MODELING OF COMBINED AEROSOL AND PHOTOOXIDANT PROCESSES IN THE MEDITERRANEAN AREA

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Abstract. The combined UAM-AERO/RAMS modeling system has been applied to study the dynamics of photochemical gaseous species and particulate matter processes in the eastern Mediterranean area between the Greek mainland and the island of Crete. In particular, the modeling system is applied to simulate atmospheric conditions for two periods, i.e., 13–16 July 2000 and 26–30 July 2000. The spatial and temporal distributions of both gaseous and particulate matter pollutants have been extensively studied together with the identification of major emission sources in the area. New pre-processors were developed for the UAM-AERO model for evaluating detailed emission inventories for biogenic compounds, resuspended dust and sea salt. Comparison of the modeling results with measured data was performed and satisfactory agreement was found for a number of gaseous species. However, the model underestimates the PM_{10} measured concentrations during summer. This is mainly due to the considerable underestimation of particulate matter emissions and in particular dust resuspension, the effect of forest fire emissions and the contribution of Saharan dust episodes.

Keywords: Mediterranean aerosol, mesoscale modeling

1. Introduction

Long-range transport and fate of photochemical gaseous air pollutants and particulate matter (PM) has been studied extensively in Europe the last decades, under the framework of several national and international efforts (EU, 1996, 1997; Berdowski *et al.*, 1998; EMEP-WHO, 1997; Beck *et al.*, 1999). A number of modeling studies for combined aerosol and gaseous species with Eulerian regional and mesoscale models have been also performed during the last several years (Binkowski and Shankar, 1995; Lurmann *et al.*, 1997; Ackermann *et al.*, 1998; Pai *et al.*, 2000). However, there is scarse information concerning consistent modeling studies in mesoscale geographical areas in Europe to reveal the

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atmospheric composition/variability of ozone and PM (Ackermann *et al.*, 1998). Furthermore, available information on characteristics of photochemical pollutants/fine particles in the Southern Europe and their effects on air quality are also limited.

In the major part of recent studies, the photo-oxidants and PM have been studied separately, even though the fine fraction of the PM is directly controlled by the airborne concentrations of photo-oxidants and other gaseous pollutants (Seinfeld and Pandis, 1998; Pandis *et al.*, 1992). Therefore, a combined intensive modeling study and comparison with available experimental data for ozone/fine particles in the Mediterranean area should offer valuable information for characterizing their dynamics and association. Furthermore, the Mediterranean is characterized by specific natural aerosol load, namely sea spray and North African Desert dust. These natural particulate matter emissions are involved in heterogeneous reactions with anthropogenic gaseous pollutants and may modify the processes leading to gas to particle conversion.

Along with the experimental work in the SUB-AERO project (European Commission grant ENVK2-1999-00052), a detailed modeling study has been performed using the UAM-AERO mesoscale air quality model (Lurmann *et al.*, 1997) including state-of-the-art modules for photochemical oxidants and fine aerosols to study the transport/chemistry interactions in the eastern Mediterranean area. Meteorological input data were provided by the RAMS (Pielke *et al.*, 1992) prognostic meteorological model, whereas regional data on background concentrations were obtained both from the EMEP trajectory oxidant model (Simpson, 1995) and the NILU-CTM model (Flatøy *et al.*, 2000). The modeling domain includes part of the SE Greek mainland, the Central and Southern Aegean Sea area and the Crete island (see Figure 1).

The combined UAM-AERO/RAMS modeling system has been applied to two simulation periods, from 13 to 16 July 2000 and from 26 to 30 July 2000. Several modifications have been introduced in the UAM-AERO mesoscale model, including new preprocessors for biogenic and natural emissions and new routines for studying new particle formation. The emission inventories are based on EMEP data (EMEP-WMO, 1997), whereas more detailed inputs for biogenic emission, resuspended dust, sodium and chlorine were calculated using newly developed methodologies combined with the UAM-AERO model.

The present study therefore represents the first application of a modified UAM-AERO model combined with a prognostic meteorological model (RAMS) for the southern Mediterranean. A better characterization of particulate matter emission inventories and determination of the background and boundary concentrations of the particulate matter in the modeling domain is a necessary step for better quantification of the chemically resolved mass of PM. Emissions from wild forest fires during the summer months and Saharan dust also contribute to the aerosol mass. Unfortunately, there were no available appropriately quantified emission inventories for these natural effects to be included in the model runs.

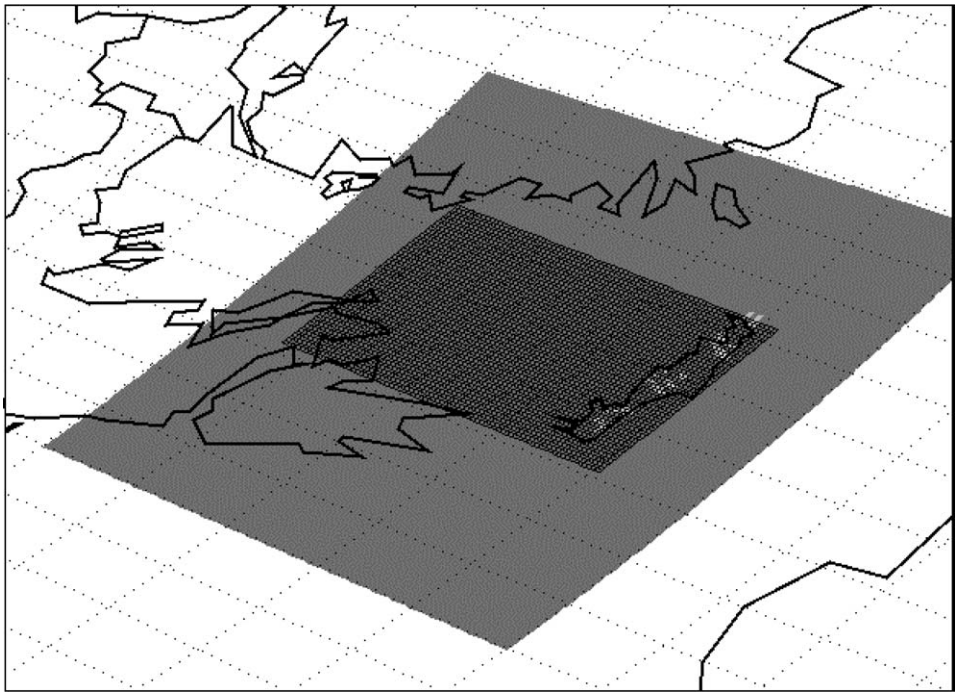


Figure 1. The UTM grid defined for the UAM-AERO model in the current modeling study.

Predicted aerosol and gaseous species concentrations patterns in the eastern Mediterranean area show the importance of the long-range transport component and the significance of biogenic and natural emissions sources. In agreement with previous studies (Lurmann *et al.*, 1997), it is observed that the model performance is sensitive to a number of parameters but mainly to particulate matter emission characteristics. The comparison with the experimental data appears satisfactory for a number of gaseous compounds. Poor agreement for a number of particulate matter chemical components reflects the uncertainty in the emission inventory data.

