

## Long-Range Transport of Anthropogenically and Naturally Produced Particulate Matter in the Mediterranean and North Atlantic: Current State of Knowledge

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#### ABSTRACT

During the past 20 years, organized experimental campaigns as well as continuous development and implementation of air-pollution modeling have led to significant gains in the understanding of the paths and scales of pollutant transport and transformation in the greater Mediterranean region (GMR). The work presented in this paper has two major objectives: 1) to summarize the existing knowledge on the transport paths of particulate matter (PM) in the GMR and 2) to illustrate some new findings related to the transport and transformation properties of PM in the GMR. Findings from previous studies indicate that anthropogenically produced air pollutants from European sources can be transported over long distances, reaching Africa, the Atlantic Ocean, and North America. The PM of natural origin, like Saharan dust, can be transported toward the Atlantic Ocean and North America mostly during the warm period of the year. Recent model simulations and studies in the area indicate that specific long-range transport patterns of aerosols, such as the transport from Asia and the Indian Ocean, central Africa, or America, have negligible or at best limited contribution to air-quality degradation in the GMR when compared with the other sources. Also, new findings from this work suggest that the imposed European Union limits on PM cannot be applicable for southern Europe unless the origin (natural or anthropogenic) of the PM is taken into account. The impacts of high PM levels in the GMR are not limited only to air quality, but also include serious implications for the water budget and the regional climate. These are issues that require extensive investigation because the processes involved are complex, and further model development is needed to include the relevant physicochemical processes properly.

## 1. Introduction

#### a. General considerations

The climatic patterns and the geographic characteristics of the Mediterranean region produce air-quality patterns with remarkable spatial and temporal variability. Concentrations of various pollutants (primary and/ or secondary) are found to be significant in remote locations as well as in multiple-layer structures up to several kilometers above the surface. Various studies in the past have identified the paths and scales of transport and transformation of air pollutants released from Europe toward the eastern Mediterranean and North Africa (Katsoulis and Whelpdale 1990; Luria et al. 1996; Kallos et al. 1997, 1998a,b; Millán et al. 1997,

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2005; Peleg et al. 1997; Gangoiti et al. 2001). Recent studies focus on the importance of long-range transport patterns of particulate matter (PM) of anthropogenic and natural origin as shown by Prospero et al. (2001), Uno et al. (2001), Rodriguez et al. (2001), Bardouki et al. (2003), and Kallos et al. (2006). There are also indications of the existence of transport patterns on larger scales toward/from the Mediterranean region (Ramanathan et al. 2001; Lelieveld et al. 2002; Carmichael et al. 2002).

The current status of knowledge on the above aspects is discussed in this paper, providing some summary remarks on the paths and scales of transport and transformation of PM in the greater Mediterranean region (GMR). The tools used for such analysis are atmospheric and air-pollution modeling techniques, together with air-quality observations. Some key issues concerning the knowledge of the air-quality status in the Euro-Mediterranean region are the identification of the geographic distribution of pollutant sources and the regional climatological description for the GMR. The PM

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FIG. 1. Mean monthly sea level pressure for August 2001, as produced by the analysis fields of ECMWF. Pressure difference along the line AB is approximately 12 hPa.

levels and composition and European Union (EU) policies addressing PM levels should be kept in mind and are hence briefly discussed herein.

Paths and scales of transport and transformation of air pollution in the GMR as identified by several studies in the past are presented and discussed as separate sections in this paper. Of the air pollutants of anthropogenic origin, fine particulate sulfate exhibits remarkable long-range transport patterns and will be the primary focus of the work presented herein. Naturally produced PM, mainly Saharan dust, is also considered as a key component in the long-range transport processes, and therefore a detailed discussion of the processes affected by dust follows. Impacts of synoptic/ regional circulation patterns on transport and transformation processes of PM are discussed thoroughly in this work.

## b. Regional climatological description

The Mediterranean Sea is closed from all sides and is surrounded by high peninsulas and important mountain barriers. The gaps between these major mountainous regions act as channels for the airmass transport toward the GMR. The climatic conditions in the GMR can be roughly divided into cold and warm periods (Maheras et al. 1999). The cold period of the year is characterized by the low-index circulation that is associated with intense cyclogenetic activity. The anticyclonic type of circulation during this period is associated with a coldcore anticyclone lying over central Europe or the Balkan region.

The warm period is characterized by the high-index circulation in which the North Atlantic Ocean low pressure centers extend over Europe and only edges of the fronts reach the GMR (Kallos et al. 1993; Kassomenos et al. 1995). The GMR is dominated by anticyclonic activity and large-scale subsidence. This period of the year is highly controlled by the balance between the North Atlantic Ocean anticyclone (that extends toward Europe and the GMR) and the monsoon activity over the Indian Ocean and the Middle East. This balance exhibits some stereotypical characteristics that define the transport at various scales. A typical case is illustrated in Fig. 1, in which the North Atlantic Ocean anticyclone and the monsoon activity over the Indian Ocean are evident. The resulting pressure gradient over the GMR is relatively strong (10-20 hPa from the western to eastern Mediterranean or even greater), and its variability defines the onset of trade wind systems like the etesians over the Aegean Sea. Figure 1 is produced with the use of mean monthly sea level pressure fields for August of 2001 from the European Centre for Medium-Range Weather Forecasts (ECMWF) analysis fields.

The general pattern of the flow over the GMR has a clear component from north to south or southeast within the lower troposphere. This pattern is illustrated in Fig. 2, in which the wind fields are shown for a typical summer case. Figure 2 has been produced with the aid of the data assimilation module of the Regional Atmospheric Modeling System (RAMS) in which the gridded  $0.5^{\circ} \times 0.5^{\circ}$  fields have been used together with surface and upper-air observations available for the entire domain. The wind field analysis illustrated in Fig. 2 is persistent over the GMR during summer and several days of the transient seasons as discussed in Kallos et al. (1998a). In this figure, the wind fields in the lower troposphere (Fig. 2a) are presented, with the north-to-



FIG. 2. Wind field at 1200 UTC 5 Aug 2001, as simulated by the RAMS at (a) 250 m, (b) 2 km, and (c) 8 km AGL.

south component of transport in the central Mediterranean and the flow field toward the Atlantic Ocean through the Gulf of Gibraltar being the most evident. Figure 2b illustrates the circulation in the free troposphere (2 km above ground), where the anticyclonic activity in the North Atlantic Ocean and the cyclonic activity in the Indian Ocean are clearly shown together with the appearance of the intertropical convergence zone (ITCZ) at the belt north and south of 25°N. In Fig. 2c, the wind fields in the upper troposphere (at 8 km above ground) have as a major characteristic the appearance of the easterlies over central Africa.

