (sodium chloride) aerosols. The chemical mechanisms supported by PMCAMx are the Carbon Bond mechanism version IV (CBM-IV) or the SAPRC99. In the simulations performed by PMCAMx the SAPRC99 chemical mechanism has been used (Gaydos *et al.*, 2007).

### 3.2 THE URBAN AIRSHED MODEL – AEROSOL

The UAM-AERO model (Sonoma Technology, Inc., 1996) is a gas/aerosol model that is based on the air quality model UAM version IV (UAM-IV) (System Applications International SAI, 1990). The procedures used in the UAM-AERO model to simulate gas-phase chemical reactions and dry deposition of gases are similar to those in the UAM-IV with the following modifications: i) Gas-phase chemistry is replaced by a more flexible interface that allows the chemical mechanism to be easily changed. For this simulation UAM-AERO was used with the SAPRC90 chemical mechanism ii) Integration of gas-phase kinetics is done by using the high accuracy LSODE solver iii) The gas-phase chemical kinetic rate expressions are calculated from 3-dimensional temperature and water vapour concentration fields, and, iv) dry deposition is calculated using an algorithm based on Wesely (1989). The model also predicts ambient concentrations of both primary and secondary PM on an hourly basis. UAM-AERO uses ISORROPIA (Nenes *et al.*, 1998; 1999) for the aerosol thermodynamic calculations, which has been chosen for its computational efficiency and comprehensive treatment of the aerosols. The physical processes considered are: advection, turbulent diffusion, condensation and evaporation coagulation, emissions, nucleation and deposition. For this simulation UAM-AERO was used with the SAPRC90 chemical mechanism. The SAPRC90 chemical mechanism is comprised of fifty chemical species, solar radiation and one hundred thirty chemical reactions.

## 4. DESCRIPTION OF THE AIR POLLUTION EPISODE USED

The size of the modelling domain of the wider region of Attica was selected to include the urban region of Athens, with the major part of anthropogenic emissions, as well as extensive surrounding rural region with important volumes of biogenic emissions. It is extended from 93 km west to 91 km east, and from 91 km south to 93 km north of Athens, covering a total area of 33856 $km^2$. The domain on which the model was applied is a 92x92 grid system, with horizontal grid increments of 2 km in both directions, covering a total area of 33856 $km^2$. The center of the domain was the National Observatory of Athens (37º, 58'; 23º, 43'). In the vertical, 10 layers of variable thickness have been used, up to 5000 m above ground level, 6 under and 4 above the diffusion break. 56% of the region that was modelled is sea, 14% forestal areas, 16% rural areas, and the rest contains urban areas. The domain is chosen in such way that the boundary concentrations can be assumed to be background concentrations, compared to the concentrations in the basin. Data from the literature has been used as initial conditions. Moreover, gridded hourly anthropogenic emissions have been estimated, based on traffic information and the industrial activities in the domain. The photochemical models were used for the four-day episode in the Greater Athens Area that started at 0000 (LST) of June 22, 2003 until 0000 (LST) of June 26, 2003. The results of the two first days were not taken into consideration, as an adaptation time for the models, so that faults from initial conditions can be eliminated.

The period of June 24 and 25, 2003 was characterized by favourable conditions for the accumulation of primary and secondary pollutants over the Athens basin: weak synoptic wind, see-breeze development and stable temperature stratification. During this episode, the synoptic flow during the night and until sunrise was from NW in the lower layers with no significant changes aloft. Only after 1100 (LST), a light (for this season) sea breeze of S to SW direction (2-4$ms^{-1}$) started to develop with three main systems in the Attica peninsula:

over the eastern Mesogea plain, in the Athens central basin and over the western Thriassion plain. There appears to be a balance between the sea breezes that meet at the convergence zones. Thus the flow out of the Athens basin through the mountains gaps is blocked and this increases the air pollution levels in the city. Wind divergence phenomena also occurred over Peloponnese, as indicated by. The atmosphere over the GAA was very stable, and the vertical mixing height, ranged between 150 to 200 m during the afternoon and 60 to 100 m during the night. A detailed description of the episode has been presented elsewhere (Sotiropoulou *et al.*, 2006).