2. Methods – Results

2.1. Simulation Period

Two periods have been analyzed during the summer of 2000, as described above. Extensive meteorological inputs were developed for this period using the RAMS prognostic meteorological model. Detailed emission inventories on a 5 km $\times$ 5 km resolution grid were obtained using the EMEP emission data. Biogenic emissions for terpenes, isoprene, dust and sea salt were also calculated for a 5 km $\times$ 5 km

resolution grid. The first day of each simulation period is serving for the initialization of background concentrations of gases and aerosol species. Accordingly, the runs were initiated 24 h before the beginning of each actual simulation period. This is necessary for the simulations to be driven by emissions in the modeling domain and its boundaries rather than by initial conditions.

The combined UAM-AERO and RAMS modeling framework has been applied to model the summer campaign of the SUB-AERO experimental campaign. The UAM-AERO model grid (58 × 74 grid points) covers SE Greece (see Figure 1). Five vertical layers were used: two below and three above the diffusion break. The grid was defined in the UTM coordinate system. The UTM zone 34 was used for the definition of the grid above Greece, which means a central meridian of 21°E. The SW corner coordinates of the grid are the following (according to the WGS84 system) – Easting: 692089 and Northing: 3868229. This corresponds to the geographical coordinates: 34°56′08″ latitude and 23°06′02″ longitude.

The UAM-AERO allows the use of various alternative chemical mechanisms. The one employed for this case study is the Carbon Bond-IV (CB-IV), where species are lumped according to the type of their C–C bonds. A large number of reactions, involving 47 species are taken into account.

Two different background air quality data sets have also been used: EMEP and NILU-CTM (NILU – Chemistry Transport Model) data. The above initial concentrations, representing 3-D hourly values (in ppm) were also used background concentrations of the domain. The main purpose of using different background conditions was to check their effect on model predictions. Sensitivity analysis using the EMEP and NILU-CTM regional models will be presented in future work. In general terms, the results show similar spatial and temporal distributions for the gaseous and particulate matter pollutants but differences up to 20% in their concentration levels (e.g., O_3). In the results presented in the current paper, the NILU-CTM model predictions were used.

2.2. Meteorological and Air Quality Input Data

The meteorological inputs were obtained using the RAMS (Version 4.29) prognostic meteorological model (Walko and Tremback, 1996; Pielke *et al.*, 1992). RAMS combines a non-hydrostatic cloud model (Tripoli and Cotton, 1982) and a hydrostatic mesoscale model (Mahrer and Pielke, 1977). It was developed for simulating atmospheric phenomena with resolutions ranging from tens of kilometers to a few meters. There is no lower limit to the domain size or to the mesh cell size of the model finite difference grid. A general description of the model and its capabilities is given in Pielke *et al.* (1992).

RAMS initialization data include topography files, sea surface temperature data, vegetation files, and initial meteorological fields. The USGS topography data set of 30 arc-seconds (about 1 km) was used. From the above topography data set, the land-water percentage was extracted. The sea surface temperature data set consists

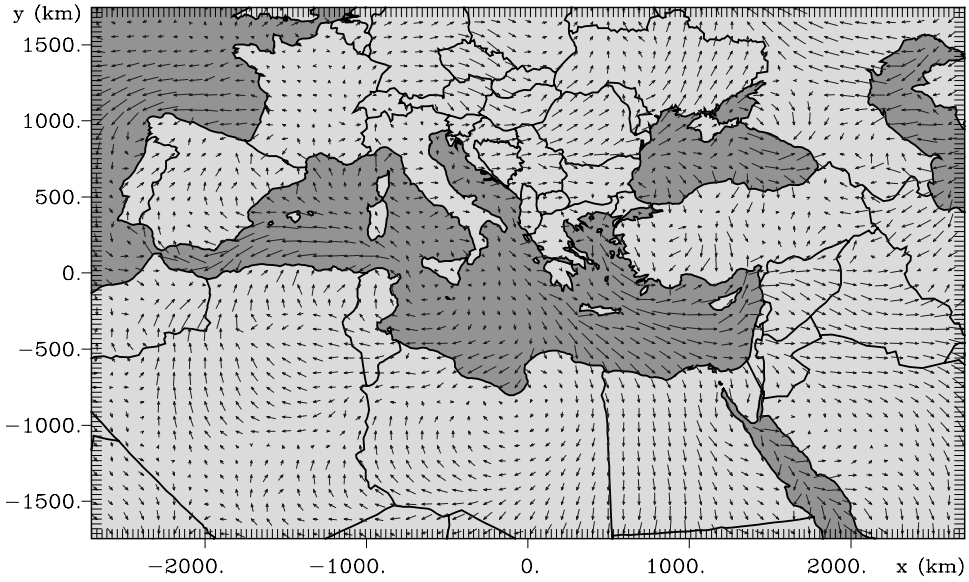
of mean climatological monthly values with a resolution of 1° (about 100 km). The vegetation data set was available in gridded form with a resolution of 30 arc-seconds (about 1 km) and global coverage. The vegetation data have been retrieved from the USGS. The 74 Olson World Ecosystem classes have been classified to 30 classes based on the Land Cover/Vegetation type from Biosphere–Atmosphere Transfer Scheme which is used by RAMS. The USGS dataset is based on 1 km Advanced Very High Resolution Radiometer data spanning April 1992 through March.

The model was initialized with gridded data sets prepared by the isentropic analysis package. The primary data sets were retrieved from the European Center for Medium Range Forecasting (ECMWF). Their horizontal increment was 0.5°, and they are available every 6 h (00:00, 06:00, 12:00 and 18:00 h UTC). In addition, surface meteorological data from the same source were used for the initialization of the model.

These sets contain the following fields: horizontal velocity components, temperature, geopotential height and relative humidity as a function of pressure. These initialization fields are used to supply a time series of observational data for the atmospheric model to assimilate during execution. The lateral boundary region of the coarser grid is nudged toward the initialization file values every 3600 s, while there is no relaxation time scale at the center of the domain.

The lateral boundary conditions on the outer grid followed the Klemp–Lilly condition which is a variant of the Orlanski condition, in which the gravity wave propagation speeds computed for each cell in the Orlanski condition are averaged vertically, with the single average value being applied over the entire vertical column. The horizontal diffusion coefficients were computed as the product of horizontal deformation rate and a length scale squared, based on the original Smagorinsky formulation. The vertical diffusion coefficients were computed according to the Mellor and Yamada parameterization scheme, which employs a prognostic turbulent kinetic energy. For both shortwave and longwave radiation parameterizations, the scheme described by Mahrer and Pielke (1977) has been used.

RAMS was initialized on the 1st of July; during this period, the synoptic conditions were characterized by the installation of a high-pressure system over the central and eastern Mediterranean and Northern Africa. The passage of relatively shallow disturbances over Southern Europe towards the Balkans and the Black Sea has as a result the strengthening of the pressure gradient over the NW Turkey and the Dardanelles Gap. As a result, a westerly flow is evident on 15th and 16th while during the following days, the Etesians were established with a persistence of about 7 days. The dissipation of the pressure gradient over NW Turkey resulted in the extension of the high pressures to the east. A weak northerly current is evident over the Central and SE Aegean while the development of mesoscale circulations is favored both at the continental and coastal areas of Greece and Turkey. Figures 2 and 3 show the simulated surface wind fields on 17 July 2000 at 12:00 h UTC. After the passage of the low pressure system, the northerly current of the Etesians is gradually established.