During both the cold and the warm periods of the year, the general trend of the winds is from north to south across the Mediterranean region with significant variations in each area. This is mainly due to the differential heating between the two land areas (southern Europe and North Africa) and the Mediterranean Sea. Owing to these complicated flow patterns, the air pollutants released from various sources located in the surrounding areas can be transferred long distances, in a complex fashion (Kallos et al. 1993, 1998a; Luria et al. 1996; Dayan and Levy 2002; Dayan and Lamb 2005). Landscape variability and, especially, land–water contrast result in the formation of thermal circulations that range from a few hundred meters to a few thousand kilometers (Millán et al. 1997, 2005).

The position of the ITCZ during summer is located over the northern part of the Saharan Desert, whereas during winter it is close to the equator. The height of the mixing layer over Europe is approximately 1-2 km during summer days and 100-1000 m during the winter and the transient seasons. The height of the mixing layer over North Africa varies between a few tens of meters during the night and 2-4 km or even deeper during the day, especially during summer (Kallos et al. 1998a). The mixing layer over the Mediterranean Sea is almost stable during the diurnal cycle ( $\sim$ 300 m) and varies slightly with the seasonal cycle (200-350 m). An important feature of the coastal zones of the GMR is the formation of the internal boundary layer. The islands and the peninsulas act as chimneys and obstacles, causing abrupt changes in the mixing depth (Kallos et al. 1997). The formation of relatively strong updrafts can inject polluted air masses from the boundary layer into the free troposphere. All these local meteorological features contribute to the formation of pollutant transport patterns with multifaceted spatial and temporal characteristics.

## c. Geographic distribution of pollutant sources

The major pollutant sources of anthropogenic origin in the GMR are located in Europe. The existence of megacities (e.g., Istanbul, Turkey; Cairo, Egypt) and other smaller urban conglomerates (e.g., Athens, Greece; Rome, Italy; Barcelona, Spain), industrial activities, and energy production/consumption in the GMR result in elevated emissions of several pollutants such as nitrogen oxides, sulfur oxides, carbon monoxide, nonmethane hydrocarbons, ammonia, and so on. Figure 3 illustrates the emission pattern for some pollutants for the GMR and Africa that are considered important for the photochemical cycles of the PM productivity. The emissions inventory was provided by the Cooperative Program for Monitoring and Evaluation of the Long-Range Transmission of Air Pollutants in Europe (EMEP) and the Global Emissions Inventory Activity (GEIA) database. It is evident that the sources of anthropogenic pollutants are located mainly in central and southern Europe, and there is a minor contribution from northern Africa.

The anthropogenically produced pollutants may result in the production of secondary pollutants like ozone and PM of various sizes and properties (e.g., sulfates, nitrates). In addition to the anthropogenic PM, another factor contributing to PM concentration is the marine environment and the production of salt spray and dimethylsulfide (DMS; Kouvarakis and Mihalopoulos 2002; Herut et al. 1999). The North African region is responsible for the mobilization and transport of Saharan dust, which is considered of major importance to the total PM loading in the GMR (Rodriguez et al. 2001).

# *d. Paths and scales of transport and transformation of air pollution*

The paths and scales of air-pollution transport and transformation have been the subject of various studies during the last two decades. Table 1 summarizes the most important projects carried out in the GMR during this period and defines project acronyms. Projects such as MECAPIP (Millán et al. 1996; Salvador et al. 1997), SECAP (Millán et al. 1997), T-TRAPEM (Kallos et al. 1997; Kotroni et al. 1999; Peleg et al. 1997), and MEDCAPHOT-TRACE (Ziomas 1998; Ziomas et al. 1998) provided initial information about the recirculation mechanisms, layering, paths, and transformation processes, mainly of the photooxidants. The mechanism of Saharan dust transport toward the GMR and Europe was the subject of other projects such as MEDUSE (Söderman 1998). This combined effort continued in the framework of the BEMA (Kesselmeier et al. 1996; Seufert 1997), MAMCS (Pirrone et al. 2003), SUB-AERO (Lazaridis et al. 2006) and ADIOS (Love-Pilot and Benyahya 2003; Kallos et al. 2004) projects. The identified paths and scales of air-pollution transport are



FIG. 3. Emissions of anthropogenic and natural pollutants on an hourly basis, as produced from the EMEP ( $16 \times 16 \text{ km}^2$ ) (Z. Zlatev 2003, personal communication) and GEIA ( $1^\circ \times 1^\circ$ ) database (online at http://www.geiacenter.org/).

mainly from southeastern Europe and the Black Sea toward the Middle East and Africa, across the Aegean Sea. From the western to the eastern Mediterranean region, the transport pattern has two branches: toward the northeastern Mediterranean and toward the southeastern Mediterranean, the Middle East, and Africa. These paths are described extensively in Kallos et al. (1998a).

During the cold period of the year, washout mechanisms are important. The photochemical processes are not at their peak because of limited insolation and cloud formation. During the warm period of the year, the wet-removal processes are very limited and strong insolation promotes the photochemical processes (Varinou et al. 1999; Varinou 2000; Kotroni et al. 1999). The photochemical processes result in the appearance of high ozone concentrations in the vicinity of urban areas as well as in remote locations (Pilinis et al. 1993; Peleg et al. 1997; Kouvarakis et al. 2000; Wagner et al. 2000; Zerefos et al. 2002). As discovered during the

| TABLE 1. The EU | project acronyms an | id representative | references. |
|-----------------|---------------------|-------------------|-------------|
|-----------------|---------------------|-------------------|-------------|

