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MODELING OF PHOTOCHEMICAL POLLUTION IN ATHENS, GREECE. APPLICATION OF THE RAMS-CALGRID MODELING SYSTEM

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Abstract—The causes of the poor air quality in Athens, Greece during the severe episode of 25–26 May 1990 has been studied, using a prognostic model (RAMS) and a three-dimensional Eulerian air quality model (CALGRID). The modeling effort indicates that the main urban area of Athens exhibited high concentrations of nitrogen oxides, the main sources of which are automobiles, while the NNE suburban area exhibited high ozone concentrations, the product of photochemical activity of the primary pollutants that were transported by the sea-breeze. The application of the models also demonstrated the need for an accurate emission inventory for improved predictions of the pollutant concentrations. It was also found that a 50% reduction of the nitrogen oxide emissions will increase the ozone levels in the downtown area substantially.

Key word index: Photochemical modeling, ozone, mesoscale modeling, air pollution.

INTRODUCTION

The problem of air pollution in Athens has been of great concern for the last two decades, because the imposed state and Commission of the European Communities (CEC) limits on O_3 and NO_2 have been violated for a significant number of days, during all seasons every year (Kallos and Kassomenos, 1992; Kallos *et al*., 1993). Despite this fact, the air quality in Athens cannot be considered as the worst among other cities in Europe and the American continent. It is well known that the relatively high concentrations of air pollutants in the Greater Athens Area (GAA) are mainly due to a combination of factors, i.e. the physiographic characteristics of this region, the meteorological conditions and the sources of air pollutants (e.g. Lalas *et al*., 1982, 1983, 1987; Gusten *et al*., 1988; Katsoulis, 1988; Kallos *et al*., 1993).

The concentrations of air pollutants within the Athens Basin are affected by the weather conditions occurring in the region as they are modulated by the regional and local physiographic characteristics. The role of some mesoscale circulations in the GAA on the formation of air pollution episodes (violation of the state imposed limits) has been the subject of several studies (e.g. Lalas *et al*., 1982, 1983, 1987; Prezerakos, 1986; Flassak and Moussiopoulos, 1989; Kallos *et al*., 1993). Although, most of these studies have emphasized the role of the sea (land)-breeze mechanisms in the GAA, it has been found, recently, that this is not always the case (Kallos *et al*., 1993). In that study, the case-by-case analysis of the 80 worst air pollution episodes that occurred in Athens during the time period 1983–1990 showed that the worst air pollution episodes in Athens occurred during the days where the synoptic and local circulations in the Athens Basin were in critical balance and/or with warm advection in the lower troposphere.

In contrast to the extensive experimental effort, and the limited atmospheric modeling that has been done to isolate the weather patterns that lead to high pollutant concentrations, the air quality modeling effort that relates the pollutant levels to emissions of primary species is very limited (Moussiopoulos, 1990). As a consequence, the effects of the different state actions, e.g. restrictions of the traffic in the center of the city, the use of cars with catalytic converters etc., have not been evaluated. Besides there is the suspicion, that, while the traffic restriction reduces locally the generation of primary pollutants, it may result in higher ozone concentrations, because of the smaller doses of NO, a major sink of ozone in Athens (Lalas *et al*., 1987).

The purpose of our research program is to study in a systematic manner the major causes of the air quality degradation in Athens, using state of the art atmospheric and photochemical modeling. In this paper the first results of this modeling effort are presented and several characteristic features of the photochemical air pollution in Athens are explained.

DESCRIPTION OF THE AREA

The cities of Athens, Piraeus and their suburbs are located in an oblong basin surrounded by high mountains from three sides and open to the sea from the

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fourth (Fig. 1). The three mountains around Athens are Hymettus to the E, Pendeli to the N–NNE and Parnitha to the N–NNW with elevations up to 1400 m. To the W of the basin is mountain Aegaleo with its peak elevation at 450 m. The length of the basin is approximately 25 km (SSW to NNE direction) and its width is 17 km. The basin has a slope of less than 1% up to the prairies of the mountains which are steep. The mountains mentioned above are the physical barriers between the Athens Basin and the plains of Thriassion and Mesogea and the hilly area of Marathon as well. There are three main gaps between these mountains: the gap of Agia Paraskevi between Hymettus and Pendeli (to the NE of the basin), the gap of Malakassa between Pendeli and Parnitha (to the N of the basin) and the gap of Ano Liossia between Parnitha and Aegaleo (to the W of the basin). Another gap at the southern edge of Aegaleo is the Dafni which is very narrow (100–200 m). Near the southern edge of Aegaleo, at a distance of approximately 1.5 km is the island of Salamis, while at a distance of approximately 20 km to the S of Piraeus is the island of Aegina. These islands are within the Saronic Gulf which ends in NE Peloponnese to the SW and is open to the Aegean Sea to the SE.

There are three hills with heights up to 340 m within the Athens Basin. These are the Pnyka, Lykabettus and Tourkovounia hills. Their tops are approximately 150 m above the bottom of the basin. They are almost along the main axis of the basin and they separate it to a western and an eastern part. Almost the entire Athens Basin can be considered as urban area. Only 3% of the urban area is covered by vegetation. The population of the urban areas within the Athens Basin is approximately 3,600,000. The rapid expansion of the city during the last 30 years was not followed by the parallel development of the necessary infrastructure (road network, parks etc.). More than one million automobiles of all types operate in the region. The lack of the necessary infrastructure and the large number of automobiles are the main reasons for the slow traffic observed every day despite the imposed restrictions by the local authorities (only civilian cars with odd or even ending numbers on their tags are allowed to enter the center of Athens alternatively during the day-hours). During days with high concentrations of air pollutants additional restrictions in the traffic, central heating and industry are imposed. The automobiles are considered as the main source of photochemical pollutants recorded in the GAA.

Most of the industrial and small business activities are concentrated in the western part of the basin. None of the industrial installations in this area can be considered as large air pollution sources other than a few chemical factories at the harbour of Piraeus. A large power plant is located at the harbour of Piraeus but it has not been in operation for the last 12 years, for environmental reasons. Two other power plants are located at the SE edge of the Attiki Peninsula (cape Sounion) and on the island of Evoia at the central Evoic Gulf, respectively. The harbour of

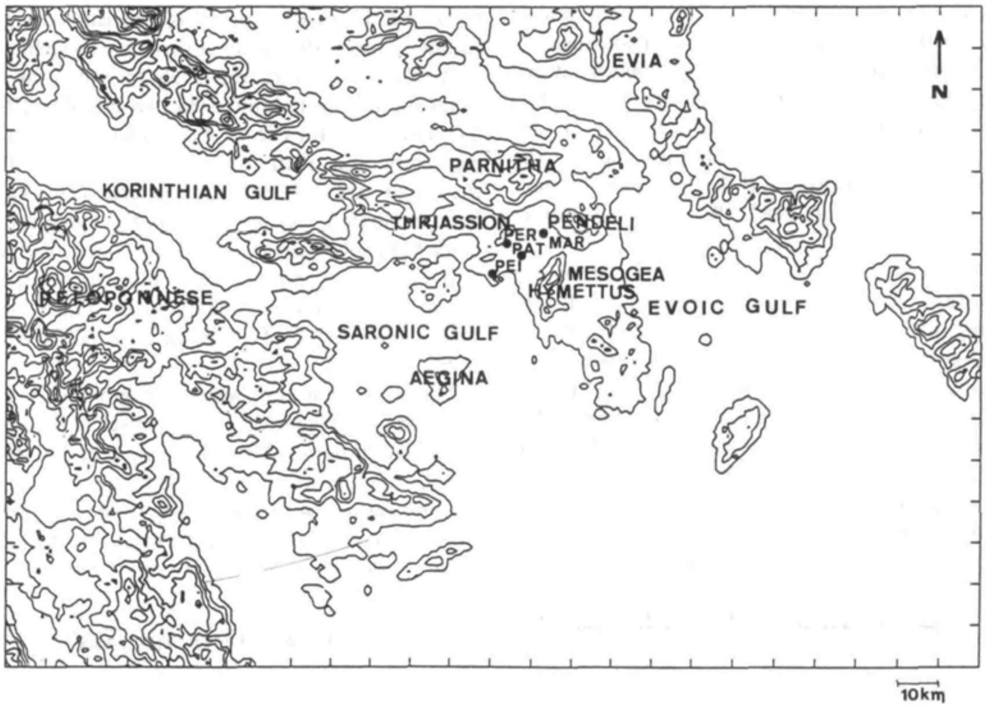

Fig. 1. The topography of SE Greece. Contours are every 250 m. The axes show the distance in km from the SW corner of the domain.

Piraeus is the biggest in the country and the air pollution from the ships is considered to be significant. The international airport of Athens, another important source, is located at the SE edge of the city of Athens, S of Mount Hymettus near the coast. Most of the industrial activities of the GAA and the country are concentrated at the Thriassion Plain and west of it. There are a number of heavy industrial operations such as refineries, steel smelters, cement plants, shipyards etc. A considerable number of industrial installations are also located in the area NNW of Mount Parnitha.