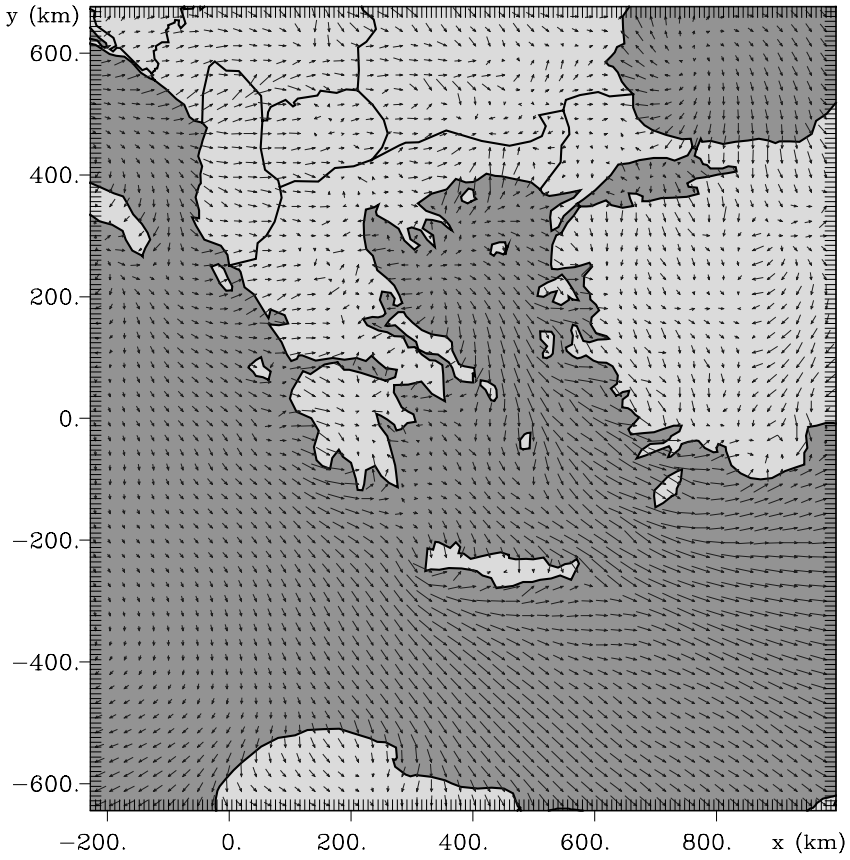


JULY 12 2000		Grid 1			
z = 43.7 m	07/17/00 1200 UTC 0 sec	min	max	inc	lab*
vectors	→ 10.0 m/s horiz 10.00 m/s vert				

Figure 2. Wind field at $z = 45$ m at grid (1), 12:00 h UTC, 17 July 2000. Wind arrows are plotted every second grid point.

The emissions inputs except for PM_{10} are from the EMEP database, the CTM-NILU regional model (background) (see Figure 4) and from detailed emission inventories for biogenic species (isoprene and terpenes), resuspended dust and sea salt. The background and initial concentrations for the 24 gaseous species considered in the model were obtained from the CTM-NILU regional model (Flatøy *et al.*, 2000). An example of the ozone background concentrations calculated with the CTM-NILU model is presented in Figure 4, where high ozone concentrations are calculated downwind of the Athens metropolitan area.

In order to include background values of PM_{10} a combination of measurements and calculated values from the EMEP (EMEP4/2001, 2001) were used. In addition, in the air quality data initial background values of 24 gaseous species were obtained for air quality data. Different concentrations for the initial conditions in all vertical layers of the atmosphere were used as specified by the CTM-NILU model (exept for PM_{10}). For the values at the boundaries of the domain (and also for the top of the domain), hourly boundary values of 24 species specified for all five layers were used. The same boundary concentrations were used during the whole period of modeling.



JULY 12 2000			Grid 2			
z = 43.7 m	07/17/00 1200 UTC 0 sec		min	max	inc	lab*
vectors	→	10.0 m/s horiz 10.00 m/s vert				

Figure 3. Wind field at $z = 45$ m at grid (2), 12:00 h UTC, 17 July 2000.

2.3. EMISSIONS

Spatially resolved emission rates of NO_x, SO_2, NMVOC, CO, NH_3 and PM_{10} on an hourly basis are required as input to the UAM-AERO model. A modified CB-IV chemical mechanism is used in the current application and the NMVOC emissions have been partitioned into the appropriate classes for this chemical mechanism. For sensitivity analysis, two separate runs were performed with/and without the ship traffic in the Mediterranean area. The relevant data were obtained from the EMEP (EMEP-WMO, 1997). The effect of the ship traffic is not important for the majority of the compounds but there is an increase of 10% in the ambient concentration of NO_x species in the areas close to the emissions (e.g., the island of Crete). A more

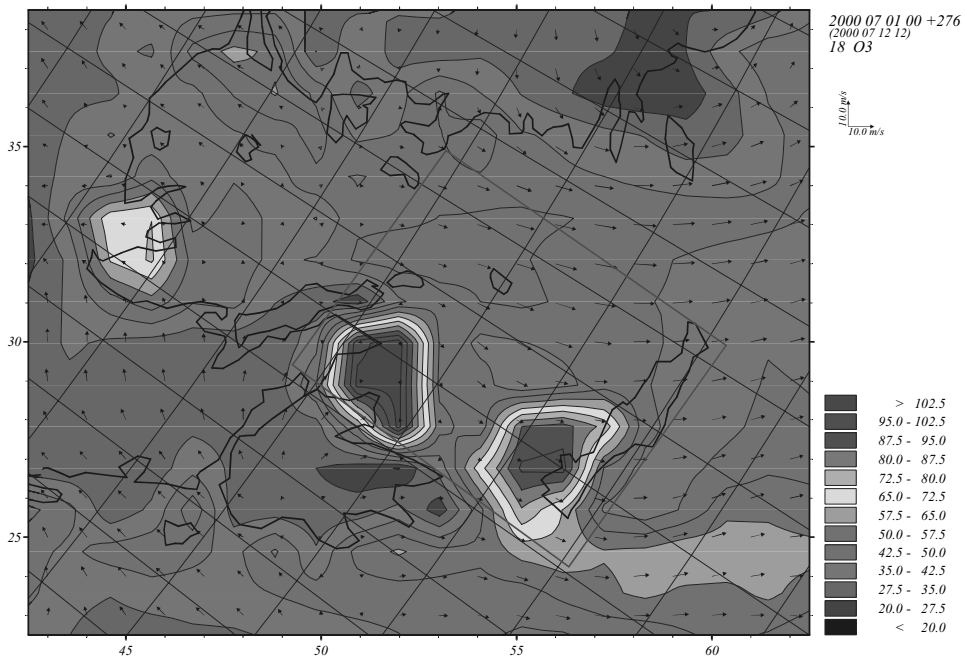


Figure 4. Simulation results of the ozone surface concentration for an extended grid using the CTM-NILU regional model (unit in ppb).

detailed analysis is beyond the scope of the current paper. In the current simulations, emission inventories from ship traffic in the Mediterranean area have been included into the modeling area emissions.