| MECAPIP         | Mesometeorological Cycles of Air Pollution in the Iberian Peninsula (1988–91) (Millan et al. 1996;  |
|-----------------|---|
| SECAP           | South European Cycles of Air Pollution (1992–95) (Millan et al. 1997)   |
| MEDCAPHOT-TRACE | Mediterranean Campaign of Photochemical Tracers-Transport and Chemical Evolution (1994) (Ziomas 1998; Ziomas et al. 1998)   |
| T-TRAPEM        | Transport and Transformation of Air Pollutants on East Mediterranean (1992–95) (Kallos et al. 1997;<br>Kotroni et al. 1999)   |
| BEMA            | Biogenic Emissions in the Mediterranean Area, Phase I (1993–95) and Phase II (1996–98) (Kesselmeier et al. 1996; Seufert 1997)  |
| MEDUSE          | Monitoring and Prediction of the Atmospheric Transport and Deposition of Desert Dust in the Mediterranean Region (1996–98) (Söderman 1998)                                |
| MAMCS           | Modeling the Atmospheric Mercury Cycles (1997–99) (Pirrone et al. 2003)   |
| SUB-AERO        | Subgrid-Scale Investigations of Factors Determining the Occurrence of Ozone and Fine Particles (2000–02) (Lazaridis et al. 2006)  |
| ADIOS           | Atmospheric Deposition and Impact of Pollutants, Key Elements and Nutrients on the Open<br>Mediterranean Sea (2000–03) (Loye-Pilot and Benyahya 2003; Kallos et al. 2004) |
| MERCYMS         | An integrated approach to assess the mercury cycling in the Mediterranean basin (2002–05) (Pirrone 2004; Voudouri et al. 2005)  |
| MINOS           | Mediterranean Intensive Oxidant Study (2001) (see special issue of <i>Atmospheric Chemistry and Physics</i> , 2003, vol. 3)   |
| PAUR I          | Photochemical Activity and Solar Ultraviolet Radiation (1996-97) (Zerefos et al. 2002)  |
| PAUR II         | Photochemical Activity and Solar Ultraviolet Radiation (1998-2000) (Zerefos et al. 2002)  |

T-TRAPEM project (see references related to its activities), ozone plumes can travel long distances over the Mediterranean Sea and affect remote locations. Ozone and aerosols exhibit similarities in their longrange transport patterns. The air quality in the GMR can generally be characterized by the multiscale transport and transformation processes. The knowledge gained from the aforementioned projects gave inspiration to study in depth the production and transport patterns of PM in the GMR.

The work performed in the framework of the research activities mentioned above is summarized in Table 1. Most of this work and analysis has been performed with the aid of the RAMS (Cotton et al. 2003), Hybrid Particle and Concentration Transport (HY-PACT) (Tremback et al. 1993; Lyons et al. 1994), Comprehensive Air Quality Model with Extensions (CAMx) (Environ 2003), and "SKIRON"/Eta (Nickovic et al. 2001) advanced modeling systems. These modeling systems have been improved and validated against observations collected from the extensive experimental campaigns carried out in the GMR. The scales and paths identified so far are summarized in Fig. 4. In this figure the blue, greenish, and gray arrows indicate the transport paths of anthropogenic pollutants in the GMR. The gray arrows demonstrate mostly upperlayer transport, and the blue and greenish arrows indicate the movement of pollutants in the lower-tropospheric layers. The red-brown arrows indicate transport of desert dust from Africa in the lower troposphere. The following specific characteristics are shown.

- Transport of air masses from southern Europe toward the Mediterranean Sea, the Middle East, and North Africa occurs during all seasons, with the summer being the most efficient (transport paths A, B, and C in Fig. 4).
- Air quality at various locations in the GMR, especially near the coast, is defined mainly by the thermal circulations (diurnal cycle) (e.g., paths M1, M2, and M3 in Fig. 4), although the long-range-transport component is also significant.
- Venting of urban or industrial plumes located near the coastal zone occurs toward the free troposphere with the aid of upslope flows during the daytime and toward the marine boundary layer (MBL) where they are trapped and travel long distances until they reach land.
- Transport over the Mediterranean Sea occurs mainly within the MBL. Polluted air masses from the MBL are injected into the free troposphere, and in several locations the existence of islands acts as chimneys, contributing substantially to the described behavior.
- Some locations in the GMR act as "temporal reservoirs" where air pollutants are "concentrated" and "aged" before they are readvected (e.g., the Black Sea and the eastern Mediterranean region; path C in Fig. 4).
- In the western Mediterranean region, vertical transport is considerable and leads to multiple layering (path M1 in Fig. 4); in the eastern part, the horizontal component of transport dominates.
- In general, the time scale of transport of air masses



FIG. 4. Characteristic paths and scales of transport of air masses in the Euro-Mediterranean region. The blue and greenish arrows (A, B, C, and D) indicate transport paths of anthropogenic pollutants in the Euro-Mediterranean region (ozone, sulfates, etc.) in the lower troposphere. The gray arrows are associated mostly with the upper-layer transport of anthropogenic pollutants; in particular, the paths denoted as M1, M2, and M3 correspond to the thermal recirculation patterns in the area. The red-brown arrows indicate transport of desert dust from the Africa region in the lower 5 km of the troposphere.

from Europe toward the Middle East is approximately 2–3 days (transport paths A, B, and C in Fig. 4). The transport from the western part of the Mediterranean toward southeastern Europe has a range of 1–2 days. The transport from the western part of the GMR toward the Middle East and northeastern Africa is, in general, longer (3–4 days) (transport path A in Fig. 4).

- The air quality in urban areas of southeastern Europe, North Africa, and the Middle East is affected significantly by the long-range transport patterns described. This is due to the fact that the time scales involved in these patterns are still within the life span of most of the air pollutants.
- During summer, the ITCZ is located over northern Africa, south of the Mediterranean coastline (25°–30°N), over southern Libya and Egypt, where there are some strong convergence lines.
- Air masses from Europe should reach the midtropospheric layers of the ITCZ within a time frame of a few (4–6) days. This results in a massive upward transport of various aged pollutants.
- Mixing these aged pollutants with dust particles present in this area can produce new types of particle formations.

The transport patterns and characteristics of the GMR that have been discussed here often lead to high PM values in several locations. These values cannot be attributed solely to anthropogenic activities, but also are attributed to the contribution from and possible convolution between anthropogenic and natural PM. The impact on air quality in urban areas resulting from increased PM values has relevance for EU environmental policy, and these implications will be discussed later.