MODELS USED FOR THE SIMULATIONS

1. *The RAMS model*

The model used for the flow simulations over the GAA is the Colorado State University Regional Atmospheric Modeling System (CSU-RAMS) (Walko and Tremback, 1991; Xian and Pielke, 1991; Pielke *et al.*, 1992). The CSU-RAMS is a highly versatile tool that can be used in air quality studies and in a wide variety of other atmospheric phenomena. Some of the RAMS features include (Pielke *et al.*, 1992):

- Two-way interactive nesting with any number of fine nest grids.
- Cartesian or polar stereographic horizontal coordinates with terrain-following coordinate surfaces.
- Non-hydrostatic or hydrostatic time-split time differencing.
- Various levels of microphysical complexity, including cloud microphysics.
- Parameterizations of shortwave and longwave radiative transfer through clear and cloudy atmospheres.
- Surface layer parametertization with a prognostic soil model and vegetative canopy.
- Several options for lateral and top boundary conditions.
- Horizontally homogeneous or variable initialization (isentropic analysis). Output from other models could be used for initialization (e.g. ECMWF).
- It is highly portable and runs on several types of computers.

Because of these capabilities, RAMS is considered as one of the most appropriate models for simulations over Athens.

2. *The CALGRID model*

In order to study the transformation, transport and removal of the various pollutants, the cause of the air quality deterioration in Athens, a three-dimensional air quality grid model should be used. The most widely known photochemical models for air basin scales are the Urban Airshed Model (UAM) (Chico and Lester, 1992), the CALTECH Model (Harley *et al.*, 1992) and CALGRID (Yamartino *et al.*, 1989). Among the three models, CALGRID has been chosen, because of both scientific and practical advantages, some of which are (Yamartino *et al.*, 1992):

- A horizontal advection scheme that prohibits negative concentrations and exhibits little numerical diffusion.
- A full resistance-based model for the computation of dry deposition rates as a function of geophysical parameters, meteorological conditions and pollutant characteristics.
- A modern photochemical scheme, based on the SAPRC mechanisms (Carter, 1988, 1990). The main advantage of the implementation of the SAPRC chemical mechanism is that it can be upgraded, relatively easily, without any major changes of the rest of the code (Pilinis *et al.*, 1990; Pandis *et al.*, 1992)
- Excellent documentation (Yamartino *et al.*, 1989).
- New structured Fortran computer code, that is highly modular and machine independent, which enabled the program to operate on both an IBM-RISC 6000 and a VAX/VMS workstations, without any major changes.
- Three options for the vertical layer structure.
- Many options for the level of complexity and detail for such input data as the emission inventory, the initial and the boundary conditions.
- A wide variety of output options.

Since the time the decision to use CALGRID was made, many of its features have been also incorporated in both the CALTECH and UAM and intercomparison studies are currently under way.

THE EPISODE OF 25–26 MAY 1990

The episode chosen for the simulation is that of 25–26 May 1990. During this episode, a high pressure system was covering the central and eastern Mediterranean and extended over Eastern Europe. This high pressure system was associated with a ridge over the Central Mediterranean and Greece and a trough over Western Mediterranean and Europe. At the surface, a weak pressure gradient was observed over Greece and the Aegean. Warm advection aloft (850 and 700 hPa) was evident during this day and the previous one. According to the upper-air observations at the airport of Athens (GAA) the synoptic flow during night was from W-WNW at the lower atmospheric layers and NW aloft with speeds ranging from 2 to 7 m s^{-1}. At noon, the flow was from WSW in the lower layers with no significant changes aloft. From 23 until 25 May the maximum rise of the temperature at the layer between 950 and 700 hPa was approximately 11°C which shows the strength of the warm advection. A strong temperature inversion was observed up to 920 hPa during the night from 25 to 26 May, which did not break up during day hours. Under these synoptic conditions, a sea breeze developed over the GAA but it was not as strong as it is usually during summer. S to SW winds usually observed over the Athens basin started to appear almost 1 h before noon and were relatively light (2–4 m s^{-1}) while previous to these hours the winds were very light from variable directions. Due to these weather conditions the atmosphere over the GAA was very stable. Mixing height calculations showed that the afternoon mixing height for the 3 days 24–26 May was from 150 to 200 m while the night mixing height was from 60 to 100 m. When this air pollution episode ended on 27 May, the afternoon and night mixing heights were 600 and 175 m, respectively. Under these conditions, the concentrations of some primary and secondary air pollutants reached such high levels, that it was considered one of the worst air pollution episodes recorded in Athens. More information about the weather conditions during these days are provided in Kallos and Kassomenos (1992).

APPLICATION OF THE MODELING SYSTEM

RAMS was used first with three model domains: the coarse one with grid increments of 16 km covering all of Greece, the Aegean Sea and a portion of Turkey, while the second domain was covering the NE Peloponnese, the Saronic Gulf and a large portion of mainland Greece with horizontal grid increments of 4 km. A third domain with a grid increment of 2 km was also used. The coarse grid was used in order to provide the necessary lateral boundary-conditions for the fine grids and some sub-regional scale variations. As it was found in Kallos *et al.* (1993), this is necessary for model simulations over the GAA because of the various mechanically and thermally forced circulations developed in this area and the interaction between them. The three domains had 45 × 43, 46 × 46, and 38 × 38 horizontal grid points. In the vertical, the troposphere was divided into 29 levels with different thicknesses ranging from 5 m near the ground (minimum) to 1 km at the uppermost model levels, up to 11 km above ground level. The simulation started at 200 LST and ended 40 h later. The data used for the initialization were the upper-air data (radiosonde) from the airport to Helliniko in Athens (station GMS). Additional data used for the initialization were the climatological values of the sea-surface temperatures for the water surfaces, and the land-use data (such as vegetation index, urban areas, soil-type etc.), which have been derived from satellite images and cartographic maps. The roughness length over land was determined at each grid cell according to the land-use.

For this simulation, RAMS was configured to estimate the subgrid-scale mixing, coefficients according to Xian and Pielke (1991) (Smagorinsky deformation K). Additional set-ups are:

- The detailed calculation of shortwave and longwave radiation.
- The calculation of land surface temperature, based on a prognostic equation for soil temperature and water content.
- The absorbing layer (Rayleigh friction) at the top five model levels.
- Zero gradient lateral boundary conditions for the coarse grid.

All the necessary meteorological data and other parameters required by CALGRID were produced by RAMS and stored hourly in the required formats (Yamartino *et al.*, 1989).

One of the most important parameters in air quality models, like CALGRID, besides the meteorological parameters is the emission inventory. Errors in the emission inventory cause air quality models to produce non-realistic results. These errors propagate in a non-linear manner, because the photochemical processes are non-linear. A good emission inventory, though, is very difficult to produce. Even in areas such as Los Angeles, California, in which several millions of dollars have been spent in an effort to develop a high quality gridded emission inventory, uncertainties of the order of 300% are still considered usual for road traffic hydrocarbon emissions (Chico and Lester, 1992; Harley *et al.*, 1992; Fujita *et al.*, 1992). Thus, it is easy to understand that the quality of the emission inventory in areas, such as Athens, in which the air pollution modeling effort is in its infancy, is quite poor.

The overall road traffic NO_x emissions used in a previous dispersion modeling study in Athens were 84,718 kg day^{-1} (Winkler *et al.*, 1992), while others estimate the same emissions as being about 50,684 kg day^{-1} (Pattas *et al.*, 1987). To this uncertainity one has to add the uncertainity of the spatial and temporal distribution of the emissions, as well as the uncertainty of the emissions of the various hydrocarbons and the question of their speciation. For this study a gridded emission inventory provided by Winkler was used, which is supposed to be an emission inventory for a typical 24-h period in Athens. This inventory was used for our base case calculations, though, due to the large uncertainties, additional runs were performed in order to evaluate the importance of the emission accuracy to the model predictions.

The domain on which CALGRID was applied was the same 46 × 46 grid system, that was used in RAMS, the center of which is Athens. The size of each horizontal grid, as described previously, was 4 km in both directions. In the vertical, 10 layers of variable thickness were used, up to 2500 m above ground level. The domain was chosen in such way so that the boundary conditions (concentrations) can be assumed to be very small compared to the concentrations in the basin. Thus, concentrations smaller than 1 ppb were used, as boundary conditions, for all the pollutants in the base case calculation. Because of the extent of the domain of the simulation it was found that an increase of the boundary conditions even by an order of magnitude did not affect the results in the basin of Athens during this specific episode.

To make the calculations easy to evaluate, concentrations smaller than 1 ppb were used as initial conditions for the pollutant species of interest. The effects of the choice of the initial conditions on the predicted concentrations were then evaluated with additional runs.

The CALGRID model was applied for 36 h, from 400 (LST) of 25 May 1990 until 1600 (LST) of 26 May 1990. Figure 2 shows the predicted and measured O_3 concentrations for the 36 h of the simulation for four monitoring stations which are shown in Fig. 1. Unfortunately all the air quality measurement sites are concentrated in the heavily populated part of the basin, thus no data is presented for areas further downwind.