In relation to the dust emissions in the current simulations (see Figure 5), we have followed standard literature methods where the vegetation, land use, and wind speed determine the amount of soil dust that is resuspended (Andreae, 1995).

Sea salt aerosols play an important role in the atmospheric chemistry, and they influence the gas concentrations of several species such as HCl, H_2SO_4, $ClNO_2$, and NH_3 (Ericson *et al.*, 1999). In addition, the sea salt is important in the radiation balance (Seinfeld and Pandis, 1998). NaCl emissions were calculated as a function of wind speed and relative humidity mainly based on the work of Monahan *et al.* (1986) for particles smaller than 7 μm and the work of Smith *et al.* (1993) for particles greater than 7 μm, respectively. As shown in Figure 1, our modeling domain covers a large maritime area where the concentration of sea salt is significant. Emission of sea salt is calculated at all relevant grid cells, and hourly values of Na^+ and Cl^- emission rates are used as inputs to UAM-AERO.

The biosphere emits a large number of volatile organic compounds (VOCs) into the atmosphere, with isoprene and monoterpenes being the most abundant species (Singh and Zimmerman, 1992). These compounds are highly reactive in the troposphere and thus plays an important role in tropospheric chemistry.

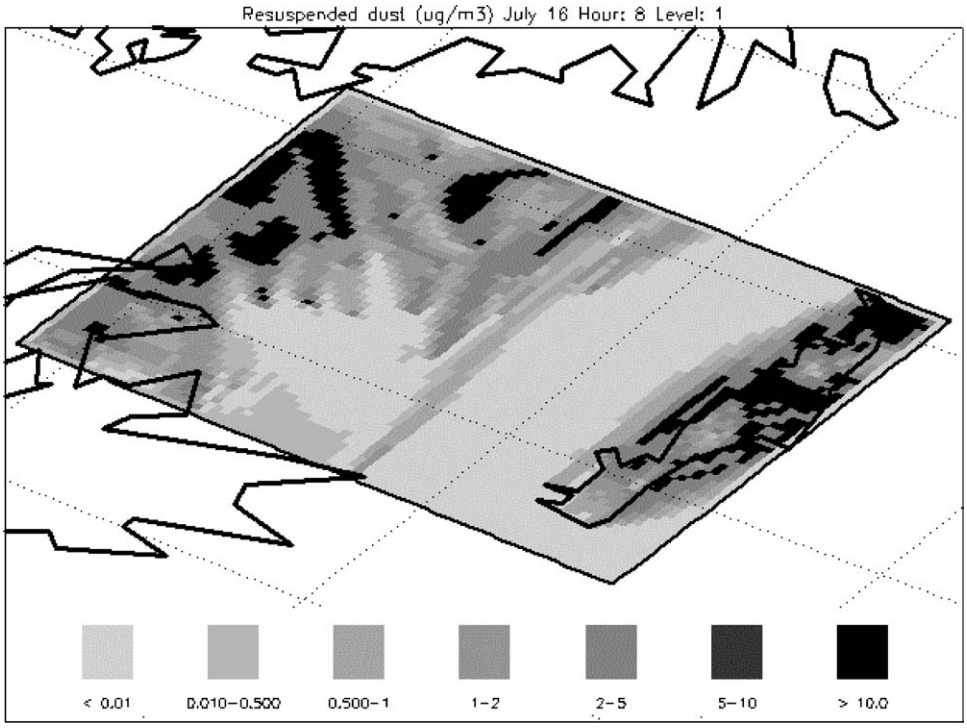


Figure 5. Surface spatial distribution of resuspended dust at 16 July 2000 at 08:00 h.

In the current modeling study, we have focused mainly on the emissions of isoprene and monoterpene (hereafter referred to as terpene) in Greece. In Figure 6 we present an average emission profile from Greece in July as calculated from the current approach. The E-94 correction factors for isoprene are obtained from the work of Simpson *et al.* (1995) and Guenther *et al.* (1993). The terpene correction factors are described by Pierce *et al.* (1990) and Simeonidis *et al.* (1999).

For the VOC emissions, hourly temperatures at each grid cell are used from the RAMS temperature predicitions, whereas the sunlight is calculated from a separate model (Thomas and Stamnes, 1999). The correction factor for sunlight dependence includes photosynthetically active radiation (PAR: 400–700 nm). In order to calculate the flux at different locations and times of the day we have used a Radiative Transfer Model (Thomas and Stamnes, 1999). The model is set to calculate flux F_λ [J m^{-2} s^{-1}] every 10th nanometer from 400 to 700 nm. We have assumed a clear sky (only a very thin layer of haze) and in ozone value of 290DU (the ozone value over Crete 19th of July 2000). The calculations are performed hourly from 1:00 to 24:00 h. Further, we have used a "midlatitude summer" atmospheric profile for our calculations. In order to find the PAR flux at all the grid cells, we have

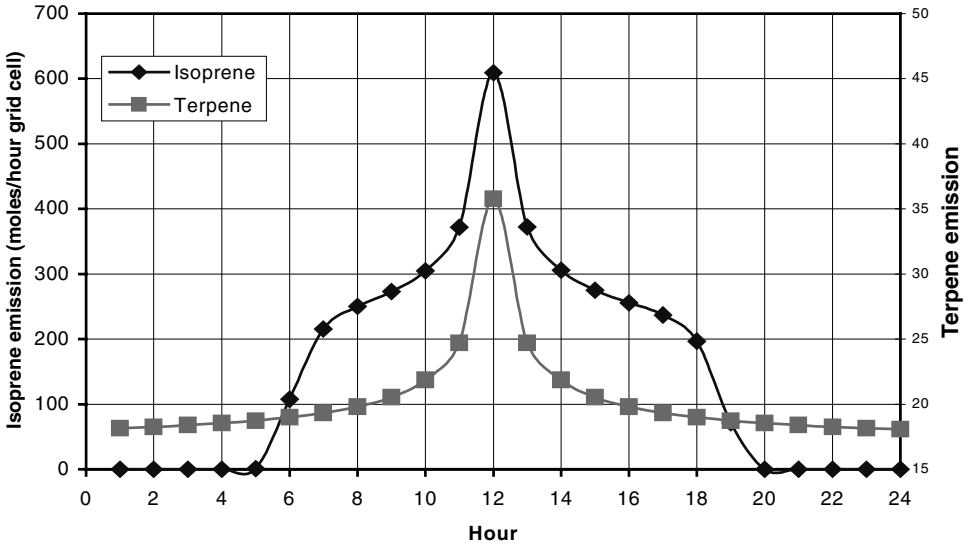


Figure 6. Emission of isoprene and terpene (moles per hour per grid cell).

made flux calculations at the four corners of the grid region (south-east, south-west, north-east, north-west) and used interpolation methods for finding flux at each grid cell.

2.4. Model Predictions

Much effort has been put in preparing input files and initializing the model. Hourly formations of several species are checked against measurements to ascertain that the model results are reasonable. Spatial patterns of predicted 1 h average O_3, NO_2, Na^+ and PM_{10} concentrations on July 30, 2000 are shown as examples in Figures 7–10. The predicted pattern of O_3 shows a band of high concentrations extending from the Athens metropolitan area to the middle of the Aegean sea and lower concentrations in a narrow area at the western part of the domain and above the island of Crete.