## 5. RESULTS AND DISCUSSION

The predicted by the PMCAMx model hourly concentrations of $O_3$, $NO_2$ and NO during the 24$^{th}$ of June, 2003 are compared with the measured concentrations from the stations and with the equivalent predicted concentrations of the UAM-AERO model as well. Measured and predicted (PMCAMx and UAM-AERO models), time series concentrations of $O_3$, $NO_2$ and NO at three monitoring stations are presented in Figure 1. Figure 1 shows the concentrations when the full emission inventory, which includes both anthropogenic and non-anthropogenic sources, is used. Peak ozone concentrations receive special attention, as they are important from a regulatory and public health point of view.

As shown in Figure 1, PMCAMx simulated the ozone diurnal pattern better than UAM-AERO. The low values of ozone during the night, when no photochemical reactions take place, are clearly depicted. UAM-AERO on the other hand predicts high ozone concentrations throughout the night. The maximum daytime concentration of $O_3$ is overpredicted in one station (Patision) and is lower in the others. At all sites there is a delay in the appearance of the maximum by both models, caused either by inaccuracies in the emission inventory or by the radiation factor provided by the meteorological model. It has to be emphasized that the maximum daytime concentrations in the UAM-AERO model are much higher than PMCAMx and way above the measured ones. It has also to be underlined that the UAM-AERO model simulates its maximum daytime concentrations in the evening hours. On the contrary PMCAMx simulates its maximum daytime concentrations around noon, which is much closer to the time the peak ozone concentrations were observed. The sharp peaks of the observed concentrations are very difficult to be predicted by models of this type, as the estimated concentrations are the average concentrations in each grid while the observed concentrations are at the location of the station (Sotiropoulou *et al.*, 2004).

PMCAMx shows signs of superiority in terms of its ability to predict $NO_2$ and NO concentrations. Its predictions are much closer to the observations for all three stations, which may indicate that PMCAMx has not the reported weakness UAM had in predicting $NO_2$ and NO concentrations (Lurman *et al.*, 1997). The Patision prediction is characteristic. PMCAMx predicts a peak of around 400 μg m$^{-3}$ for $NO_2$, while UAM-AERO predicted only 95 μg m$^{-3}$, with observed value of around 500 μg m$^{-3}$.

The predicted spatial distribution of the concentrations of ozone and secondary anthropogenic PM10, $SO_4^{=}$, $NO_3^{-}$, $NH_4^{+}$, OC at 1500 LST, as well as their increase when biogenic emissions are considered, are presented in Figures 2(a), 3(a) and 2(b), 3(b), respectively.

Figures 2 (a) and (b) show the spatial distributions of the predicted by PMCAMx ozone, PM10, OC and $SO_4^{=}$ concentrations at 1500 LST. The spatial distribution of ozone reveals higher concentrations in the suburbs, north of the Athens city-center. In particular, Figure 2 shows that ozone gathers in great amounts between the mountains Penteli and Hymettus, because of the existing winds, and that has a great effect in the areas of Marousi and Kifissia at the same time. Aerosols seem to be higher at the north region of GAA, because of the wind direction. The spatial distribution of the secondary anthropogenic OC, the only organic aerosol constituent in our model, indicates the high emissions of its precursors, such as alkanes, alkenes and aromatics, at that part of the domain. Figures 3 (a) and (b) show the PMCAMx spatial distribution of $NO_3^{-}$ and $NH_4^{+}$ at the same time. The nitrate distribution depicts two maxima, one at the centre of the city of Athens and the other southeast of the first. The first peak is due to the $HNO_3$ produced from NOx emitted from cars and other vehicles in downtown Athens, and a second peak is due to the $HNO_3$ produced from NOx of

industrial origin. The profile of the spatial distribution of $NH_4^-$ concentrations is a combination of the spatial distributions of $SO_4^=$ and $NO_3^-$.

