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Air pollution modeling in the Mediterranean Region: analysis and forecasting of episodes

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Abstract

Air pollution modeling in the Mediterranean Region is in its third decade. The first step beyond the Gaussian-type plume dispersion model was the combined use of mesoscale atmospheric models with three-dimensional dispersion models. During the last decade, availability of computer power has increased tremendously, and high-resolution configurations of atmospheric and photochemical models have been applied in regional studies. Advanced modeling techniques have opened new opportunities in air pollution studies. Capabilities are available for forecasting air pollution episodes along with capabilities for traditional analyses of air quality measurements for specific cases of severe episodic atmospheric pollution. In this study, we have elaborated on capabilities for analyzing and forecasting air pollution episodes, mainly for desert dust and photochemical pollutants like ozone and mono-nitrogen oxides (NO_x). Advanced modeling techniques were used with the SKIRON/Eta atmospheric and dust-modeling system, RAMS atmospheric modeling system, and air quality model CAMx. The methodology for analyzing air pollution episodes and products for the day-to-day forecasting application were analyzed with available measurements from the Mediterranean Region and Greece. The results from air quality forecasts have shown acceptable agreement with the observations. For photochemical pollutants, the necessity for higher model and emission resolutions and boundary conditions has become evident. The gas-to-particle conversion mechanisms and separation of anthropogenic and natural sources of origin for episodes of extreme pollution are still open issues. Nevertheless, simulating and forecasting air pollution processes has proven a useful and adequate methodology for investigating air quality degradation in various scales and locations.

Keywords: Air pollution, Modeling, Particulate matter, Photochemistry, Dust

1. Introduction

In the beginning of three-dimensional dispersion models, techniques were either Eulerian or Lagrangian. Implementation of complex, physicochemical processes led to the first photochemical models, which were applied with the Eulerian approach. The mesoscale models were initialized in a rather simplistic way by considering horizontally uniform fields, and domain sizes were on the order of 100 km. This type of modeling was appropriate for conceptual types of analyses or for thermally driven local circulations and was applied in urban simulations. The problems have proven to be significant, and several of these simulations have led to wrong conclusions. A typical example is the greater Athens area where the conceptual models of the 1980s were reproduced by dispersion models, and these early simulations widely adopted the recirculation of air pollutants over a few tens of kilometers. During the 1990s, the new generation of atmospheric models was introduced, and their initializations were performed in a nonuniform way. Multi-scale types of approaches have been adopted (Kallos et al., 1993, Millan et al., 1997), and these simulations have revealed the role of various scale flow interactions in air quality degradation. Conceptual models for air pollutant recirculation within a few tens of kilometers have shown their limitations.

During the last decade, computer power has increased tremendously, and high-resolution configurations of atmospheric and photochemical models have been applied to regional studies. These simulations have demonstrated the role of long-range transport of air pollutants on air quality degradation in various locations around the Mediterranean Region. Key issues that influence long-range transport of air pollutants include recirculation patterns, multiple layering of different ages of air pollutants, horizontal transport within or above the marine boundary layer, and thermal circulations over mountainous regions (Kallos et al., 1997, 1999, 2007). Despite progress related to the description of these physical processes within atmospheric models, several problems still arise when performing quantitative analyses of air pollutant concentrations. Resolution of the emission inventory is partially responsible for the biases that are often introduced in comparisons of measured with simulated values. Emission inventories of greater detail (higher-spatial resolution of the area sources) (Bossioli et al., 2007) together with greater accuracy in representing meteorological features that influence the production/destruction/removal of air pollutants could decrease these biases (Sistla et al., 1996, Biswas and Rao 2001, Hogrefe et al., 2001).