The NO_2 concentrations are also higher closer to the Athens metropolitan area with higher values at the northern part of the domain. The Na^+ concentrations are higher in areas with higher wind speed in the middle of our domain and south of Crete. The PM_{10} total mass distribution follows the patterns of O_3 close to Athens but also its spatial distribution encounters the production of sea salt and the regional contribution.

Examples from the obtained results for all simulation periods and scenarios are shown in Figures 11 and 12, where a comparison between modeled and measured results for O_3 is shown for the two periods. The measurements were performed

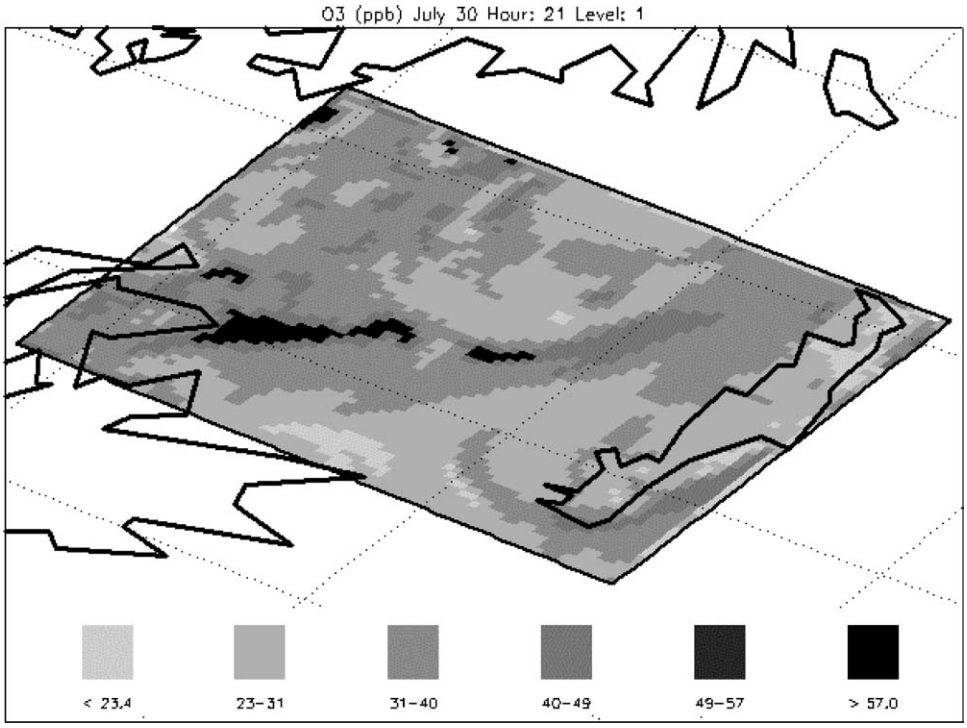


Figure 7. Surface spatial distribution of O_3 at 30 July 2000 at 21:00 h.

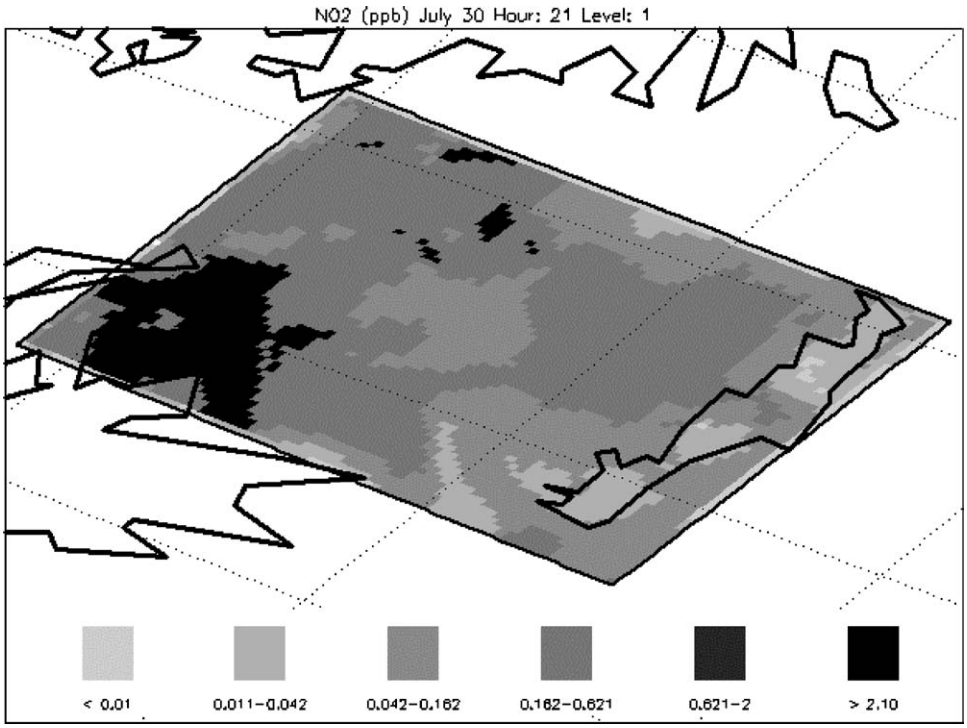


Figure 8. Surface spatial distribution of NO_2 at 30 July 2000 at 21:00 h.

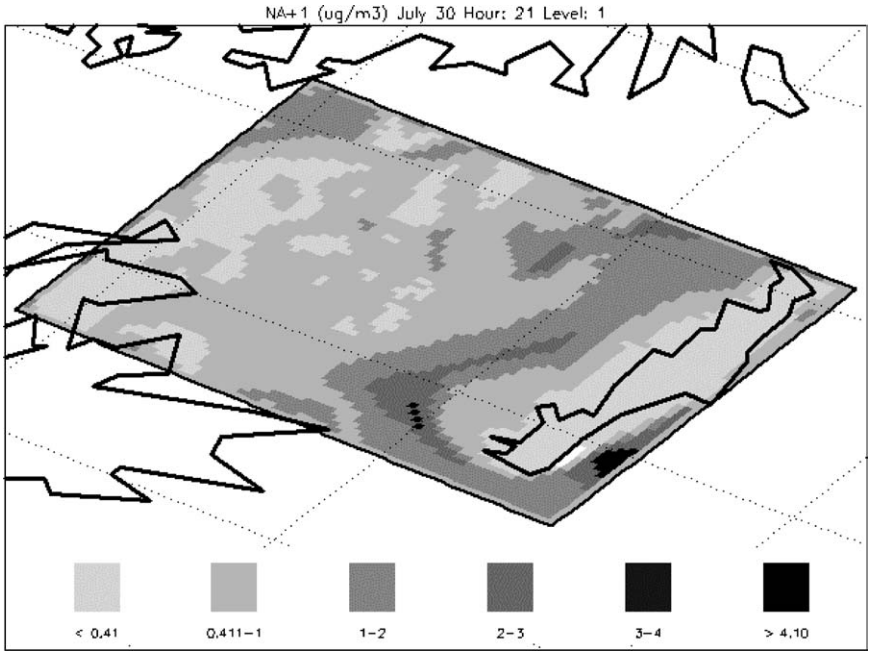


Figure 9. Surface spatial distribution of Na^+ at 30 July 2000 at 21:00 h.