## e. Transport patterns from/to the Euro-Mediterranean region

As was discussed in the previous section, long-range transport of pollutants of anthropogenic and/or natural origin can contribute significantly to air-quality degradation in various regions. Several studies and experimental campaigns in the past have shown evidence and speculation about long-range transport paths on the transcontinental scale. Such paths are summarized in Table 2. The first transport path, from Europe to Africa, has been well documented in past studies and is summarized in the previous section and in Table 1. Indicative references are listed in Table 2. The second type of transport is from Europe toward the Atlantic Ocean and North America and is considered to be an

TABLE 2. Summary of transport paths from/to Euro-Mediterranean region and related references.

|   | Transport paths  | References  |
|---|--|---|
| 1 | From Europe toward Africa  | Kallos et al. (1997, 1998a,b); Luria et al. (1996); Millan et al. (1997, 2005); Duncan and Bey (2004) |
| 2 | From Europe toward Atlantic Ocean and North America                        | Not yet well documented   |
| 3 | From North America toward Europe   | Li et al. (2002); Derwent et al. (2004)   |
| 4 | From Asia to Europe  | Li et al. (2002); Lelieveld et al. (2002); Galperin and Sofiev (1998)                                 |
| 5 | From the sub-Sahel region to Europe and/or Atlantic<br>Ocean–North America | Thompson et al. (2001)  |
| 6 | From Saharan region toward the GMR, Europe, and Atlantic Ocean             | Kallos et al. (2005, 2006); Ozsoy et al. (2001); Nickovic et al. (2001)                               |
| 7 | From Indian Ocean toward North America and Europe                          | Ramanathan et al. (2001); Krichak et al. (2002); Lelieveld et al. (2002)                              |

extension of the previous path. This transport pattern has not yet been well documented. Preliminary results concerning particulate sulfate are discussed in the next sections of this paper.

The third path is from North America toward Europe. This path has been documented through experimental campaigns and modeling during the last two decades. This transport can occur over the North Atlantic Ocean with the aid of the North Atlantic Ocean anticyclone (see references in Table 2). The transport path mainly concerns PM of anthropogenic origin.

The fourth transport path is from Asia to Europe. Despite the fact that such transport is not common and is not considered to be responsible for severe airpollution episodes in Europe, it can occur under certain circumstances, and areas of eastern Europe can be influenced (see related references in Table 2). It is related mainly to transport of anthropogenic pollutants from old power plants and industrial sources in central Asia. In addition, dust is also transported from steppes in Asia during the summer months.

The fifth transport path is from the sub-Sahel region toward the GMR and/or the central Atlantic Ocean and North America. It is mainly related to biomass burning. As documented in the work of Thompson et al. (2001) and Prospero et al. (2001), this path is very effective toward the Atlantic Ocean and America with the aid of the predominant easterlies, but transport toward the GMR is highly unlikely.

The sixth path of transport is from the Saharan Desert toward the GMR. It is related mainly to the transport and deposition of Saharan dust. It can occur during all seasons. Almost every day there is Saharan dust transport somewhere in the GMR. Guerzoni et al. (1999) and Kallos et al. (2005) found that approximately  $10^8$  metric tons of dust are deposited annually over the Mediterranean Sea, and a similar amount is transferred toward Europe. Some representative publications are listed in Table 2.

The seventh transport path is related to potential transport of air pollutants (mainly from biomass burning) from the area of the Indian Ocean toward Africa, the GMR, and the Atlantic Ocean. Experimental results in the framework of the Indian Ocean Experiment (INDOEX) and MINOS projects (see references listed in Table 2) provided indications of such potential paths. According to this work, there is adequate evidence of trans-African transport in the upper-atmospheric layers with the aid of the easterlies. Nevertheless, there are doubts about the transport toward the GMR because the ITCZ acts as a northern barrier that prevents air masses from shifting northward.

In the next sections, recent findings concerning the transport and transformation of PM over the following five paths are discussed:

- 1) from Europe toward Africa,
- 2) from Europe toward the Atlantic Ocean and North America,
- 3) from the sub-Sahel region to Europe and/or the Atlantic Ocean–North America,
- 4) from the Saharan region toward the GMR, and
- 5) from the Indian Ocean toward North America and Europe.

The analysis of transport along these paths and the identification of characteristic spatial and temporal scales have been done with the aid of the SKIRON/Eta, RAMS/HYPACT, and CAMx modeling systems.

## f. PM levels and composition—EU policies

Air-quality standards in several European locations (mainly around the GMR) are frequently violated not only as a result of the anthropogenic activities but also as a result of the surges of Saharan dust that are superimposed. The EU Air Quality Directive 1999/30/EC states clearly that for several atmospheric pollutants the natural background levels may cause difficulties for several member states to meet the requirements of the limit values. Such problems have been already reported in all southern European countries (Rodriguez et al. 2001; Astitha et al. 2006). The implementation of the EU air-quality directives will require key legislative measures to be taken in member states as well as at the European level for reducing the risks associated with the exposure to high PM concentrations (especially "PM2.5") and the impact on human health and the environment. Similar problems are associated with extremely high ozone concentrations in southern Europe and the GMR.

## 2. Pollutants of anthropogenic and natural origin

As described in previous sections and with respect to the GMR, the most important sources of air pollutants of anthropogenic origin are located in Europe. Eastern European sources are considered to be significant because of the lack of strict emission caps and because of their location. Therefore, the transport from eastern Europe toward the GMR and North Africa is significant. This path follows the trade winds system that results in the transport of air masses from north to south. This is especially true during the warm period of the year when the trade system of the "etesian winds" dominates over the area.

A characteristic pattern that lasts for most of the days during the warm period of the year is the transport of air pollutants from eastern European sources toward the Black Sea or the Balkan Peninsula and then over the Aegean Sea (and secondarily over the Adriatic and the Ionian Seas) toward North Africa and the Middle East. Such transport had been initially described in Kallos et al. (1998b). This transport exhibits some unique characteristics. Some of them are illustrated with the aid of the RAMS/HYPACT modeling system (Pielke et al. 1992; Cotton et al. 2003; Tremback et al. 1993; Lyons et al. 1994). The case selected for analysis involves the summer of 1995 (Varinou 2000). Lagrangian particle releases were performed from the locations of major power plants in the vicinity of the Black Sea. As is illustrated in Figs. 5a and 5b, the general pattern of transport is over the Aegean Sea and Asia Minor, following the typical flow pattern of the season [etesian winds; see Kallos et al. (1993) and references therein]. The vertical mixing along this path exhibits some unique features. Figures 5c and 5d illustrate the vertical projection along the north-south axis for the boxes marked in Figs. 5a and 5b. During the daytime, boundary layer growth over land extends to several hundreds of meters. This is also evident over the Aegean Sea where the islands act as chimneys, transferring polluted air masses from the MBL upward (Fig. 5c). During the

night, most of the transport occurs within the MBL (around 300 m deep), as is illustrated in Fig. 5d.