With the experience gained over previous years of modeling research and development, the first attempts at operational air quality forecasts were realized by the Atmospheric Modeling and Weather Forecasting Group of the University of Athens during the Athens Olympics (summer 2004). Meteorological and air quality forecasts were accomplished with implementation of the SKIRON/Eta weather and Saharan dust-forecasting system, Regional Atmospheric Modeling System (RAMS) weather-forecasting system, and Comprehensive Air Quality Model with Extensions (CAMx). These modeling systems have been applied in the nested mode in order to cover the multi-scale flow and air pollution processes in the Mediterranean Region and Greece. The chemical processes included gaseous and particulate pollutants. Anthropogenic and natural sources of origin were taken into account, including Saharan dust as the naturally produced pollutant and ozone and nitrogen dioxide as the anthropogenically produced pollutants. In this paper, the previously stated issues are summarized, and the experience gained is discussed.

2. Model description

The modeling systems used for performing simulations include the SKIRON/ETA modeling system developed at the University of Athens by the Atmospheric Modeling and Weather Forecasting Group (Kallos 1997, Nickovic et al., 2001, Kallos et al., 2006). This system possesses enhanced capabilities with the unique capability of simulating the dust cycle (uptake, transport, deposition). RAMS is most frequently used to simulate atmospheric phenomena on the mesoscale (horizontal scales from 2–2000 km) and for various other applications. This model also includes a two-way, interactive, nested grid structure and detailed cloud microphysics. Detailed information about the RAMS modeling system can be found in Cotton et al. (2003).

CAMx (Environ, 2003) is an Eulerian photochemical model that allows integrated assessment of air-pollution over many scales ranging from urban to super regional scales (http://www.camx.com). CAMx also has structures for modeling aerosols, processes linked to the CB4 (Carbon Bond 4 mechanism) gas-phase chemical mechanism, science modules for aqueous chemistry (RADM-AQ), inorganic aerosol thermodynamics/partitioning (ISORROPIA), and secondary organic aerosol formation/partitioning (SOAP).

3. Air pollution modeling: analyzing episodes

Anthropogenic gases, particulate matter (PM), and natural aerosol-like desert dust are the subjects of modeling studies in conjunction with measurements of atmospheric pollutants. Ozone is a well-known secondary pollutant, which has been the primary target in several studies during the past 20 years. Ozone formation, destruction, transport, and deposition patterns have been identified in various projects in the past. During the last years, a great number of studies have focused on the important role of aerosols in the air quality of a specific area due to the potential impact on human health and ecosystems (di Sarra et al., 2001, Rodriguez et al., 2001). Some European Union (EU) research projects on air quality in the Mediterranean Region with representative publications follow: MECAPIP (Millan et al., 1996); SECAP (Millan et al., 1997); T-TRAPEM (Kallos et al., 1997); MEDCAPHOT-TRACE (Ziomas 1998;, BEMA (Seufert 1997); MEDUSE (Soderman 1998); MAMCS (Pirrone et al., 2003); SUB-AERO (Lazaridis et al., 2006); ADIOS (Loye-Pilot and Benyahya 2003); PAUR I, II (Zerefos et al., 2002); and MINOS (ACP special issue, Vol.3, Editors: Mihalopoulos and de Reus, 2003). Today, scientific interest focuses on the patterns that characterize aerosols in the atmosphere, as well as the interaction between gases and PM, mainly of small sizes.

Advanced modeling systems are considered useful tools for analyzing air pollution episodes and evaluating the impact of different sources from various locations on air quality degradation. Focusing on anthropogenic aerosols and particulate sulfate production and transport are of major importance to impacts on climate and environment. The first step in identifying the paths and transformation of SO_2 (sulfur dioxide) to particulate sulfate was performed by calculating the sulfate ratio with the air quality model CAMx (Kallos et al., 2004). The sulfate ratio has been used in previous studies (Luria et al., 1996) to define the chemical age of air masses based on measurements of sulfur dioxide and particulate sulfate. The sulfate ratio is characterized as the ratio of sulfate concentration to total sulfur concentration ($RATIO = \frac{PSO4}{SO2 + PSO4}$), leading to a dimensionless value from zero to unity. According to Luria (1996), the higher values for sulfate ratio (greater than 0.1) correspond to aged air masses, and the closer the ratio is to unity, the older the air mass and the longer the travel distance.