Comparison between Figures 2(a), 3(a) and 2(b), 3(b) reveals the importance of biogenic emissions. As it is shown in these figures, the contribution of biogenic emissions to total ozone is more important in the suburbs, and dominates in the agricultural areas surrounding the GAA. This increase is caused by the interaction of biogenic VOCs and NOx. One can also observe in these figures a slight reduction of the maximum daytime concentrations of ozone at the centre of Athens. Ozone is formed through complex reactions involving sunlight, nitrogen oxides (NOx), and volatile organic compounds (VOCs). The concentration of $O_3$ in a given area depends on many factors including temperature, meteorology, and the presence of precursors. Due to this complex chemistry, increases in either NOx or VOCs could potentially result in decreased ozone levels, depending on the concentrations of precursors and the region's ozone isopleth plot (Seinfeld and Pandis, 1998; Bell and Ellis, 2004). Therefore, the response to reductions in the VOC's emissions is remarkably non- linear and any resultant reduction in the level of VOC's may potentially be accompanied by an increase in the concentration of ozone (Clapp and Jenkin, 2001; Mazzeo *et al.*, 2005).

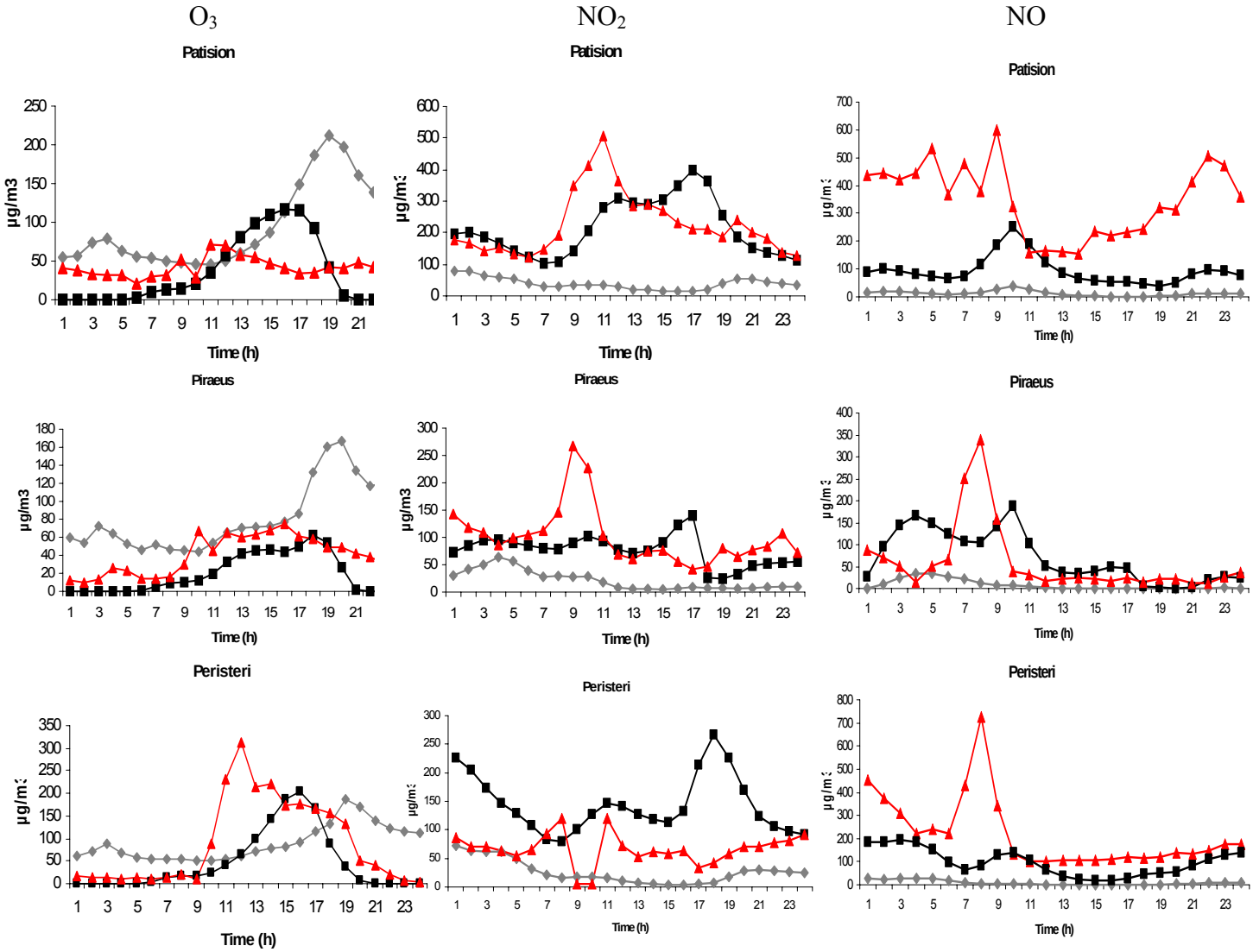


*Figure 1*. Predicted by PMCAMx (-■-) – UAM-AERO (-♦-) and observed (-▲-) time series concentrations of $O_3$, $NO_2$ and NO, during the 24$^{th}$ of June, 2003, for three monitoring stations in the GAA taking into consideration both biogenic and anthropogenic emissions