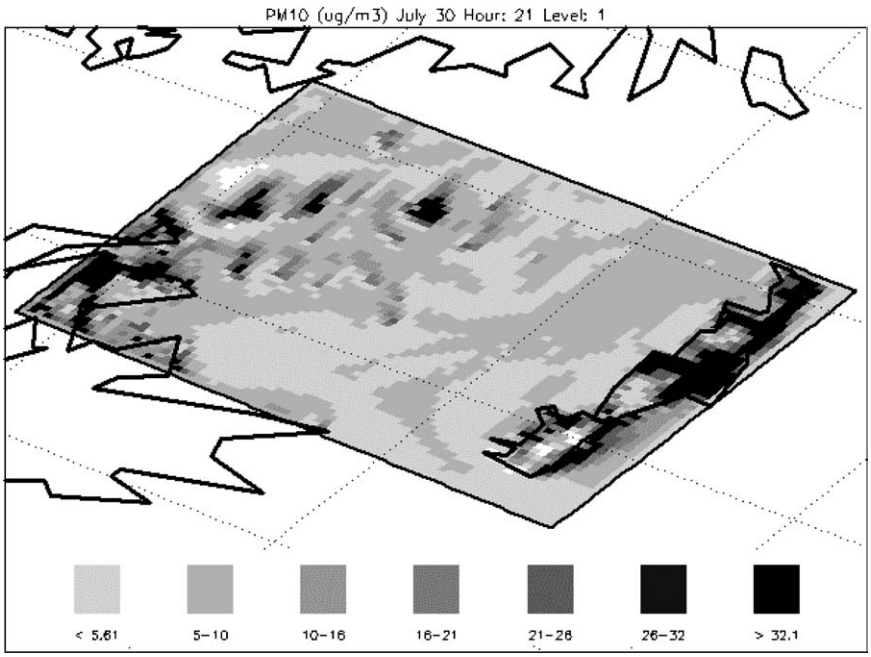


Figure 10. Surface spatial distribution of PM_{10} at 30 July 2000 at 21:00 h.

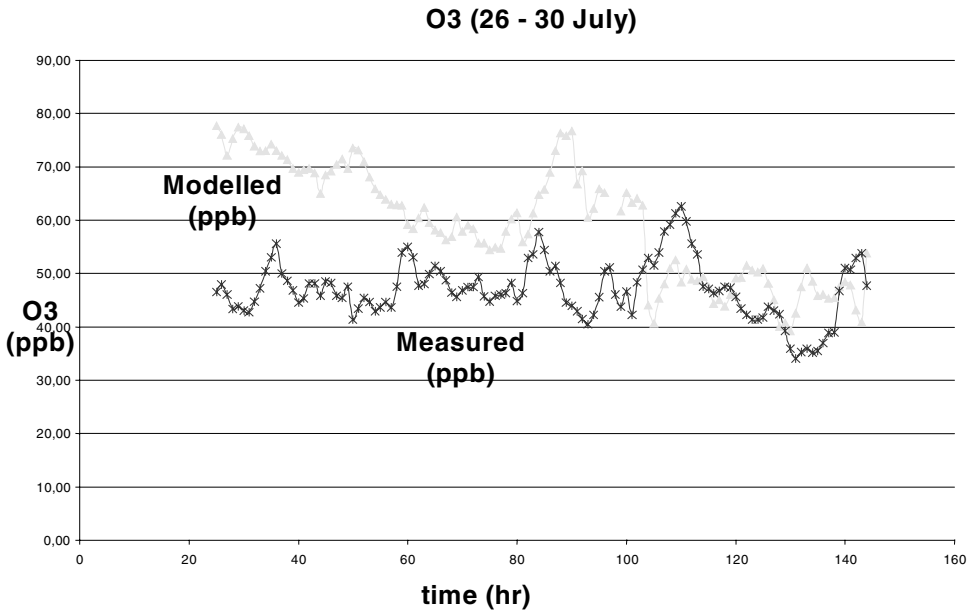


Figure 11. Comparison between modeled and measured ozone concentrations at the Finokalia station, Crete, Greece for the period 26–30 July 2000.

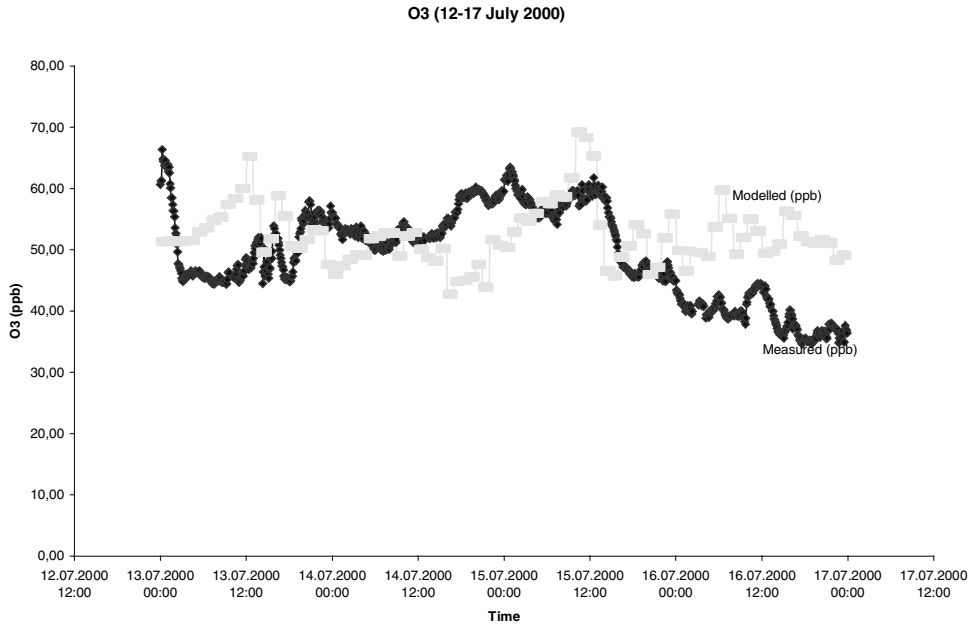
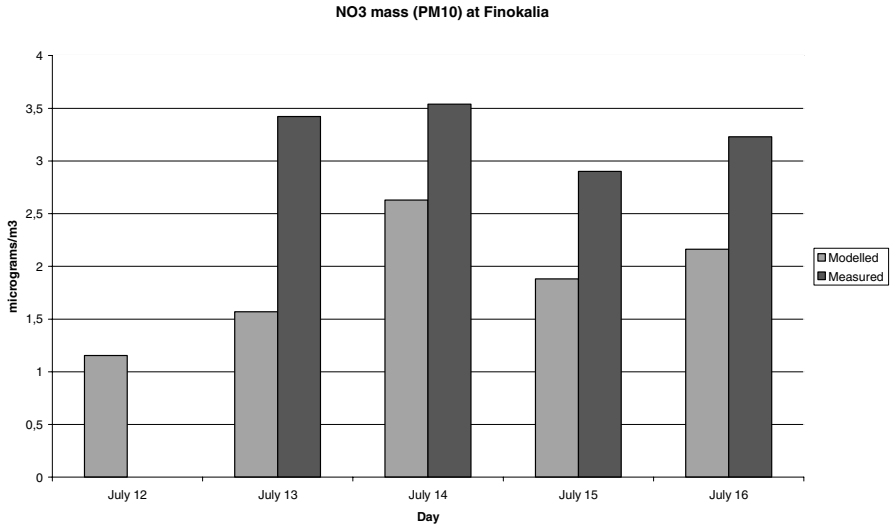
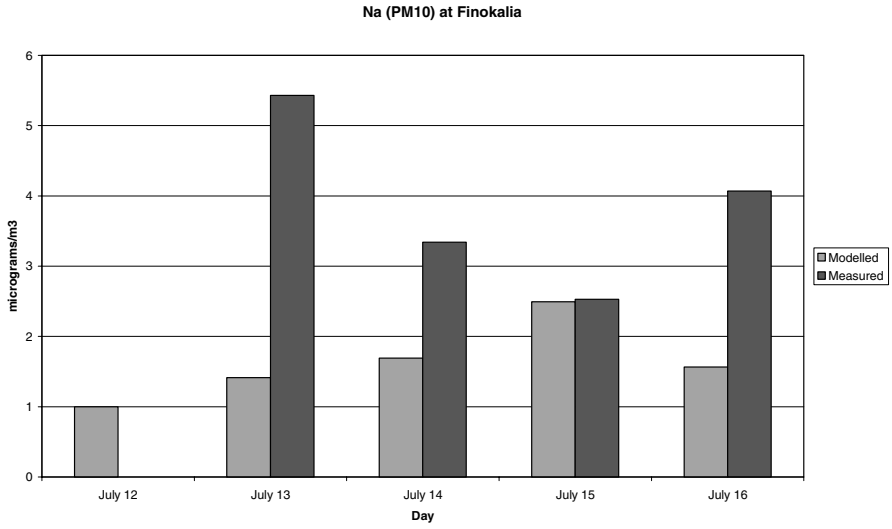