These results are presented as an indication of the adequate behaviour of the modeling system in this specific application. An extensive and scientifically sound evaluation of the system cannot be performed, though, due to the lack of a broad database and the poor quality of the emission inventory. For two of the sites, specifically the port city of Piraeus and Peristeri further inland, there is a good agreement between predictions and measurements. At Maroussi and Patissia, on the other hand, there is a delay in the predicted O_3 peak for both days, which can be attributed to errors in the spatial distribution of the emission inventory, or in local conditions not accurately resolved, due to the size of the grid increment.

Figure 3 shows the evolution of the NO concentrations over the domain of the simulation, both spatially and temporally. The high NO emissions in the urban areas of the basin, combined with the strong thermal inversion, caused a continuous increase in the NO concentrations in the coastal and main metropolitan area of Athens and nitrogen oxide concentrations as high as 300 ppb are predicted by the model, by 1900 (LST) of that day. In general the mixing height calculations showed that the afternoon mixing height was from 150 to 200 m. The situation becomes even worse later at night and early next morning and justifies this episode as one of the worst, with mixing heights in the range of 60–100 m. During that night the very strong inversion combined with the termination of the sea-breeze caused extremely unhealthy conditions, with NO concentrations over 650 ppb in many parts of the basin. As shown in Fig. 3d, the light WNW drainage winds moved, by 600 (LST) of the next day, substantial quantities of NO along the southeastern coastal line, resulting in concentrations higher than 100 ppb. The rise of the inversion the next morning, that is the second day of the episode, caused a decrease of the NO levels, but even so, concentrations higher than 100 ppb are predicted by the model to persist in the northern, residential suburbs of Athens.

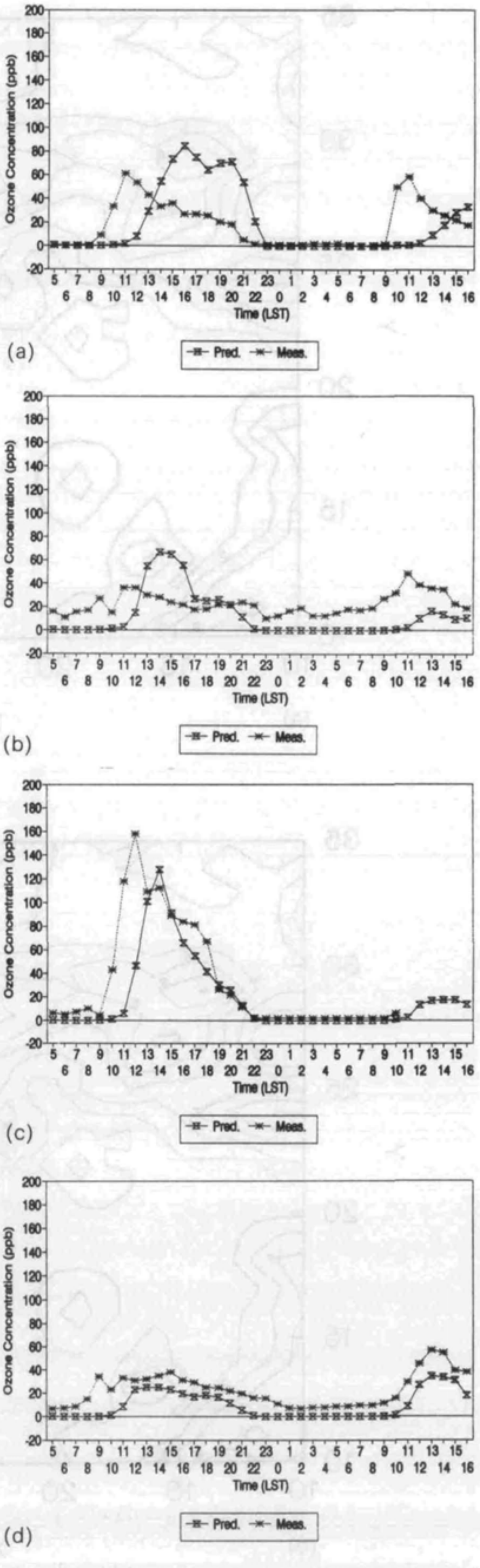

Fig. 2. Predicted and measured O_3 concentrations at (a) Maroussi, (b) Patissia, (c) Peristeri and (d) Piraeus.

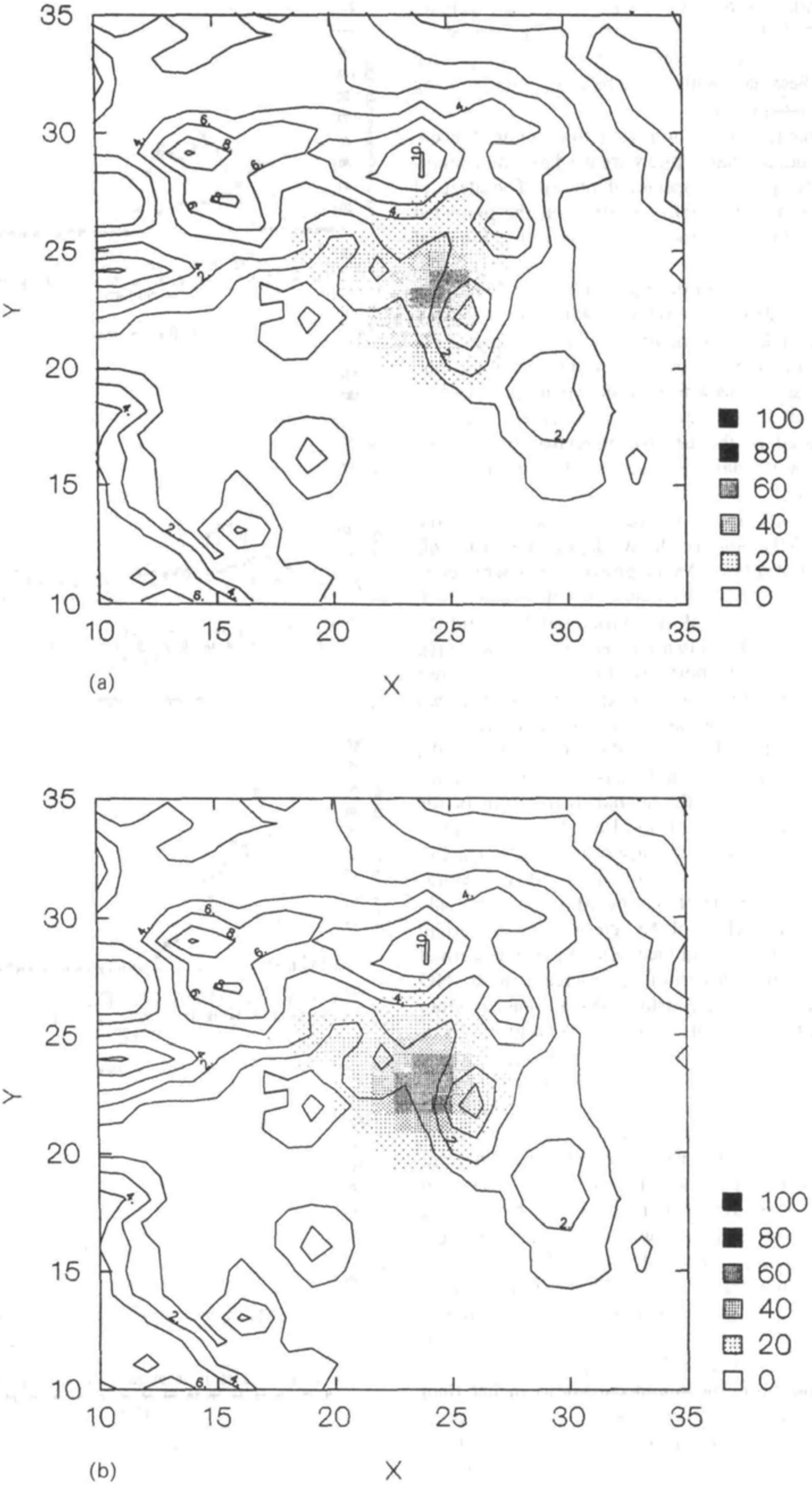

Fig. 3(a–b).