An example of the methodology used in analyzing episodes based on the comparison of modeling results with observations of particulate sulfate in southern Greece (station located in Finokalia at northeastern Crete) during August 2001 is presented in Fig. 1. The modeled sulfate concentrations appear to be lower than the measured sulfate values because of the coarse resolution of the model ($0{,}235^{\circ} \times 0{,}18^{\circ}$), which tends to under predict in situ observations. In addition, the emission inventory has a coarse resolution (16,67 × 16,67 km) that leads to insufficient information about the mass emitted in the modeled domain. The sulfate ratio calculated from the modeled values agrees well with the sulfate ratio calculated from the observed values showing that the processes of chemical transformation in the air quality model are well reproduced.

Sensitivity tests for the gas-to-particle conversion over southern Europe and the Mediterranean have shown that the temporal scales of conversion are on the order of 4–6 h until the sulfate ratio reaches unity. The spatial scale is approximately a few hundred kilometers with no significant differences for the paths within the marine boundary layer and free troposphere. This result can be attributed mainly to the strong horizontal transport component and relatively good mixing due to topographic incongruities. Remarkable differences occurred over the North African Coast because of the abrupt changes in the depth of the atmospheric boundary layer (from approximately 300 m above the seawaters to 3000–4000 m above the land during day hours). Extensive discussion on gas-to-particle conversion processes driven by sulfur dioxide oxidation to particulate sulfate are included in Kallos et al., (2004).

In addition to the anthropogenically produced aerosols, such as sulfates and/or nitrates, desert dust contributes significantly to the air quality degradation due to the episodic character of increased desert dust concentrations (Mihalopoulos et al., 1997, Ozsoy et al., 2001). In general, air pollution episodes originating from anthropogenic activities occur together with desert dust transport episodes due to the prevailing synoptic conditions favorable for dust transport (ahead of a trough or behind an anticyclone in the Mediterranean Region). Such synoptic conditions are most frequently associated with stable atmospheric conditions and stagnation (transport of warm air masses aloft that suppress vertical developments like updraft and convergence zones). Air pollution episodes in southern Europe occur with the transport of continental tropic air masses originating over northern Africa (Kallos et al., 1993). This air mass transport leads to stabilization of the lower troposphere and usually is associated with dust plumes from northern Africa (Alpert et al., 1998).

The amount of Saharan dust deposited on the Mediterranean waters or over the European land exhibits significant seasonal and inter-annual variability (Papadopoulos et al., 2003), having a rather episodic character (Fig. 2). In Figure 2, the intense dust episode of April 17, 2005, is demonstrated. Figure 2a illustrates the dust plume over the eastern Mediterranean Region from the NASA/GSFC satellite. Figure 2b illustrates the integrated dust column (dust load, mg/m^2) as predicted from the SKIRON/Eta-forecasting cycle from the previous day. During that day, measurements from several stations in Athens showed peak values of PM_{10} at about 2500 $\mu g/m^3$, whereas the model predicted a total dust concentration approximately 500 $\mu g/m^3$ near the previous measured value. The peak concentrations in the dust plume were predicted approximately 100 km south of Athens. The PM_{10} measurements included the contribution from local sources and from a variety of species in the specific size range, whereas the SKIRON/Eta model included only dust sources, and the differences in the comparison with the observations were more or less presumable.

The strength and frequency of Saharan dust episodes define annual deposition and aerosol patterns to a high degree, alternating the mean annual values. This leads to the point that long-term modeling and measurement data are essential in understanding the synergetic effects of anthropogenic and natural PM in the atmosphere of the Mediterranean Region. Qualitative assessment of the contribution from excessive dust concentrations in the total PM_{10} levels in Greece is demonstrated with the aid of Table 1.