Figure 12. Comparison between modeled and measured ozone concentrations at the Finokalia station, Crete, Greece for the period 12–17 July 2000.

at the northern part of the island of Crete, at the Finokalia station. As can be seen, the comparison is satisfactory and the variation of the modeled results follow the same characteristics as the measured data. Also, the diurnal variation of the predicted concentrations has been found to be in good qualitative agreement with the



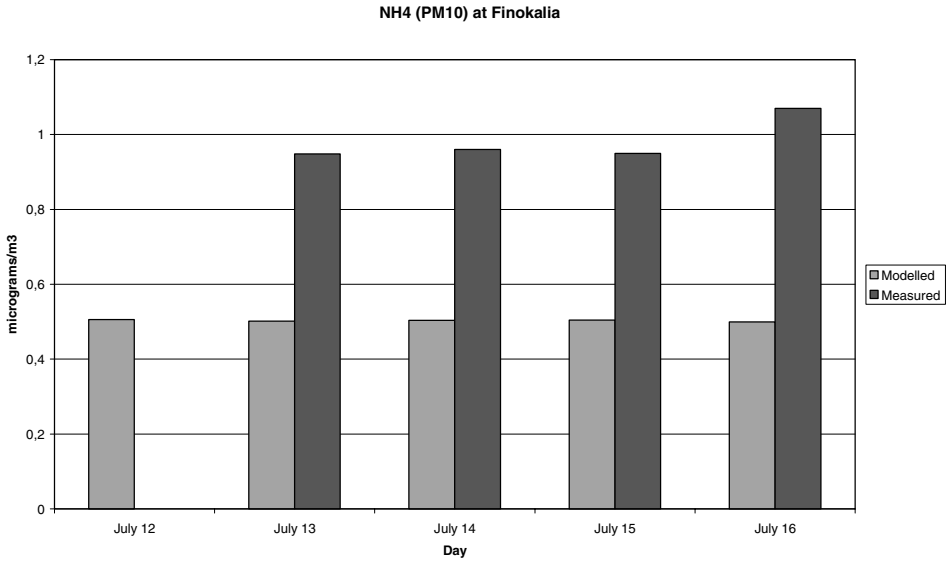
(a) NO_3



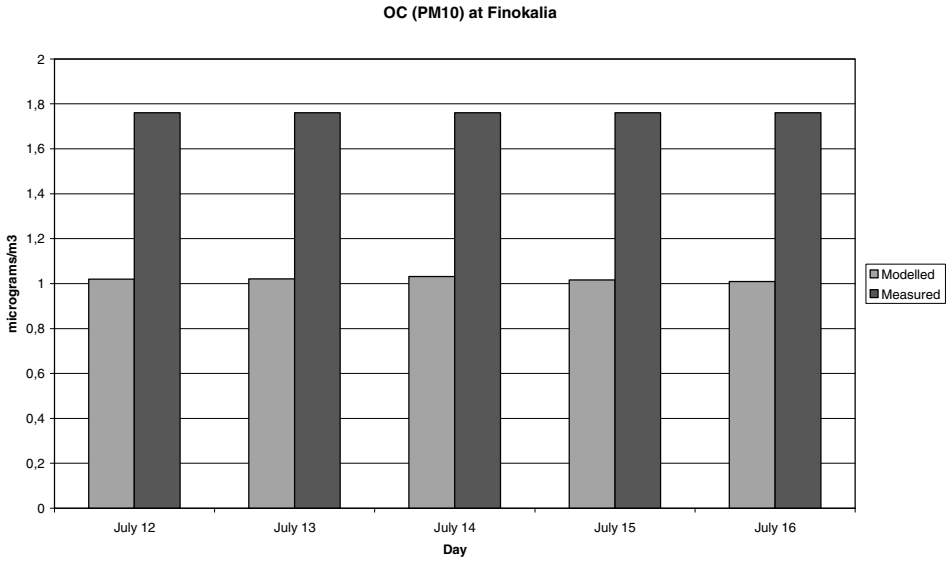
(b) Na

Figure 13. Comparison between modeled and measured PM_{10} chemical composition data (Finokalia, Crete, Greece): (a) NO_3 (b) Na (c) NH_4 (d) OC (e) Cl (f) SO_4 (g) soil dust.