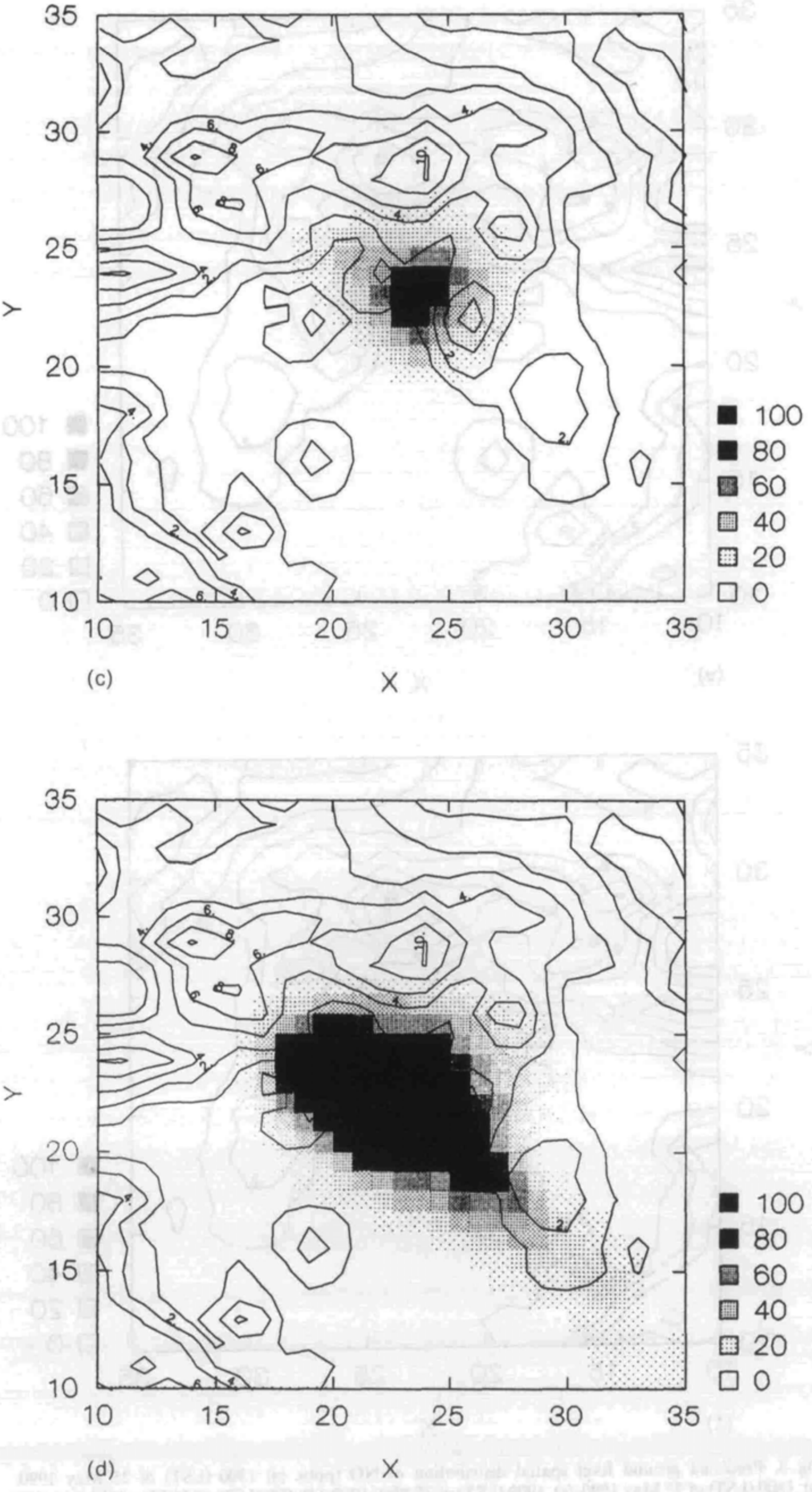
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Fig. 3(c–d).

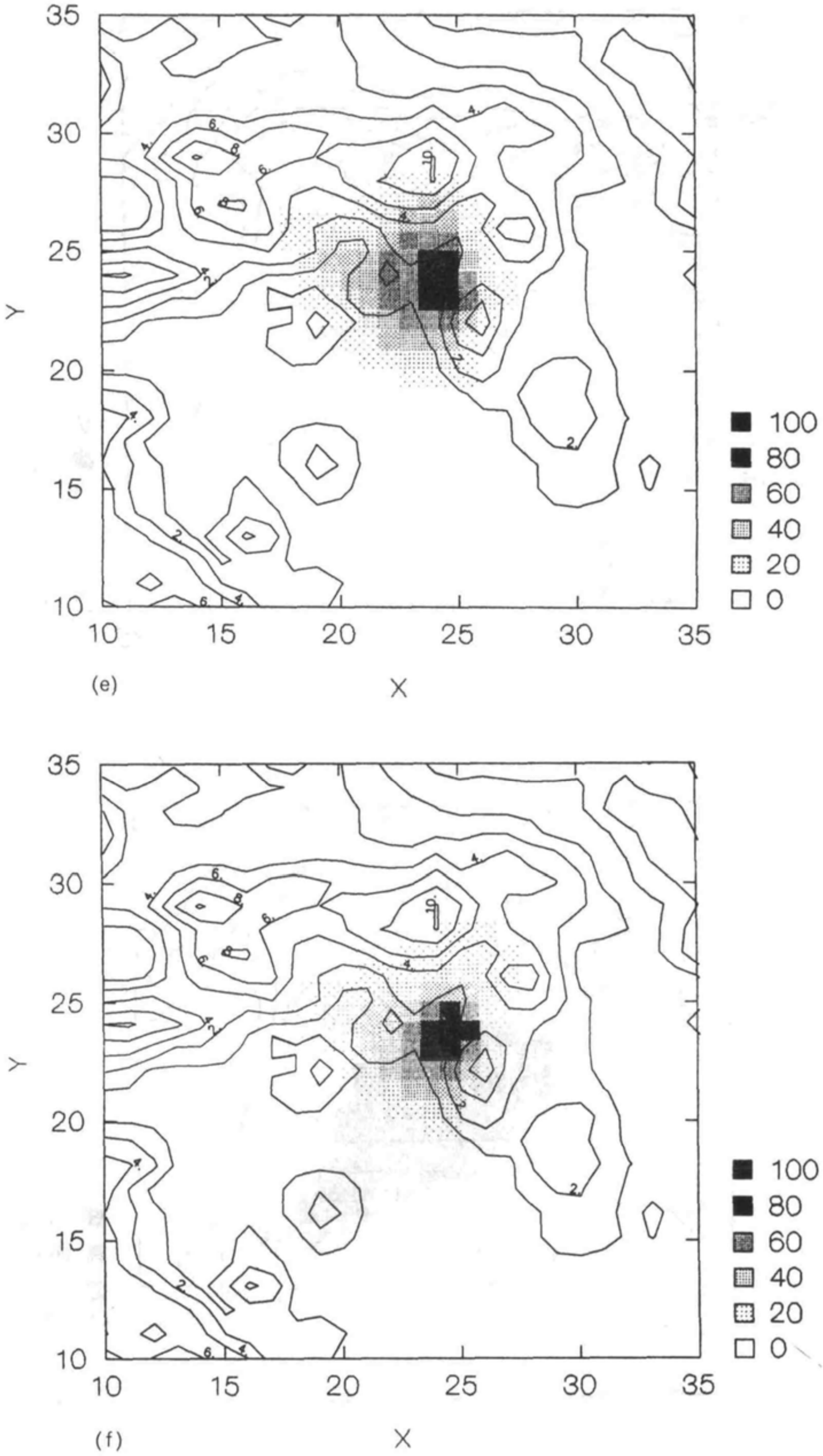

Fig. 3. Predicted ground level spatial distribution of NO (ppb). (a) 1300 (LST) of 25 May 1990, (b) 1600 (LST) of 25 May 1990, (c) 1900 (LST) of 25 May 1990, (d) 600 (LST) of 26 May 1990, (e) 1200 (LST) of 26 May 1990 and (f) 1600 (LST) of 26 May 1990.