During 2001–2005 in Athens, PM records showed that there were 135–223 days where at least one monitoring station recorded PM_{10} concentrations above the EU imposed limits. For those days with PM exceedances, a systematic day-to-day analysis was performed based on PM_{10} measurements from several stations in the Attika Region (provided by the Hellenic Ministry for the Environment, Physical Planning, and Public Works); SKIRON/Eta dust forecasting system; SeaWiFs satellite images (Sea-viewing Wide Field-of-view Sensor Project) provided by Norman Kuring, scientific programmer at NASA/Goddard Space Flight Center; and TOMS satellite images (ftp://toms.gsfc.nasa.gov/pub/eptoms/images/aerosol/). The analysis showed a synergetic contribution of anthropogenic (urban, long-range transport) and natural sources (Saharan dust) in 53–70% of the cases.

The paths and scales of transport and transformation of air pollutants in the Mediterranean Region have been identified in previous studies conducted in the framework of various EU projects as discussed previously. Focusing on PM of natural origin (Saharan dust) and of anthropogenic origin (particulate sulfate), the current state of knowledge on the transport/transformation paths for PM in the Mediterranean Region are given in detail in Kallos et al. (2007), along with the comparison of dust concentrations from the SKIRON/Eta forecasting system with available measurements for 2 years, 2001 and 2003.

A conceptual diagram of the transport paths for air pollutants in the Mediterranean Region resulting from analysis of air pollution episodes with the aid of the modeling systems described in section 2 is provided in Fig. 3. The synoptic/regional circulation during summer favored long-range transport of anthropogenic air pollutants released from southern and Eastern Europe and the central Mediterranean Region toward the eastern Mediterranean Region, northern Africa, and the Middle East (broken grey arrows in Fig. 3). Saharan dust transport followed the path toward the eastern and central Mediterranean Region mostly during the transient seasons, whereas during the summer, high amounts of dust were transported mainly to the western Mediterranean (solid arrows in Fig. 3).

4. Air pollution modeling: ozone and dust forecasting

Knowledge gained from years of modeling atmospheric and photochemical processes have provided the ability to forecast weather phenomena and air pollutant concentration in the Mediterranean Region. Since January 1998, the SKIRON/Eta model has operated covering the Mediterranean Region and providing three-day forecasts of dust load and deposition (http://forecast.uoa.gr) among other meteorological parameters. The ozone-forecasting system applied to the Mediterranean Region has operated since July 2004 for the Athens Olympics (http://forecast.uoa.gr). The operational use of atmospheric and air quality models provides the opportunity to study photochemical activity and particle formation and transport in various scales, from mesoscale to regional. During the Athens Olympics, in the summer of 2004, the SKIRON/Eta system was running twice daily in combination with the CAMx model, covering the entire Mediterranean Region with horizontal grid increments of 0.1° latitude/longitude, for a forecasting horizon of 48 h. A higher-resolution system (0,0235 × 0,018° horizontal grid resolution) has been utilized over southeastern Greece covering the greater Athens area and using an emission inventory of 2 × 2 km horizontal resolution. The meteorological fields have been adapted from the RAMS model, because of its nesting capabilities. The high-resolution air quality forecasting system has been nested within the coarser system (SKIRON/Eta–CAMx).

The modeling system has been validated for its performance for a number of cases, especially for those with interesting features. An interesting event was the extreme dust transport episode toward the eastern Mediterranean Region during April 2005, as discussed in the previous section. The operational performance of the model is illustrated in Fig. 4. The time plots for hourly ozone and nitrogen dioxide concentration against measured values in several stations in Athens for April 15–19, 2005, are presented in this figure. Measurements were available during that specific period for the following stations: Galatsi (urban background station), Maroussi and Patission (urban traffic stations), and Zografou (suburban background station). In general, the model underpredicted hourly ozone concentrations at all stations. The monitoring station located in an area with heavy traffic (Patission) overpredicted the very low ozone concentrations. Especially during nighttime, ozone titration by the model significantly altered the diurnal variation as compared to the measurements. Nitrogen dioxide seemed to be overpredicted by the model especially during the nighttime except for the Patission station.