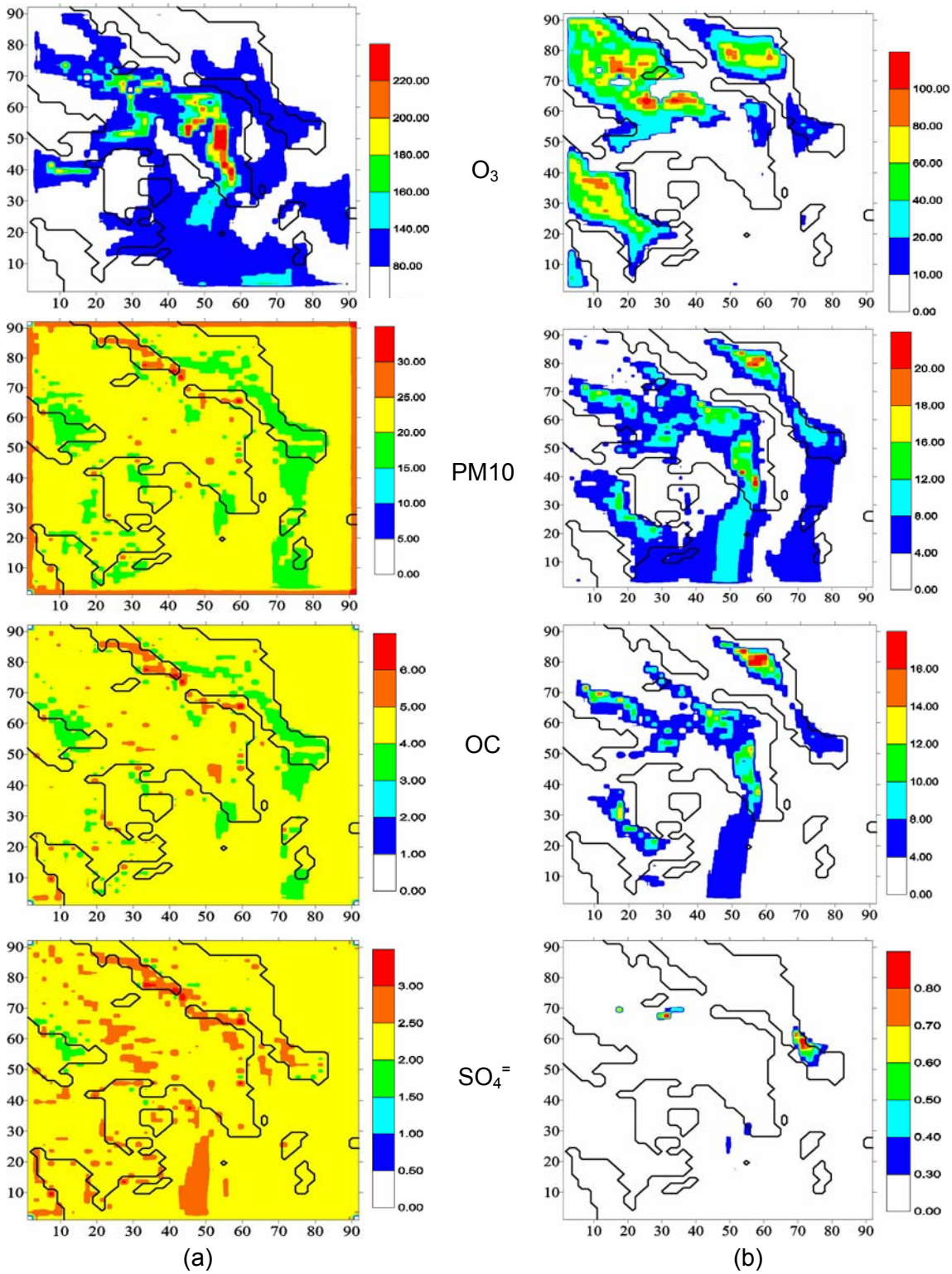


*Figure 2.* Predicted by PMCAMx spatial distribution for the episode of the 24$^{th}$ June 2003 at 1500 LST (μg m$^{-3}$) **(a)** when biogenic emissions are not included, **(b)** the increase when biogenic emissions are considered