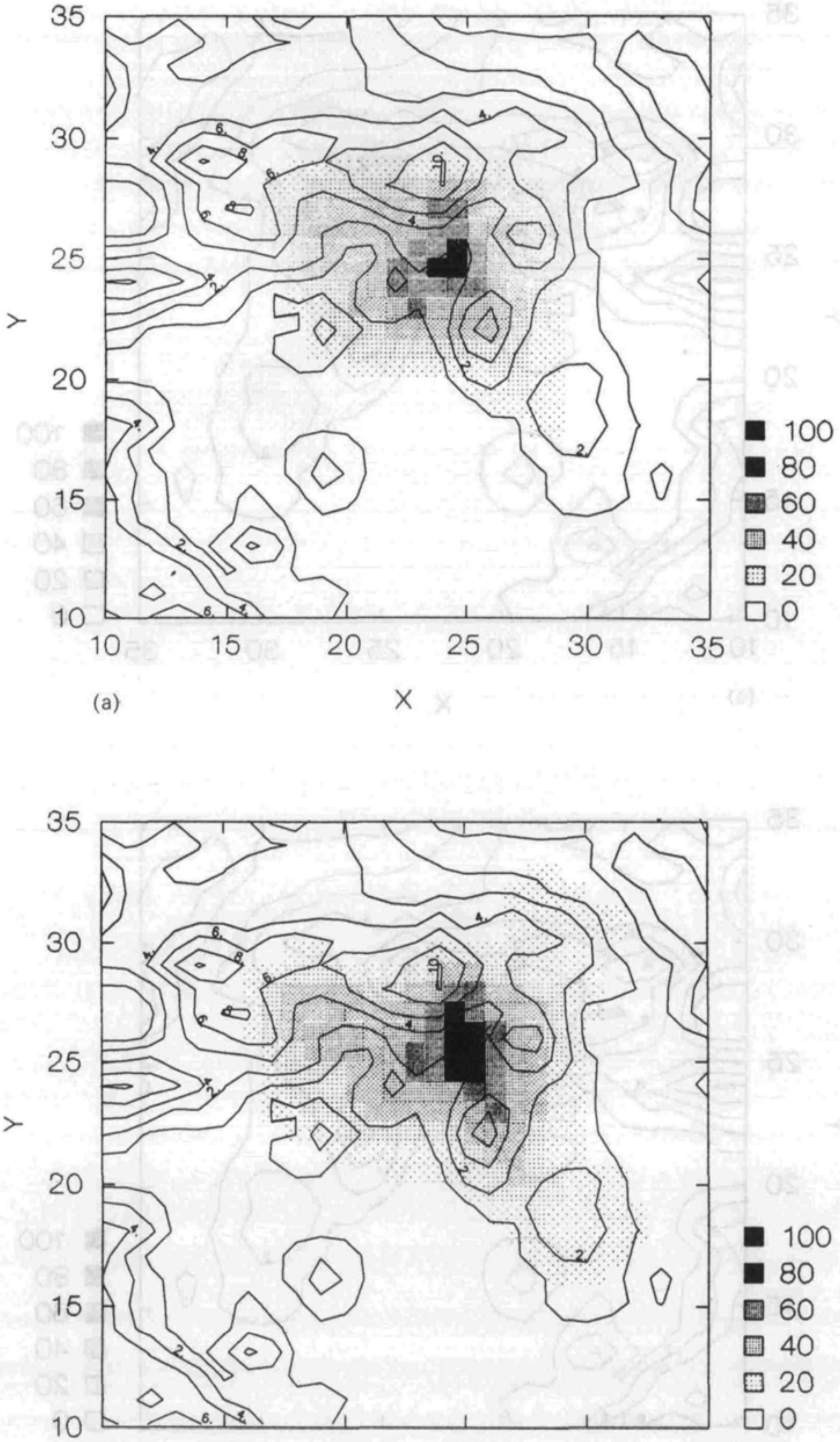

Fig. 4(a–b).

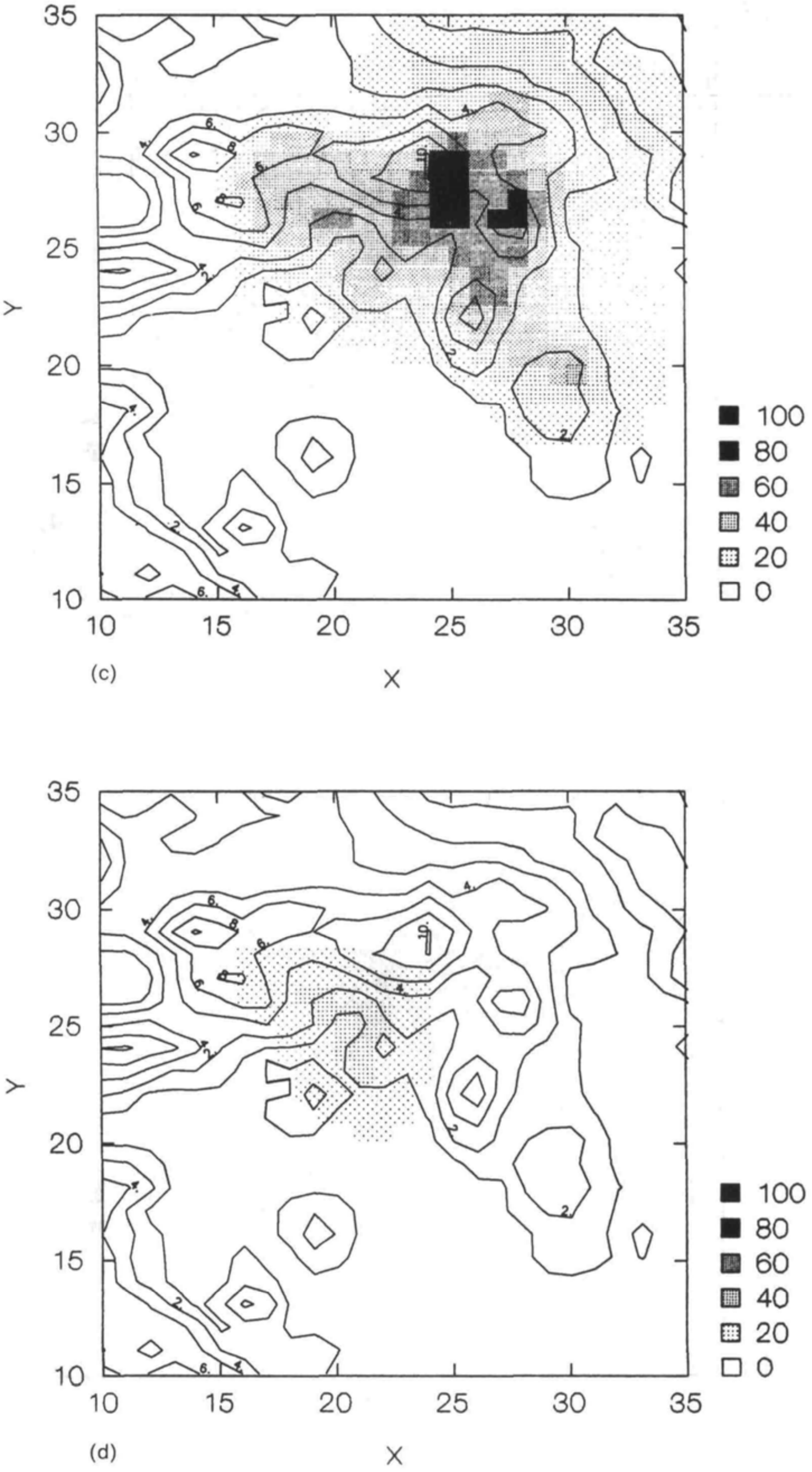
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Fig. 4(c–d).

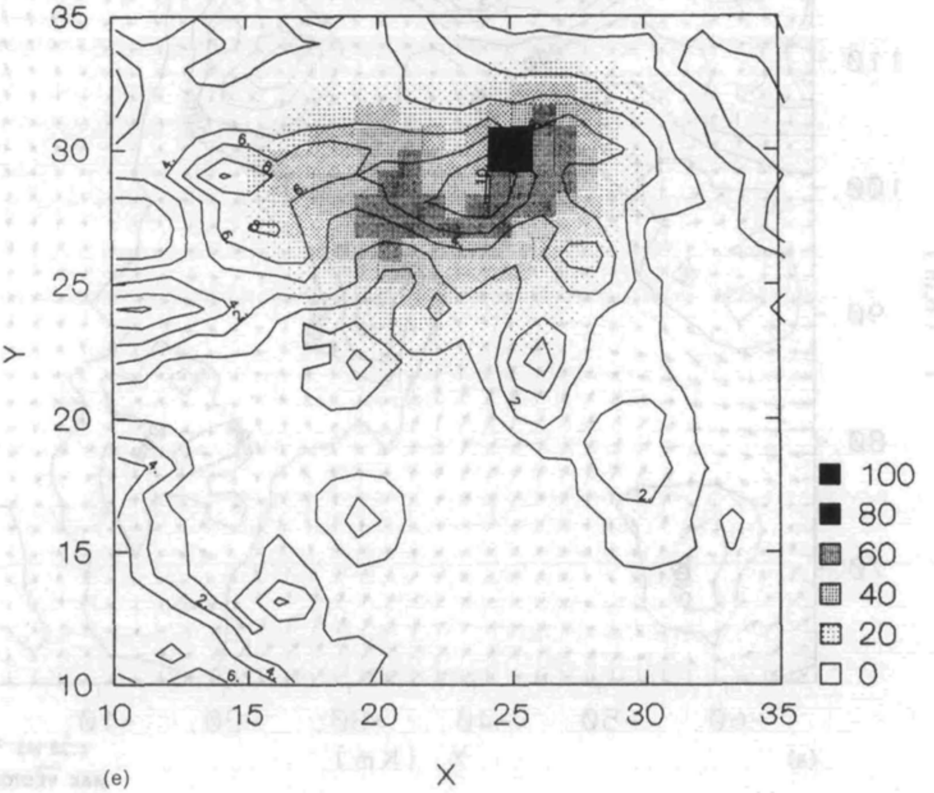

Fig. 4. Predicted ground level spatial distribution of O_3 (ppb). (a) 1300 (LST) of 25 May 1990, (b) 1600 (LST) of 25 May 1990, (c) 1900 (LST) of 25 May 1990, (d) 1200 (LST) of 26 May 1990, and (e) 1600 (LST) of 26 May 1990.

Figure 4 shows the evolution of the ozone concentrations throughout the basin. The strong photochemical activity caused an increase of the ozone levels above 100 ppb by 1300 (LST) of the first day of the simulation in downtown Athens. Later in the afternoon of the same day the light SSW winds, caused by the sea-breeze moved the ozone, which had already formed towards the gaps at the northern edge of the basin. This prediction is consistent with the near ground (20 m) wind fields, presented in Fig. 5. As shown in these figures, the sea-breeze over the GAA started before 1000 (LST) of the 25 May 1990 and ended in the evening, having direction SSW.