The two stations identified as urban traffic (Maroussi and Patission) were resolved differently by the model. This occurred because the Patission station was located in the heart of Athens and was influenced directly by heavy traffic from public and private transportation, whereas Maroussi was affected by traffic but at the outskirts of the city. Insufficient information on NO_x emissions in the area may have been the reason that the model underestimated NO_2 concentration at the Patission Station. In general, the differences between the modeled and observed values can be attributed to a number of factors such as emission inventory, model configuration, and quality of observational data.

As suggested by van Loon et al. (2007), use of the total oxidant O_x (sum of ozone and nitrogen dioxide concentration, $O_x = NO_2 + O_3$) can provide an estimation of the diurnal cycle of these oxidants without the effect of errors in the titration processes. Comparison of O_x from the model with O_x from the measurements is illustrated in Fig. 5 for one station in Athens. The comparison reveals again the underestimation by the model, although the diurnal cycle of the oxidants is better represented than the diurnal ozone cycle. The previously mentioned air quality forecasting systems are still operational with slightly different configurations, and the results are available daily from http://forecast.uoa.gr.

The effort to produce reliable air quality predictions is a well-based task and is subjected to continuous development and evaluation. Research, sufficient measurements of air pollutants, and statistical evaluation techniques should aid this effort into the future for accurate predictions.

5. Conclusions

The present study summarizes the multi-year research modeling efforts to identify the characteristic spatial and temporal scales of photochemical activity along the previously defined main transport routes over the Mediterranean Region. Air quality degradation in the area is influenced by photochemical pollutants like ozone and aerosols, such as particulate sulfate and desert dust. The use of advanced modeling systems with the aid of observations is a research tool providing remarkable capabilities to analyze episodes of air pollution. The operational use of these modeling systems can be a significant tool for predicting air quality degradation in several spatial scales from urban to regional. The remarks concluded from this work are summarized as follows.

The combined use of limited-area, meteorological models with chemical transport models can form systems that provide reliable predictions of atmospheric gas-phase pollutants. Nevertheless, there are remaining issues that require further investigation and refinement. The issues pertaining to adequacy in the emission inventories and land-use need to be better described and continuously updated. The meteorological fields together with turbulence in urban environments must be improved to aid in better describing pollutant cycles. Long-range transport of atmospheric pollutants is also a key-factor in air quality degradation, which is equally as important as correctly implementing local emission sources.

The Mediterranean Region is an area sensitive to Saharan dust transport, and during extreme episodes, the amounts of Saharan dust can influence the measured levels of PM (PM_{10}) leading to values higher than the imposed EU limits. Analysis of those episodes for Athens (Greece) showed a synergetic contribution of dust to PM_{10} measurements in 50–70% of the cases. This type of analysis provides insight in the need to discriminate the origin of pollutants (natural or anthropogenic) and hence evaluate high PM concentrations and air quality degradation in urban environments based on varying natural origins.

The preceding conclusions are a result of on-going research based on modeling techniques for assessing air quality over specific areas. Modeling tools are subject to continuous development in order to eliminate as many errors as possible. Thus, on-going development has shown the way to produce real-time forecasts of air pollution episodes in different scales from urban to regional. The operational use of advanced atmospheric and air quality models has provided reliable predictions of several air quality episodes in the Mediterranean Region. Nevertheless, continuous research and sufficient measurements of air pollutants should aid this effort toward accurate predictions in the future.