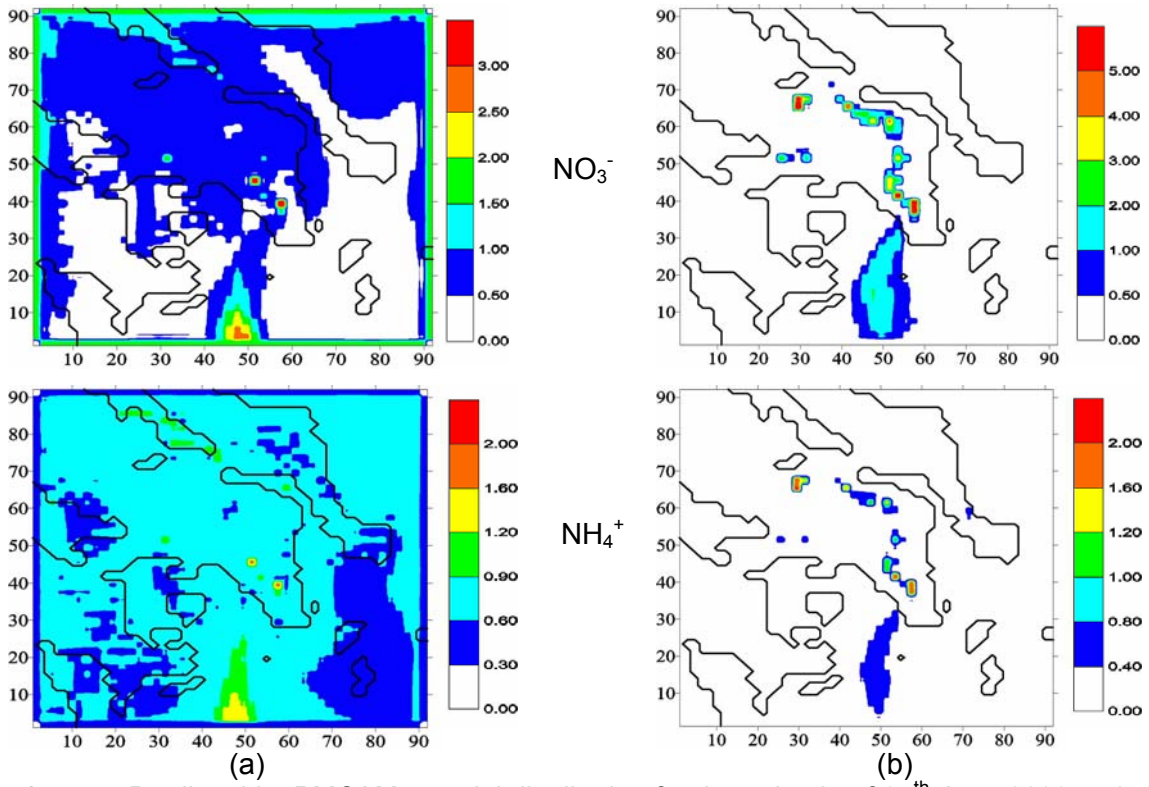


*Figure 3.* Predicted by PMCAMx spatial distribution for the episode of 24th June 2003 at 1500 LST (μg $m^{-3}$) **(a)** when biogenic emissions are not included, **(b)** the increase when biogenic emissions are considered