The concentrations in the center of Athens and the port of Piraeus are predicted to be low in the afternoon, as presented in Figs 4b and 4c. The main reasons for the low O_3 levels are: (1) the reduced photochemical activity, (2) ozone produced earlier is transported away of the center and (3) the nitrogen oxide rich environment, due to the continuous production, causes the destruction of O_3 and the formation NO_2. Later in the evening and during the next morning the ozone concentrations are predicted to be very low, for the same reasons described above. The sea-breeze of the 26 May 1990 causes the motion of the primary pollutants towards the northern part of the domain, while photochemical reactions increase the ozone levels above 100 ppb, by 1600 (LST) of that day, in the NNE. As a consequnce, suburban surface ozone concentrations exceed the concentrations in the downtown areas of Athens. This result is consistent with the findings of other investigators (Lalas *et al.*, 1987; Moussiopoulos, 1990).

In order to verify the behavior of the ozone concentrations a Lagrangian Particle Dispersion Model (LPDM) has been used to track the trajectories of parcels originated in the main source areas of Athens and Piraeus. The necessary input data for the particle model were available from the RAMS simulation. Three continuous ground level sources were used in the LPDM simulation. Figures 6a and 6b show the results of the horizontal location of particles below 100 m at 1600 and 1900 (LST) of 25 May 1990. As shown in the two figures, polluted parcels, in which photochemical reactions continuously take place, move towards the gaps in the northern part of the GAA, causing the increased O_3 concentrations predicted by CALGRID.

The evolution of the NO_2 concentrations is presented in Fig. 7. Concentrations as high as 100 ppb are predicted to exist throughout the urban parts of the basin by 1300 (LST) of 25 May 1990. As photochemical activity intensifies and the dirty parcels move NNE, NO_2 concentrations higher than 200 ppb are

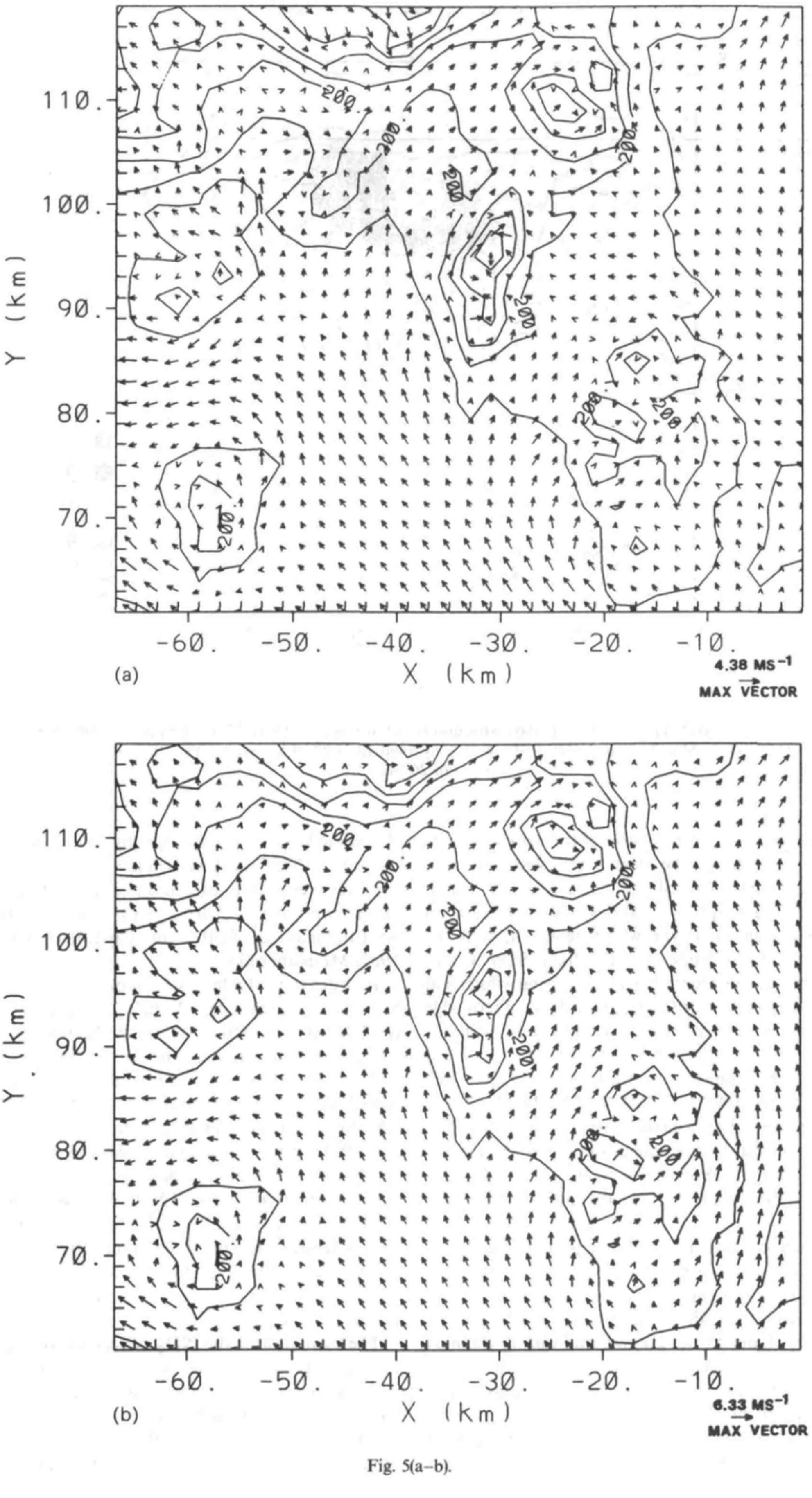

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Fig. 5(a–b).

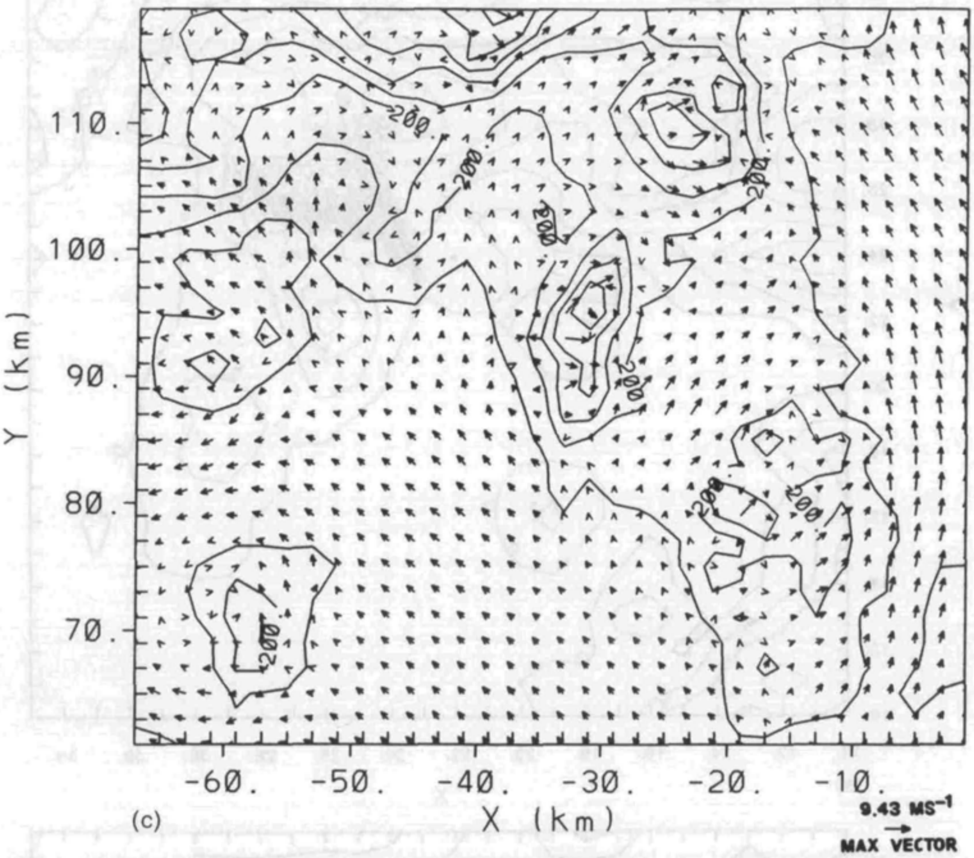

Fig. 5. Predicted ground wind field. (a) 1000 (LST) of 25 May 1990, (b) 1400 (LST) of 25 May 1990, (c) 1800 (LST) of 25 May 1990. The grid increment is 2 km.

predicted to exist by 1600 (LST) of the same day over the northern suburbs of Athens. Later in the afternoon of 25 May 1990 the oxidation of NO and the very low inversion height cause a sharp increase in the NO_2 concentrations throughout the GAA. As shown in Figs 7d and e, more moderate values, less than 100 ppb, are predicted for the next day of simulation.

SENSITIVITY ANALYSIS

An initial sensitivity analysis of the air quality model has been performed in order to examine the importance of uncertainties in the emission inventory and the initial conditions. A more systematic analysis of both RAMS and CALGRID will be presented in subsequent publications.

The first sensitivity run corresponds to an across-the-board increase of the NO_x emissions by 100%. The second run corresponds to a decrease of the NO_x emissions by a half, while the third sensitivity run corresponds to an increase of all the emitted hydrocarbons by 100%.