6. Acknowledgments

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Figure Captions

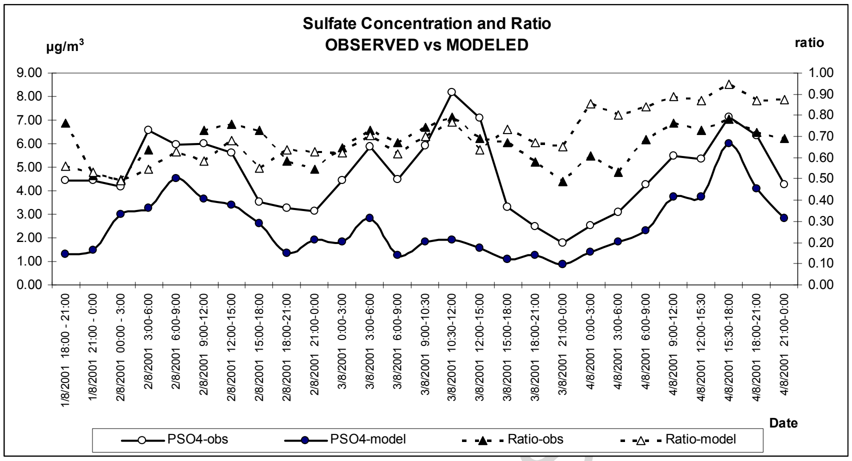
Figure 1. Intercomparison between measurements and model calculations of particulate sulfate and the sulfate ratio at the Finokalia Station, Crete, Greece, during August 1–4, 2001.

Figure 2. Saharan dust episode for April 17, 2005. Left, dust over Greece;, picture taken from NASA/GSFC satellite (2005/107–04/17 at 1140 UTC) (http://earthobservatory.nasa.gov). Right, total dust load (mg/m^2) simulated from SKIRON/Eta dust modeling system for 1200 UTC.

Figure 3. Characteristic paths and scales for transport of air masses in the Mediterranean Region. Grey interrupted arrows are associated with the transport of anthropogenic pollutants in the lower troposphere; solid arrows indicate Saharan dust transport in the Mediterranean Region.

Figure 4. Comparison of observations from stations in the Athens area (Hellenic Ministry for the Environment, Physical Planning, and Public Works) with results from the 48-h air quality forecast (hourly average) with the CAMx model. The left column presents ozone concentrations, and the right column presents nitrogen dioxide concentrations. Time period April 15 to 19, 2005.

Figure 5. Comparison of $NO_2 + O_3$ (ppb) observations from the Galatsi Station (Hellenic Ministry for the Environment, Physical Planning, and Public Works) with results from the 48 h air quality forecast (hourly average) with the CAMx model. Time period April 15 to 19, 2005.



Sulfate Concentration and Ratio
OBSERVED vs MODELED
μg/m3
ratio
Date
PSO4-obs
PSO4-model
Ratio-obs
Ratio-model

Figure 1

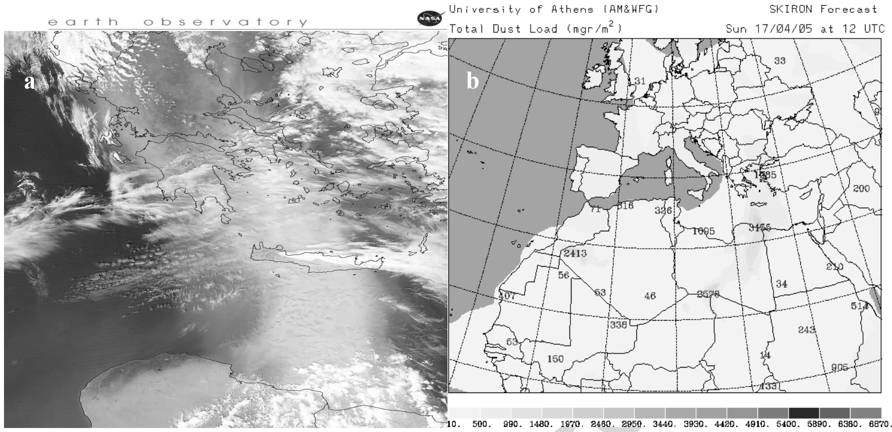

earth observatory
a
University of Athens (AM&WFG)
SKIRON Forecast
Total Dust Load (mgr/m^2)
Sun 17/04/05 at 12 UTC
b
10. 500. 990. 1480. 1970. 2460. 2950. 3440. 3930. 4420. 4910. 5400. 5890. 6380. 6870.