Because the GMR exhibits the unique regional characteristics described in previous sections, it is considered to be important to identify the potential paths of transport from sources located thousands of kilometers away. Equally important is the transport of pollutants outside the GMR. Kallos et al. (2004) made the first attempt to identify such transport patterns of PM with the aid of the Lagrangian dispersion system RAMS/ HYPACT. The model simulations performed during the summer period were several days in duration. The domain extended from the Indian Ocean to the Atlantic Ocean and from the equatorial zone to the polar region, with a resolution appropriate for meso- $\alpha$ -type simulations ( $25 \times 25 \text{ km}^2$ , 33 vertical layers up to 17 km). The model configuration for these simulations is summarized in Table 3. Lagrangian particle releases were performed from key regions known for their unique characteristics and processes. The specific regions selected are

- the Indian Ocean, because it is known for the problems caused by biomass burning in the region and the production of large amounts of aerosols,
- the sub-Sahel region, known for biomass burning and aerosol production,
- the Iberian Peninsula, known for air-pollution production and transport (multiple layering and transport toward the Atlantic Ocean, the Mediterranean Sea, and/or Europe), and
- central and eastern Europe because the air-pollutant sources in the region are relatively unrestricted and result in high emissions.

The intention is to identify potential paths of transport over long periods of time (longer than the synoptic scale) from the areas under consideration. For this reason, the wet-removal processes were not considered in these dispersion simulations. The type of area source selected for each simulation was a rectangular box of 400-m height situated at 100 m above ground level. Continuous emissions of a passive tracer were used, setting the emission box in the four different locations mentioned above and performing one simulation at a time. The horizontal dimensions of each emission box were 1650  $\times$  550 km<sup>2</sup> for Africa and Europe, 1000  $\times$ 550 km<sup>2</sup> for the Iberian Peninsula, and  $200 \times 1600 \text{ km}^2$ for the Indian Ocean. Several simulations have been performed, each for several days. Indicative results are discussed for the simulation period of 1-10 August 2001. The weather patterns during the warm period of the year, as described in section 1b, are very common and persistent for a period of 3-4 months. Photochemi-



FIG. 5. Horizontal projection of particle position from Lagrangian releases in various locations around the Black Sea for 4 Jul 1995 during the (a) day and (b) night hours. Vertical projection (north–south) of particles located within the box marked in (a) and (b) for 4 Jul 1995 during the (c) day and (d) night hours.

cal processes are at their peak and complex thermal circulation patterns are important during the summer period. When the particle release is from central and eastern Europe, the dispersion pattern after 10 days of continuous release is illustrated in Fig. 6a. In Fig. 6a, two paths are clearly shown—one path is toward Asia following the westerlies in the area, and the other is along the well-defined path toward the Black Sea, the Balkans, the eastern Mediterranean, and northeastern Africa where the released particles entered the ITCZ zone and started being transported westward toward the Atlantic Ocean. These two paths have been illustrated with the letters A and C in the conceptual diagram of Fig. 4.

The particle-release experiment from the Iberian Peninsula for the same simulation period shows the pattern illustrated in Fig. 6b. Poor dispersion conditions prevent a portion of the particles from moving quickly

|                          | RAMS   |  |  |
|--------------------------|--|--|--|
| Input data               | ECMWF initial and lateral boundary conditions $(0.5^{\circ} \times 0.5^{\circ})$ on 12 isobaric levels; ground and upper-air observations assimilated; topography $(30'' \times 30'')$ : U.S. Geological Survey (USGS) dataset; vegetation $(30'' \times 30'')$ : USGS dataset; sea surface temperature $(1^{\circ} \times 1^{\circ})$ |  |  |
| Horizontal resolution    | $25 \times 25 \text{ km}^2$  |  |  |
| Domain dimensions        | 13 725 km $\times$ 7475 km horizontal  |  |  |
|                          | 17.32 km vertical  |  |  |
| No. of vertical layers   | 33 (terrain following)   |  |  |
| Microphysics             | Full microphysics scheme with eight categories of hydrometeors   |  |  |
| Nudging                  | Nudging only on lateral boundaries every 3 h   |  |  |
| Simulation period        | 1–10 Aug 2001  |  |  |
|                          | НҮРАСТ   |  |  |
| Type of area source      | Rectangular box  |  |  |
| Box position for release | 100–500 m  |  |  |
| Emissions                | Continuous, inert species  |  |  |
| Horizontal dimension of  | $1650 \times 550 \text{ km}^2$ for Africa and Europe   |  |  |
| area sources             | $1000 \times 550 \text{ km}^2$ for Iberian Peninsula   |  |  |
|                          | $20 \times 1600 \text{ km}^2$ for Indian Ocean   |  |  |
| Simulation period        | 1–10 Aug 2001  |  |  |

TABLE 3. Configuration of RAMS/HYPACT modeling system.

away from the source area. The path of transport toward central and eastern Europe is clearly dominant (path D in Fig. 4). The other path toward the central Mediterranean and North Africa and the ITCZ zone is also clearly defined (path A in Fig. 4). The third dispersion experiment was performed with particle release from the sub-Sahel region where intense biomass burning occurs during the dry period of the year. As shown



FIG. 6. Horizontal projection of particle positions after 10 days of continuous release from various locations (10 Aug 2001) for emission source boxes located in (a) central Europe, (b) the Iberian Peninsula, (c) the sub-Sahel region of Africa, and (d) the Indian Ocean. The simulations were performed with the use of the RAMS/HYPACT modeling system.