The maximum ozone concentrations predicted by the model for the four sites are presented in Table 1. Ozone seems to be very sensitive to both the NO_x and hydrocarbon emissions. The non-linear behavior is apparent. The increase of the nitrogen oxide emissions by 100% causes a reduction of the ozone levels by as much as five times, while their decrease by 50% results in the doubling of the ozone concentrations. Ozone is predicted to triple in the run with the increased hydrocarbon emissions. The comparison between predicted and observed maximum ozone concentrations indicates that alternative emission inventories may improve the predictability of the model. A more extensive sensitivity study is needed though before this result can be finalized.

Figure 8 displays the comparison of the predicted concentrations when the initial NO_2 is set to 50 ppb and those of the Base Case simulation, in which the initial NO_2 was 1 ppb. No visible differences are observed after the first 4 h of the simulation, indicating that multiday simulations with CALGRID are not sensitive to the initial conditions.

CONCLUSIONS

A prognostic mesoscale model (RAMS) and an Eulerian air quality model (CALGRID) have been coupled and they have been applied in the GAA to study the air pollution episode of 25–26 May 1990. The major finding of this effort is that the combina-

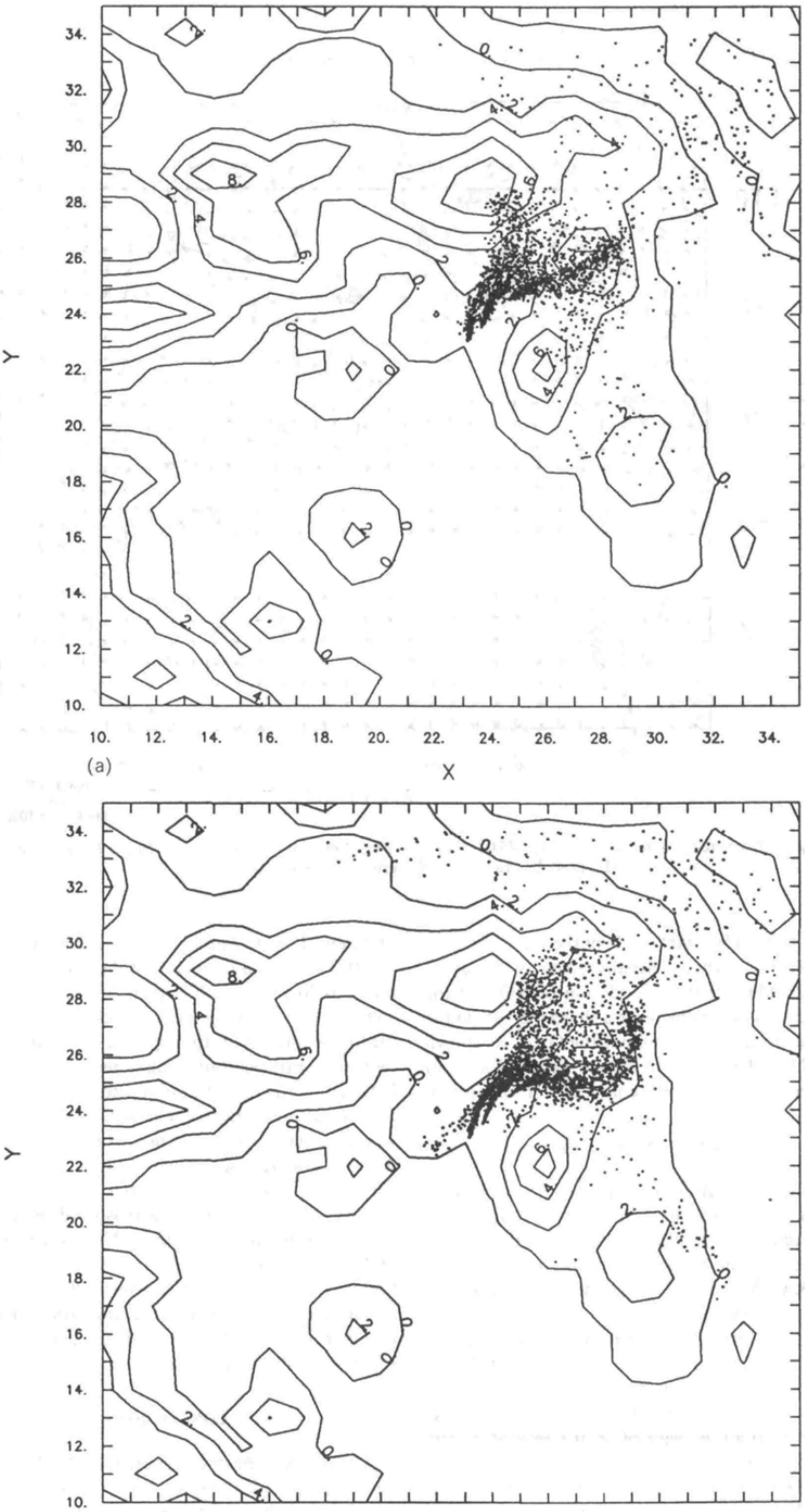

Fig. 6. Predicted position of particles at (a) 1600 and (b) 1900 (LST) of 25 May 1990. Only particles below 100 m above ground level are shown.

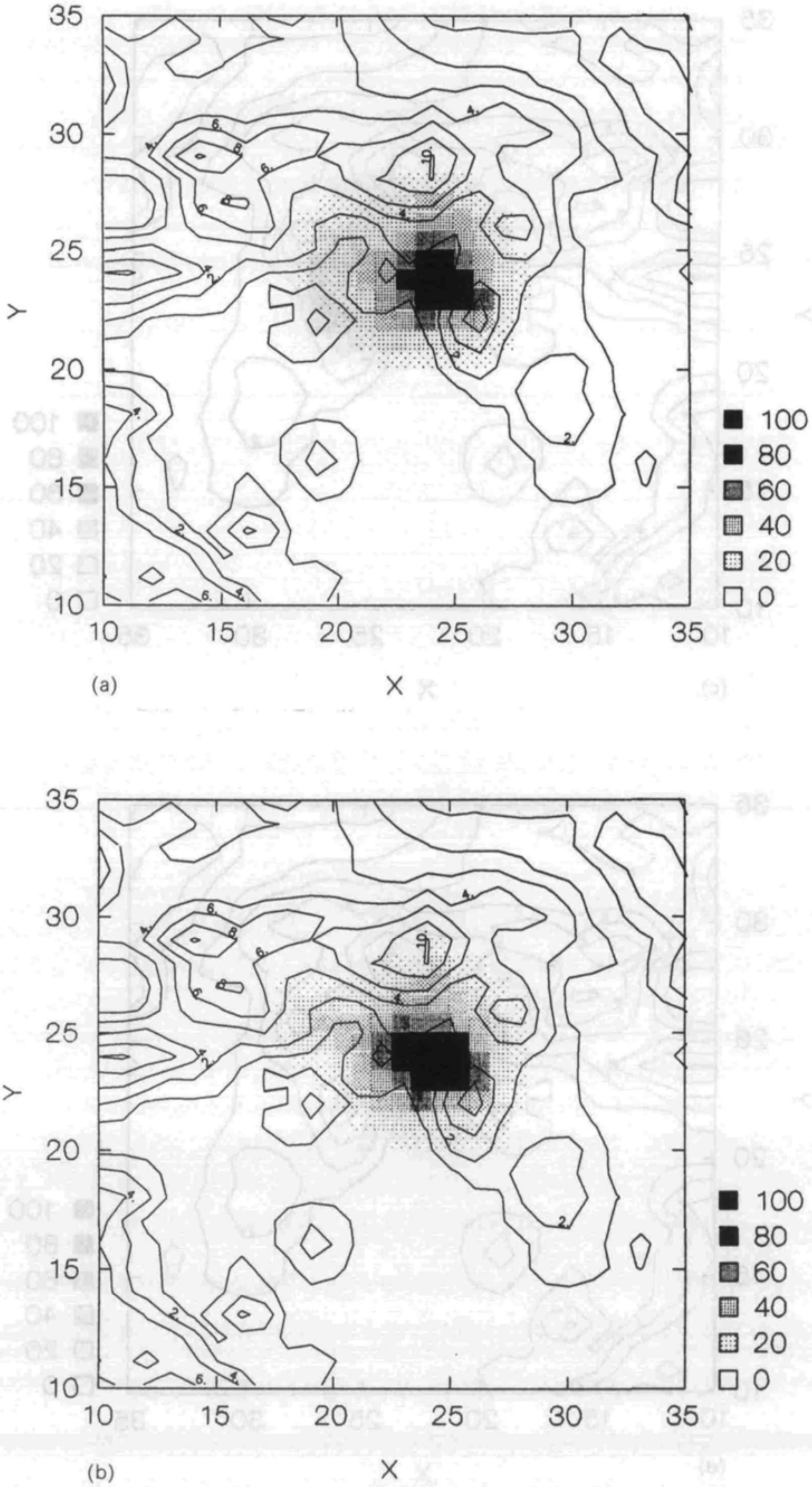

Fig. 7(a–b).