Figure 2

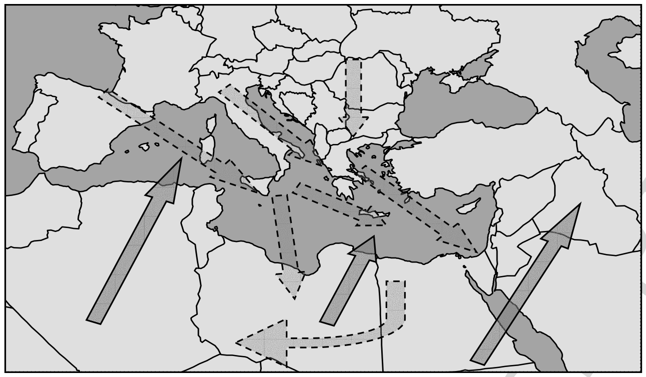
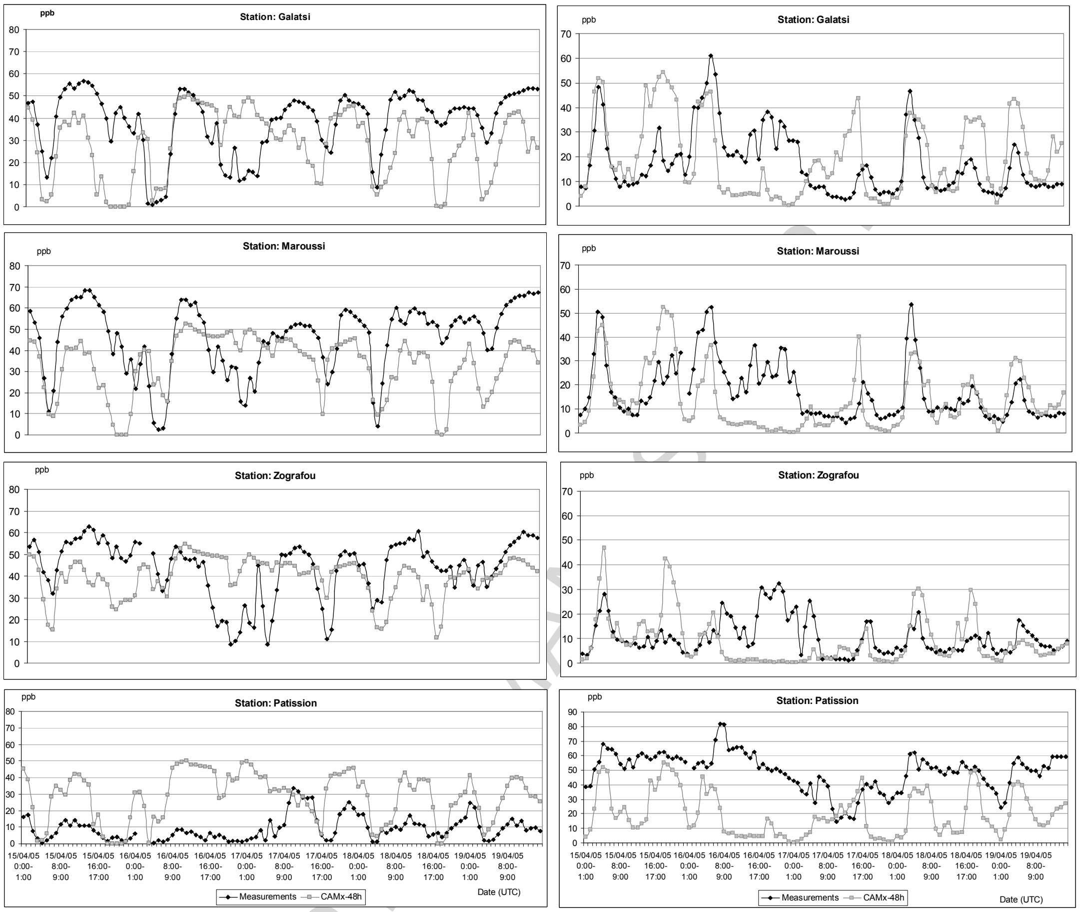