The increased concentration of OC is due to the production of condensable organic compounds from the reaction of terpenes with $O_3$, OH and $NO_3^-$. Moreover, the ozonolysis of terpenes is a significant source of OH radicals in the troposphere (Pfeiffer *et al.*, 1998). From the ozonolysis of biogenic compounds the produced OH reacts with $SO_2$ and NOx, leading to the formation of inorganic aerosol species.

The spatial distribution of the increase of both the ozone and aerosol concentrations, as a result of the inclusion of biogenic emissions, indicates that, not only the concentrations of OC are affected, but also the concentrations of both ozone and inorganic aerosol species. A moderate increase is predicted in the concentrations of ozone and aerosol species in the city of Athens and its suburbs, while a higher increase is predicted in the rural areas where the biogenic emissions are placed. Due to the calm winds, biogenic gasses remain in the rural areas and react with ozone.

In order to examine how ozone, $NO_2$ and secondary PM concentrations respond to changes in anthropogenic VOC emissions in the GAA we repeated the PMCAMx simulations with 25% (scenario 1) and 50% (scenario 2) reduction of anthropogenic VOCs.

*Table 1.* Reduction of the predicted maximum daytime concentrations of ozone, $NO_2$ and PM10 for the two emission scenarios

| | $O_3$ | | $NO_2$ | | Secondary PM10 | |
|---|---|---|---|---|---|---|
| | **-25% VOC's** | **-50% VOC's** | **-25% VOC's** | **-50% VOC's** | **-25% VOC's** | **-50% VOC's** |
| **Patision** | -57.3% | -81.1% | -5.6% | -21.1% | -14.3% | -20.7% |
| **Piraeus** | -7.1% | -13.6% | -2.8% | -5.8% | -4.5% | -6.4% |
| **Geoponiki** | -36.6% | -55.9% | -1.7% | -12.9% | -11.9% | -19.4% |
| **Peristeri** | -29.2% | -52.7% | -0.8% | -10.6% | -9.6% | -17.1% |
| **Marousi** | -71.0% | -87.9% | -10.4% | -27.8% | -19.2% | -29.5% |

Table 1 shows, that the decrease of the VOC emissions during the first scenario, leads to a significant reduction of the predicted daytime maximum concentrations of ozone in four out of the five locations. The reduction varies from 29 to 71%. The only station that has a small negative change is Piraeus, with only 7.1%. These changes appear to be greater when the VOC emissions are decreased by 50%. In that case the rate of reduction varies from 52 to 88%, for the same four stations, but Piraeus continues to have the smallest change. The reduction of VOC's has only a small effect to $NO_2$ concentrations. During the reduction of VOC's by 25%, the $NO_2$ concentrations show a reduction from 0.8 to 10.4%, while the reduction varies from 6 to 28% for the second scenario. The formation of ozone is chemically linked to the emissions of nitrogen oxides (NOx) through oxidation of gaseous precursors such as nitrogen dioxide ($NO_2$) and volatile organic compounds (VOC). This chemical interdependence is highly complex and gives rise to non-linear and coupled pollutant formation processes, as presented in the form of isopleth plots (Stein *et al.*, 2005). According to these plots there are NOx-sensitive and VOC-sensitive regimes (Sillman, 1999; Stein *et al.*, 2005). Factors that affect the split into NOx-sensitive and VOC-sensitive chemistry include VOC/NOx ratios, VOC reactivity, biogenic hydrocarbons, photochemical aging, and rates of meteorological dispersion. The results presented in Table 1 indicate that different parts of the Greater Athens Area reside in different regimes of these isopleths. As a result, to investigate the control measures, necessary to improve air quality one has to repeat model calculations with different pairs of VOC/NOx to construct local isopleths for typical episodes, and then extract conclusions on what is the right policy for the whole area.