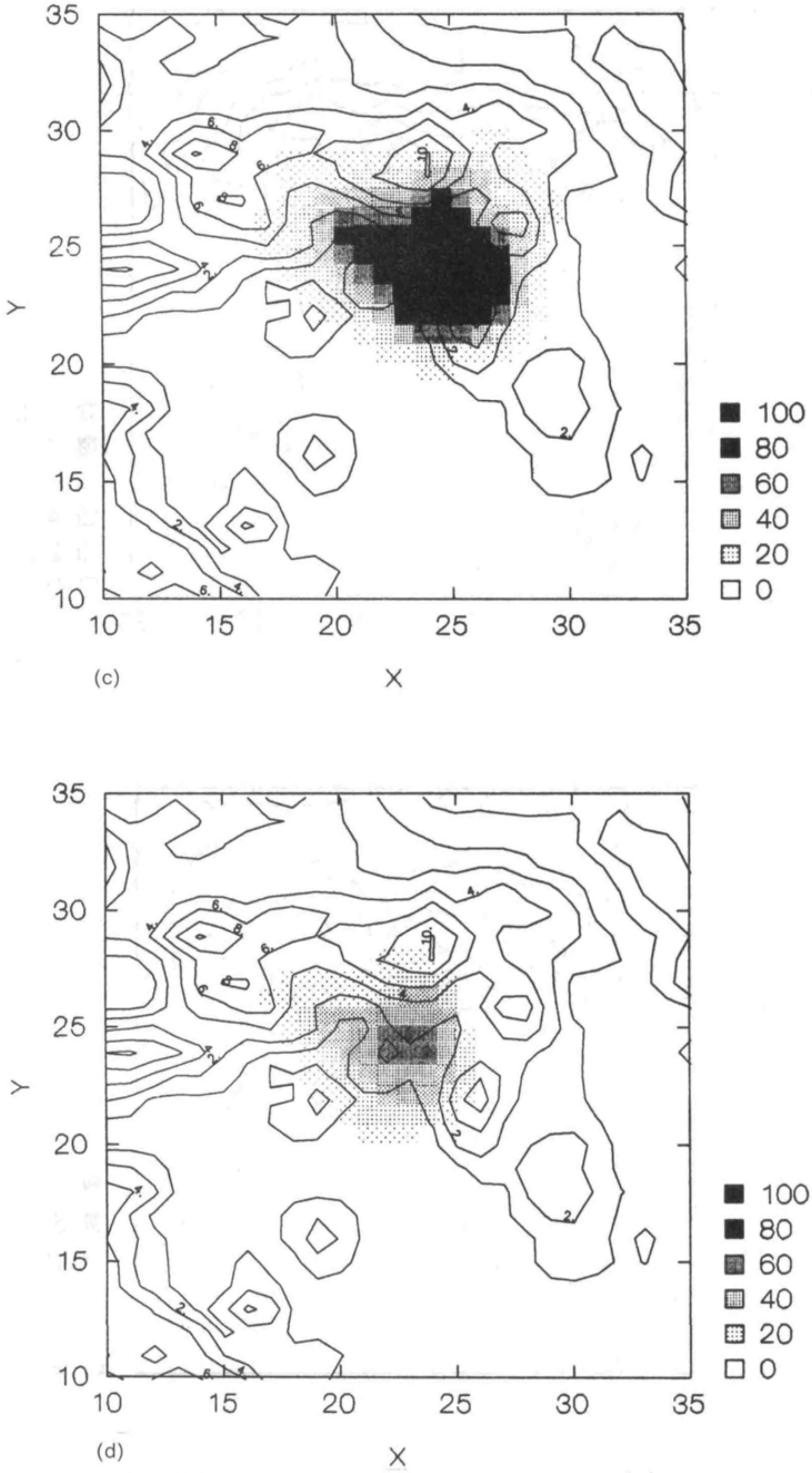

Fig. 7(c–d).

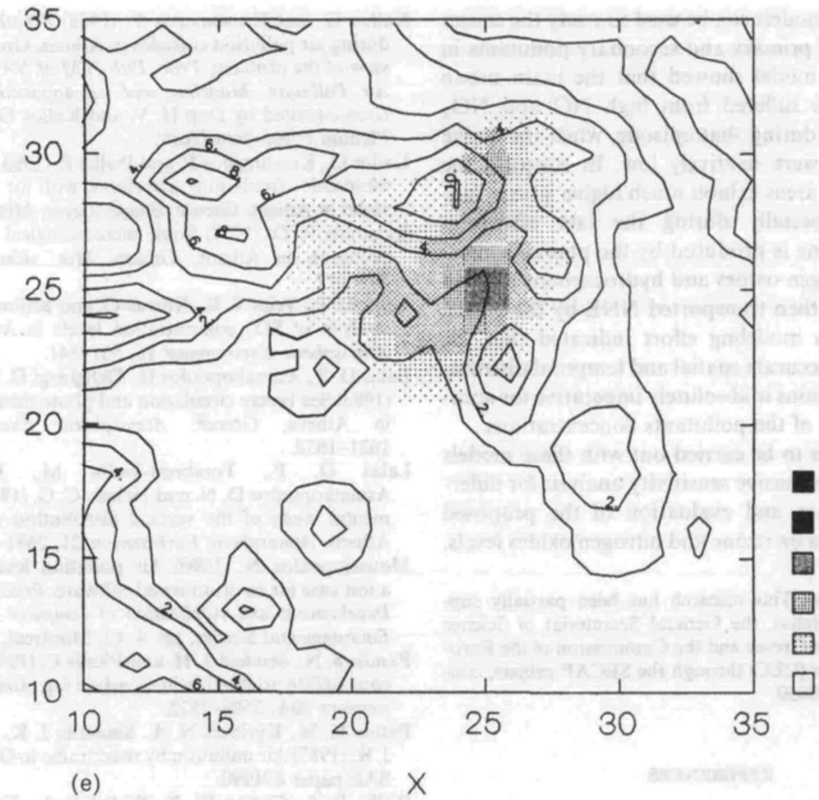

Fig. 7. Predicted ground level spatial distribution of NO_2 (ppb). (a) 1300 (LST) of 25 May 1990, (b) 1600 (LST) of 25 May 1990, (c) 1900 (LST) of 25 May 1990, (d) 1200 (LST) of 26 May 1990 and (e) 1600 (LST) of 26 May 1990.

Table 1. Maximum hourly ozone concentrations (ppb)

Study	Parameter	Variation	Piraeus	Maroussi	Peristeri	Patissia
Measured			38	62	158	36
Base Case	—	—	25	85	128	67
Sens. 1	NO_x Emission	100% Increase	10	30	28	15
Sens. 2	NO_x Emission	50% Decrease	42	150	236	183
Sens. 3	HC Emission	100% Increase	68	260	390	290

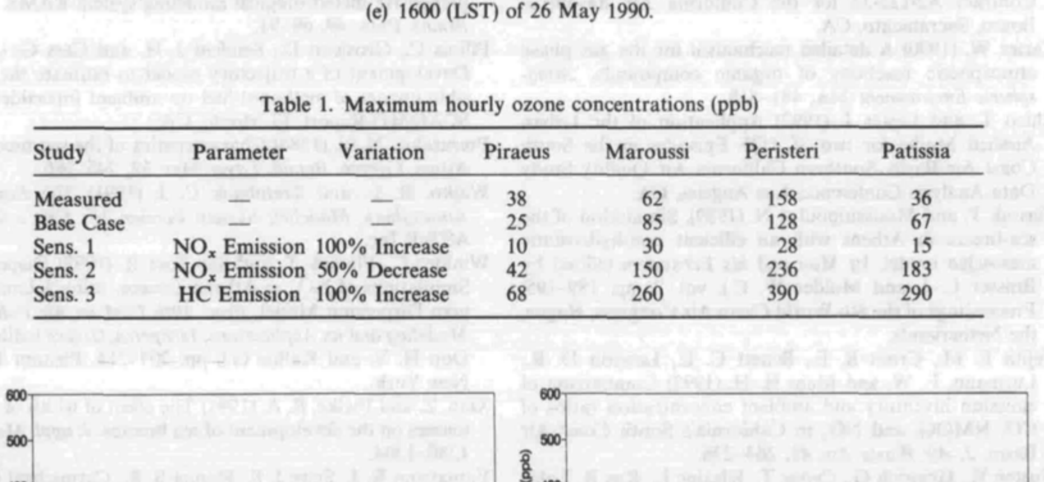

Fig. 8. Predicted NO_2 concentrations at (a) Patissia and (b) Peristeri for both the Base Case simulation and the simulation with 50 ppb initial NO_2 concentrations.

tion of the two models can be used to study the causes of high levels of primary and secondary pollutants in the basin. The model showed that the main urban areas of Athens suffered from high NO and NO_2 concentrations, during that episode, while the ozone concentrations were relatively low. In contrast, the suburban NNE areas exhibit much higher ozone concentrations, especially during the late afternoon hours. This ozone is produced by the photochemical activity of nitrogen oxides and hydrocarbons emitted downtown and then transported NNE by the strong sea-breeze. This modeling effort indicated that the existence of an accurate spatial and temporal distribution of the emissions is absolutely imperative for accurate predictions of the pollutants concentrations.

Future studies to be carried out with these models include a more extensive sensitivity analysis for different episodic days and evaluation of the proposed control strategies on ozone and nitrogen oxides levels.

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