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| Input meteorological data | 3D gridded fields from RAMS model: horizontal wind components, temperature, pressure, water vapor, vertical diffusivity, cloud and precipitation water content, cloud optical depth |
|---------------------------|---|
| Input air-quality data    | Gridded initial concentrations  |
|                           | Gridded boundary concentrations   |
|                           | Time/space constant top concentrations  |
| Input emission data       | Gridded area sources from EMEP database [refined emissions $(16 \times 16 \text{ km}^2)$ ]  |
|                           | Gridded area sources from GEIA [global emissions inventory $(1^{\circ} \times 1^{\circ})$ ]   |
| No. of vertical layers    | 22 (up to 8 km)   |
| Horizontal resolution     | $20 \times 20 \text{ km}^2$   |
| No. of grid points        | $430 \times 275$  |
| Chemical mechanism        | Carbon Bond-IV with full aerosol chemistry [Aerosol Thermodynamic Model (ISORROPIA),<br>Regional Acid Deposition Model, and Secondary Organic Aerosol (SOAP)]                       |
| Simulation period         | 1–10 Aug 2001   |

TABLE 4. Configuration of CAMx air-quality model.

in Fig. 6c, the transport pattern is toward the Atlantic Ocean following the flow field defined by the easterlies. The dispersion pattern indicates a path of transport along the northwest African coast, toward the western Mediterranean region and the Iberian Peninsula. This path depends mainly on the position and strength of the North Atlantic Ocean (Azores) anticyclone and the Icelandic low pressure center. It can become considerable under certain circumstances, and it can also transport Saharan dust as illustrated in Fig. 4. This will be discussed later.

The last simulation experiment of transport with temporal scale longer than the synoptic was performed to illustrate the paths of the air masses flowing away from the Indian Ocean where intense biomass burning and/or anthropogenic pollution from the neighboring Asian countries (e.g., Indonesia, India, and Pakistan) can create high aerosol concentrations. The simulation is performed during the summer period when monsoon activity is common in the vicinity of the Indian Ocean. The path followed by the released Lagrangian particles is upward (with the aid of the strong updrafts in the area of deep convection) and toward the west (with the aid of the strong easterlies), as is illustrated in Fig. 6d. Of course, the washout mechanisms are strong in this region, but the purpose of the simulation is to illustrate the potential paths of aerosols and not to estimate their concentration and deposition rates.

According to the results of the aforementioned simulations (and especially the latter two) as well as others performed in the framework of this analysis, it is suggested that the position and the strength of the ITCZ over North Africa prevent the transport of aerosols from the African continent and the Indian Ocean region toward the GMR and Europe. This is especially true during the warm period of the year. The opposite is true for the transport from these regions toward the Atlantic Ocean, especially along the equatorial zone. In the work of Lelieveld et al. (2002) it is stated that transport of aerosols from central Africa and the Indian Ocean toward Europe is possible, but neither their evidence nor our findings are in favor of such path. Nevertheless, this needs detailed analysis based on both experimental and modeling evidence. It is worth mentioning that, from work performed in the past by various researchers and the authors of this paper, the aerosols produced in Europe are transported southward, mainly within the boundary layer, and eastward, mainly along paths in the middle troposphere. The sulfate amounts recorded in the GMR cannot be produced only from local sources; hence, there must be a significant contribution from long-range transport. This will be discussed further in the next section.

#### a. Production and transport of particulate sulfate

As discovered in the past (Katsoulis and Whelpdale 1990; Luria et al. 1996; Mihalopoulos et al. 1997; Kouvarakis and Mihalopoulos 2002), long-range transport is at least partially responsible for the large amounts of sulfates observed in the eastern Mediterranean during the warm period of the year. According to this earlier work, a path of the long-range transport has been identified from eastern Europe toward the Middle East over the Aegean Sea. Because the lifetime of particulate sulfate is very long, simulations were performed with the aid of the CAMx photochemical model (Environ 2003) over a large domain. CAMx was driven with meteorological fields retrieved from the RAMS. Both models have been configured over a large domain that covers the area from the Mid-Atlantic Ocean to India and from the equator to northern Europe. CAMx was configured with a horizontal grid resolution of 20 km and 22 vertical layers up to 8 km. The detailed CAMx configuration is listed in Table 4. The RAMS configuration is as described in Table 3.

The emissions inventory used was an ensemble of EMEP emissions (16 km  $\times$  16 km grid resolution) for Europe with GEIA emissions (1°  $\times$  1°) for the other



FIG. 7. Validation of the model results (CAMx) against observations for fine particulate sulfate for August 2001. The station is located at Finokalia, Crete (25.67°N, 35.32°E).

areas. The simulation period was 1-10 August 2001, and the full aerosol chemistry was applied in CAMx (Nenes et al. 1998, 1999; Chang et al. 1987; Gery et al. 1989; Strader et al. 1998). The model results have been compared with available observations in the GMR. No observations from other locations were available for this period. Several sensitivity tests and adjustments have been made before the final simulation. Figure 7 illustrates the modeled and monitored particulate sulfate concentrations in Finokalia. Crete, for the simulation period. The underestimation observed during the third day of the simulation can be attributed to the intense forest fires that occurred in the eastern part of Greece during the two previous days. In general, the model performed reasonably well, with an overestimation during the last 3 days of the simulation. Several possible reasons are suggested for the overestimation, including model resolution (20 km), which is not adequate to capture the local meteorological features, and the nearby emissions from the city of Heraklion, which were not treated in an appropriate way (a power plant is situated on the coastline approximately 70 km to the

west of Finokalia, near Heraklion). Details on the meteorological conditions encountered at this ground station are provided in Mihalopoulos et al. (1997) and Kouvarakis et al. (2000).

Because our intention is to provide the major characteristics of the production and paths followed by particulate sulfate, 3D plots have been prepared. Figure 8 shows the areas where anthropogenically produced sulfates exceed 6  $\mu$ g m<sup>-3</sup> during the 5th and 10th day of the simulation. Two major paths of transport are clearly indicated. One path is from Europe toward the GMR initially and then either toward the Atlantic Ocean through the Gibraltar Straits or toward the eastern Mediterranean and North Africa entering the ITCZ zone. Another important transport path of sulfates is from the Persian Gulf and the Arabian Peninsula toward Egypt and the Saharan region with the aid of the easterlies. These patterns are discussed below, because they were not analyzed in detail previously.