The coupling of aerosols and gaseous species like ozone are also complex and highly nonlinear (Meng *et al.*, 1997). A reduction is predicted in both scenarios (25% and 50% VOC's reduction) for secondary $PM_{10}$. Table 1 shows that the reduction varies from 5 to 19% for the first scenario and from 6 to 30% for the second. The Piraeus station still has the smallest negative change in both scenarios. Secondary $PM_{10}$ seem to be little less sensitive than ozone to reduction of VOC's. Secondary inorganic PM is formed in the atmosphere by reactions of precursor gases such as nitrogen oxides (NOx), sulphur oxides (SOx) and ammonia ($NH_3$) (Seinfeld and Pandis, 1996; de Leeuw, 2002; Foresman *et al.*, 2003). NOx is a precursor to the formation of particulate nitrate, SOx is a precursor to particulate sulphate, $NH_3$ is a precursor to particulate ammonium, and VOC's are precursors to particulate secondary OC. So, any significant reduction of VOC's and NOx leads to a reduction of ozone and secondary $PM_{10}$. However, to understand the role of NO*x* and VOC's in aerosol formation, detailed isopleths are necessary to gauge the sensitivity of aerosol concentrations upon NO*x* and VOC's emission levels. The correlation between $PM_{10}$ and VOC's and NOx is nonlinear (Nguyen and Dabdub, 2002), which is consistent with Table 1.

## 6. CONCLUSIONS

Two models, PMCAMx and UAM-AERO, were applied in the Greater Athens Area, Greece, as tools to simulate a recent air pollution episode. To assure a fair comparison, the models were applied on the same domain and grid size, using the same emission inventory, diagnostic meteorology, initial and boundary conditions. Comparison of the results reveals that PMCAMx simulated the ozone diurnal pattern better than UAM-AERO, as the predicted by UAM-AERO maximum daytime concentrations were much higher than those of PMCAMx and way above the measured ones. Moreover the UAM-AERO model simulated its maximum daytime concentrations in the evening hours. On the contrary PMCAMx simulated its maximum daytime concentrations around noon, which is much closer to the time the peak ozone concentrations were observed. PMCAMx also shows signs of superiority in terms of its ability to predict the $NO_2$ and NO concentrations. Its predictions are much closer to the observations for all stations, which may indicate that PMCAMx has not the reported weakness UAM-AERO had in predicting $NO_2$ and NO concentrations.

Two simulations were performed, the first with no biogenic emissions and the second with biogenic emissions, in order to present the importance of biogenic emissions in the atmospheric chemistry of the region. Comparison of the results of those two simulations reveals the important role biogenic emissions play the in the formation of $O_3$, NOx and aerosol species. The simulations show that a fraction of the observed ozone concentrations in the

suburbs is caused by the interaction of biogenic VOCs and NOx. This “biogenic” ozone dominates in the agricultural regimes of the Greater Athens Area. However as ozone is formed through complex reactions, decreases in either NOx or VOCs could potentially result in decreased ozone and $NO_2$ levels, depending on the concentrations of precursors and the region’s ozone isopleth plot. The spatial distribution of the increase of ozone and aerosol concentrations, as a result of the inclusion of biogenic emissions, indicates that, not only the concentrations of organic carbon are affected, but also the concentrations of both ozone and inorganic aerosol species. However, despite the role biogenic emissions play, they should not be considered responsible for atmospheric pollution episodes. Anthropogenic emissions are the main causes of high levels of atmospheric pollution in urban areas. Biogenic emissions, though, should be taken under consideration when models for the prediction, as well as for the enforcement of abatement strategies of atmospheric pollution are applied, even if biogenic sources are placed mostly in the countryside. Taking into consideration the shown importance of species of biogenic origin, further research is necessary, in order to improve the biogenic chemistry that is used by the models, as well as to refine biogenic emission inventories.

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