Figure 3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ppb
Station: Galatsi
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Station: Maroussi
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Station: Patission
ppb
Station: Patission
ppb
15/04/05 0:00-1:00
15/04/05 8:00-9:00
15/04/05 16:00-17:00
16/04/05 0:00-1:00
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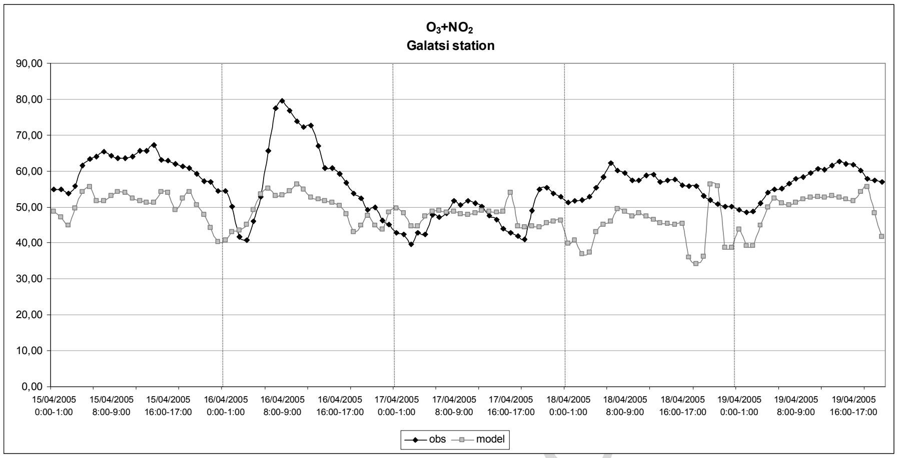
16/04/05 16:00-17:00
17/04/05 0:00-1:00
17/04/05 8:00-9:00
17/04/05 16:00-17:00
18/04/05 0:00-1:00
18/04/05 8:00-9:00
18/04/05 16:00-17:00
19/04/05 0:00-1:00
19/04/05 8:00-9:00
Measurements
CAMx-48h
Date (UTC)

Figure 4



O$_3$+NO$_2$
Galatsi station
90,00
80,00
70,00
60,00
50,00
40,00
30,00
20,00
10,00
0,00
15/04/2005 0:00-1:00
15/04/2005 8:00-9:00
15/04/2005 16:00-17:00
16/04/2005 0:00-1:00
16/04/2005 8:00-9:00
16/04/2005 16:00-17:00
17/04/2005 0:00-1:00
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19/04/2005 0:00-1:00
19/04/2005 8:00-9:00
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obs
model

Figure 5

Table 1. Air quality violations in Athens, Greece, and association with Saharan dust transport for 2001–2005.

	2001	2002	2003	2004	2005
Number of days with exceedances	141	201	135	223	213
Number of days with available SKIRON dust forecasts	102	178	135	223	213
Number of days with available SeaWiFs images	59	192	128	176	174
Number of days with available TOMS images	141	201	135	223	213
Number of days where SKIRON forecasted dust transport in Athens	60	86	59	123	116
Number of days where SeaWiFs images showed dust transport in Athens	45	90	75	96	103
Number of days where TOMS images showed dust transport in Athens	67	75	55	88	60
Percent of exceedance due to dust according to available SKIRON forecasts	58,82%	48,31%	43,70%	55,16%	54,46%
Percent of exceedance due to dust according to SeaWiFs images	76,27%	46,87%	58,59%	54,54%	59,19%
Percent of exceedance due to dust according to TOMS images	47,86%	37,31%	40,74%	39,46%	28,17%
Percent of exceedance due to dust where at least **two** of the systems (SKIRON, SeaWiFs, TOMS) showed dust transport in Athens	**53,57%**	**54,72%**	**51,85%**	**46,64%**	**47,42%**
Percent of exceedance due to dust where at least **one** of the systems (SKIRON, SeaWiFs, TOMS) showed dust transport in Athens	**62,14%**	**63,68%**	**65,92%**	**70,85%**	**52,58%**