(*Continued*)



(c) NH_4



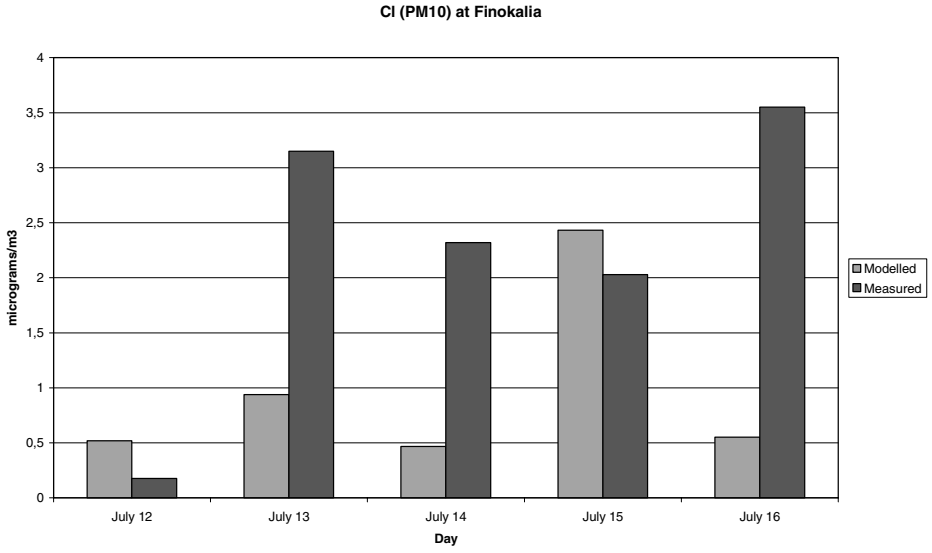
(d) OC

Figure 13. (*Continued on next page*)

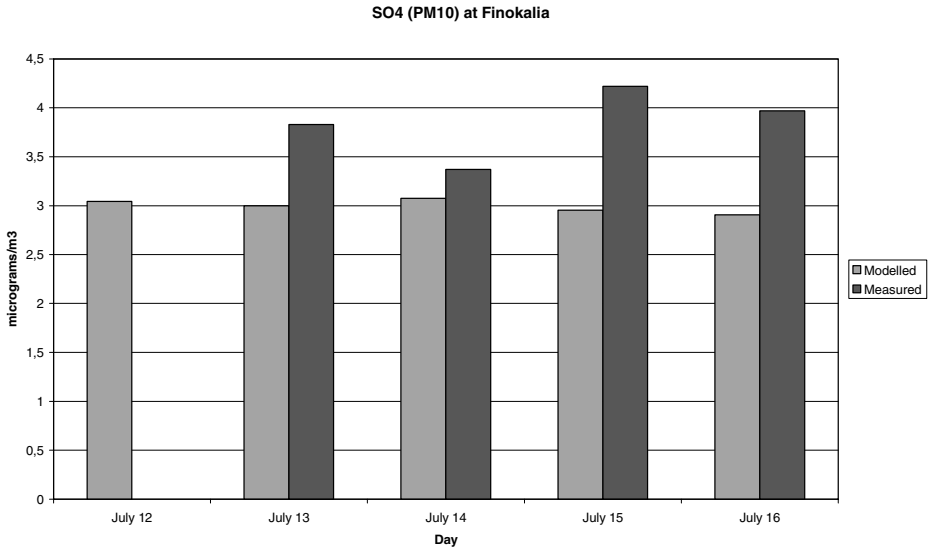
theoretical and measured results. A detailed comparison between modeled results and measured data will be performed in a future paper.

In Figure 13 comparisons between modeled and measured PM_{10} chemical resolved values are presented. The observed data were obtained from the Berner

impactor measurements and further analyzed with Ion Chromatography (IC), particle-induced X-ray emission (PIXE) and elementary carbon/organic carbon (EC/OC) speciation. Data for Saharan dust and elementary carbon are based on the measurements, only because in the UAM-AERO modeling runs are not included emission or background data for these components. The modeling results

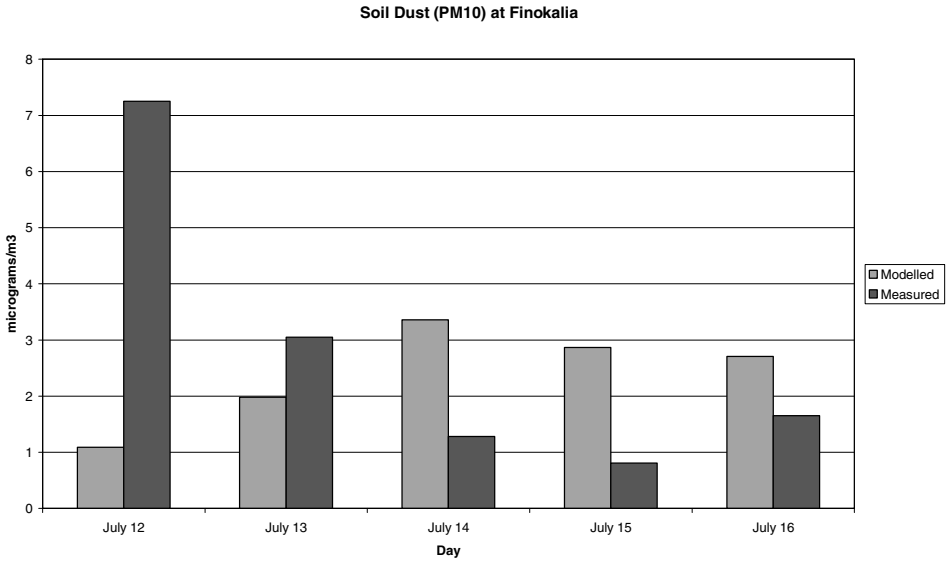


(e) Cl



(f) SO_4

Figure 13. (*Continued*)



(g) Soil Dust

Figure 13. (*Continued*)

underestimate in general the PM_{10} measured data. A large uncertainty remains in the size resolved emission inventories for particulate matter as well as detailed data on the regional component of the PM_{10}. The model results are in close agreement with the measurements for sulfates, the nitrate results are within 50% and the ammonium data are underpredicted. The comparison for sodium and chlorine is also within 50%. Future work will be based on winter simulations where the measured particulate matter concentration is considerably lower than the summer values. In general, the large deviations between modeling results and experimental data exist for those components where reliable emission inventory data do not exist. Sensitivity calculations for different emission scenarios, as performed in the work of Lurmann *et al.* (1998) will be presented in a future paper.

3. Conclusions

The combined UAM-AERO/RAMS modeling system is an efficient platform for simulating the transport/dynamics of particulate matter and photo-oxidant precursors. The UAM-AERO/RAMS modeling system has been successfully applied to simulate the dynamics of PM and photo-oxidants in the eastern Mediterranean area. Considerable effort has been invested in the model initialization and the determination of the emission inventories and the background concentrations of both gaseous and particulate matter species. Furthermore, a number of modifications

were introduced in the UAM-AERO model including new modules for sea salt production, terpene and isoprene emissions, soil dust resuspension and new particle formation.

The modeling studies with the combined UAM-AERO and RAMS system reveal the importance of photo-oxidant and fine aerosols dynamics in the Mediterranean area. Comparison of the modeling results with measured data is satisfactory but underestimation of the PM_{10} measured concentrations during summer was observed.

The simulation results show that the plume from Athens and other urban areas, as well as long-range transport contribute to the aerosol mass in the greater area of eastern Mediterranean. The results from the current simulations show the capabilities of the modeling platform used, which is able to characterize the dynamics in the Mediterranean area.

Discrepancies between modeled and measured data for the particulate matter are mainly due to soil dust emissions and Saharan dust. The measured values show that resuspension from soil is an important factor in determining aerosol size distribution during the summer. This fact, together with the considerable underestimation of resuspended dust from the modeling effort, demonstrates the need for chemical quantification of particulate matter emission inventories in Europe, as well as the need for better determination of the effect of Saharan dust, wild forest fires, and resuspended dust emissions in Southern Europe.

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