Particulate sulfate concentrations were integrated in the atmospheric column of the modeling domain (from the surface up to 8 km). The sulfate load in the atmo-



FIG. 8. Isosurface of fine particulate sulfate concentration (6  $\mu$ g m<sup>-3</sup>) for (left) 1200 UTC 5 Aug 2001 and (right) 1200 UTC 10 Aug 2001.

spheric column shows a pattern of the fine particulate sulfate transport in the GMR. Figure 9a clearly verifies transport path 2 of Table 2, in which sulfates produced mainly over the western Mediterranean are transported to the southwest through the Gibraltar Straits with the aid of the trade wind system dominant in this region during this season. The other common branches of the general path from north to south in the GMR are evident during all days of the simulation as shown in Fig. 9. Sulfates originating from sources in central Europe move toward the eastern Mediterranean and then North Africa (transport path 1 in Table 2). This pattern was evident during the entire simulation period, mostly as a result of the climatic patterns already discussed for the summer in the GMR (etesian winds over the Aegean Sea).

Another important path of transport appeared in the eastern part of the domain where anthropogenic sulfates from the Middle East and the Arabian Peninsula were transported westward. The relatively high amounts of sulfates observed over this area have their origin in the Persian Gulf and in general the oil industry in the area. Another important source is the megacity of Cairo in Egypt, where the urban and industrial emissions are relatively unrestricted. Such patterns occur mainly in the midtropospheric layers because of the deep mixing when air masses pass over the heat sources of the Arabian and Erythraean Peninsulas where deep, dry convection dominates. This transport continues toward the west with the aid of the easterlies. Part of the sulfates found over the Middle East and the Red Sea have their origin in southern Europe, where they were produced several days prior and have been transported to the southeast.

The polluted air masses from the Middle East are mixed with polluted air masses from Europe at the ITCZ region. It is worth mentioning that the ITCZ is located over Africa at a latitude of about 25°–30°N during the summer season. The sulfates, which originate from both the Middle East and Europe, end up over the Atlantic Ocean because of the absence of wetremoval processes over the Saharan region. The position of the ITCZ during summer does not permit the reentrance of sulfates into the atmosphere over the GMR. Nevertheless, exceptions may occur under certain synoptic conditions, together with air masses rich in desert dust. The desert-dust outbreaks are discussed in the following section.

## b. Saharan dust transport patterns—Potential impacts

Mineral dust, produced by wind erosion over the arid and semiarid areas of North Africa, can be transported away to the Middle East, the GMR, Europe, and even into and across the Atlantic Ocean (Kallos et al. 2006 and references therein). This material transported away from its origin is considered as an important climate and environmental modifier (Alpert et al. 1998). As was estimated by Guerzoni et al. (1999) and recently by Kallos et al. (2005), almost 10<sup>8</sup> metric tons of dust are deposited annually over the Mediterranean Sea while the same order of magnitude is transferred and deposited over Europe. Almost every day there is an area over the Mediterranean Sea where Saharan dust is deposited with various consequences.

Dust particles modify the earth's radiation budget by absorbing and backscattering both the incoming solar radiation and the outgoing infrared radiation (Andreae 1996). In addition, they alter the cloud microphysical processes because they act as cloud condensation nuclei (CCN) and play a role in the neutralization of acid rain because of their pH (>7.0) (Hedin and Likens 1996). In addition to the long-range transport of dust particles, important nutrients are moved from their sources to other regions, causing a possible modification of the biogeochemistry of marine and terrestrial ecosystems (Martin and Fitzwater 1988; Ozsoy et al. 2001). For example, the deposition of North African dust on the Mediterranean Sea provides important nutrients, such as nitrogen species, phosphorus, and iron, that may enhance the marine productivity. Some summer algal blooms in the Mediterranean Sea can be explained by such Saharan dust deposition (Dulac et al. 1996; Markaki et al. 2003).

As determined from satellite observations, groundbased measurements, and operational modeling forecasts, there is a large seasonal variability of the dust mobilization that depends on the source characteristics as well as the global atmospheric circulation (di Sarra et al. 2001; Ozsoy et al. 2001). During winter and spring, the GMR is affected by two upper-air jet streams: the polar-front jet stream, originally located over Europe, and the subtropical jet stream, which is typically located over northern Africa. The combined effects of these westerly jets in winter and spring support the propagation of extratropical cyclones toward the east and southeast, resulting in dust-plume intrusion in the GMR. During summer the transported aerosols are almost 2 times as large as in winter (Luria et al. 1996; Husar et al. 1997), but the highest amount of dust transport is within the tropical easterly jet from Africa toward the tropical Atlantic Ocean, reaching the Caribbean Sea and North America (Perry et al. 1997; Karvampudi et al. 1999; Kallos et al. 2006).

Most of the Saharan dust events that transport significant amounts of dust toward the Mediterranean Sea



FIG. 9. Fine particulate sulfate concentration integrated in the 0–8-km vertical layer of the atmosphere (g  $m^{-2}$ ) for 1200 UTC (a) 4, (b) 5, (c) 6, (d) 7, (e) 8, (f) 9, and (g) 10 Aug 2001. The dates shown here are in accordance with the dispersion simulations for the warm period of the year, during which photochemical activity is considered high in the area and the weather patterns are very common and persistent for several months in the GMR.

and Europe occur during the low-index circulation period of the year [cold and transient seasons as described in Kallos et al. (1998a) and Rodriguez et al. (2001)] (transport path 6 in Table 2). Driven by the trade wind circulation, Saharan dust is often moved across the Atlantic Ocean (Karyampudi 1979; Perry et al. 1997; Karyampudi et al. 1999; Prospero et al. 2001). During such events, dust has been observed in Barbados (Li et al. 1996) and in the eastern and southeastern parts of the United States (Savoie and Prospero 1977).

Using trajectory analysis and a conceptual atmospheric circulation model, Perry et al. (1997) postulated that the easterly winds of the summer ITCZ in the Tropics can provide a mechanism for successful longrange transport across the Atlantic Ocean. Once in the vicinity of the American continent, the dust can be farther driven toward the southern and eastern United States as it enters into the zone of the semipermanent "Bermuda" high pressure center. This case has been successfully simulated by Kallos et al. (2006).

Aerosols of anthropogenic origin, such as particulate sulfate, coexisting with aerosols of natural origin, such as desert dust and/or sea salt, in relatively high moisture environments produce PM of different sizes and properties. This situation can result in significant disturbance of cloud and precipitation patterns (Levin et al. 1996; Rosenfeld 2000; Givati and Rosenfeld 2004). As was recently discovered by Levin et al. (2005), the mixture of Saharan dust, sea salt, and sulfates can create gigantic CCN and ice nuclei that can significantly affect the rain production from continental-type clouds.

Another important role of desert dust is the airquality degradation in urban areas. Rodriguez et al. (2001) found that the cities of the Iberian Peninsula are significantly affected by Saharan dust outbreaks. Analysis of PM records in Athens during the period of 2001-04 showed that there were many days (ranging from 140 to 220) for which at least 1 of the 17 monitoring stations had "PM10" concentrations that were above the EU-imposed limits. A systematic day-to-day analysis with the SKIRON/Eta dust forecasting system (Kallos et al. 2006; Nickovic et al. 2001), Sea-Viewing Wide Field-of-View Sensor (SeaWiFS), and Total Ozone Mapping Spectrometer satellite images showed that for 60%-70% of the cases a synergetic contribution of anthropogenic (urban and long-range transport) and natural (Saharan dust) sources is evident (Astitha et al. 2006). This finding is in agreement with the composition analysis of PM performed by Mihalopoulos et al. (1997) on the island of Crete and by Herut et al. (2001) in Israel, where the mixture of Saharan dust, long-range transport of PM, and sea salt coexists

and is mainly associated with southerly and southwesterly winds.

The coexistence of PM from both origins (anthropogenic and natural) is favored by the same type of synoptic weather: the formation of a southerly or southwesterly flow in the lower troposphere. This kind of synoptic flow is responsible for stabilizing the lower troposphere by transferring warm, dry air masses (continental-tropical type) and creating poor dispersion conditions in urban areas. At the same time, these air masses are rich in desert particles and, under favorable conditions, rich in sea salt.

To support such coexistence of PM in the GMR, a discussion of the desert-dust transport patterns for the experimental period of 1–10 August 2001 follows. Figure 10 illustrates the dust load for the second, third, fourth, and eighth day of the simulation. These figures have been produced with the aid of the SKIRON/Eta weather and dust prediction system. Plumes from both origins (anthropogenic and natural) coexist over the west Mediterranean and Europe. Such coexistence can create air-quality conditions that exceed the imposed air-quality limit values.

The SKIRON/Eta weather and dust prediction system has proved to be a very useful tool for predicting desert-dust outbreaks in the GMR and Europe. SKIRON/Eta was developed at the University of Athens by the Atmospheric Modeling and Weather Forecasting Group, in the framework of the SKIRON, MEDUSE, and ADIOS projects. The system was evaluated by utilizing various types of observations for specific episodes as well as for longer periods. Evaluation of the system was performed over the Iberian Peninsula by Rodriguez et al. (2001) and Escudero (2006). The same atmospheric and dust prediction system is in operation in Malta, where it is known as the Dust Regional Atmospheric Model (DREAM). This operational version of the SKIRON/Eta system was evaluated by comparing its forecasts with lidar observations in the framework of the European Aerosol Research Lidar Network (EARLINET) project (Balis et al. 2006). The same system has recently been in operational use in Barcelona under the same name (Perez et al. 2006).

The most complete dataset available for model validation was provided from B. Herut (2004, personal communication) for the location of Tel-Shikmona (near Haifa, Israel). A description of the area is provided in Herut et al. (2001). The evaluation was performed for 2001 and 2003. The dataset included particulate aluminum (a basic component of desert dust). The measured particulate concentration of aluminum (Al) in the air was used as a precursor of dust concentrations. The 1246



0.0 0.4 0.8 1.2 1.6 2.0 2.4 2.8 3.2 3.6 4.0 4.4 4.8 5.2 5.6 0.0 0.4 0.8 1.2 1.6 2.0 2.4 2.8 3.2 3.6 4.0 4.4 4.8 5.2 5. FIG. 10. Dust load for August 2001, from the SKIRON/Eta system.

total mass of Al accumulated on the filter was normalized per 1 m<sup>3</sup> of air. Therefore, the Al concentration can be considered as an average value for each specific time interval (B. Herut 2004, personal communication). The Al content in desert dust is approximately  $7.09\% \pm 0.79\%$  (Guieu et al. 2002). The calculated Saharan dust concentrations were directly compared with simulated dust outputs corresponding to the sampling dates. The one-to-one comparison of the available observations with the model estimates is not an easy task and contains several uncertainties. Nevertheless, the evaluation performed is illustrated in Fig. 11. The scatterplot in Fig. 11a illustrates that the model has a tendency to underestimate the higher observed values for 2001 because the regression line deviates from the diagonal that represents the best fit of the simulations to the data (Wilks 1995). The correlation coefficient r indicates the strength of the fitting between the simulated and measured values of dust concentration (higher correlation is achieved when r is closer to 1). The coefficient of determination  $R^2$  shows how closely the estimated values for the trend line correspond to the actual data (most reliable when it is close to 1). The correlation coefficient is high (0.78),



FIG. 11. Measured vs simulated dust concentrations during the recorded episodes at the monitoring site of Haifa, Israel, during (a) 2001 and (b) 2003;  $R^2$  is the coefficient of determination, and r is the correlation coefficient.

which can be attributed to the agreement of the lower observed values with the predicted ones. The  $R^2$  is considered to be reliable for both years, but for 2003 the comparison shows better results. This occurs because the measurements are 1.3 times as high as the simulated values, whereas for 2001 the measured values are 2.6 times the simulated values. In the comparison of modeled versus measured concentrations, the measurements with values lower than 0.01 mg m<sup>-3</sup> have been excluded because there is a high possibility for a contribution from local dust sources in the measurements.

The difference between 2001 and 2003 can be attributed to the fact that during 2001 the SKIRON/Eta Model used one particle size scheme (average diameter of the dust particles was centered at 1.5  $\mu$ m) whereas during 2003 the model was implemented with a foursize distribution scheme (centered diameters of 1.5, 12, 36, and 76 µm). A single-size dust scheme is not adequate, because the behavior of particles with diameters outside the 1.5- $\mu$ m bin could not be forecast. The mismatch is more serious for the lighter particles that play a major role in the levels of dust concentration, because they are the ones that usually manage to travel longer distances from the source areas before they are deposited (Tegen and Fung 1994; Perry et al. 1997). Also, as described above, the desert-dust concentration was calculated through empirical formulas and ratios between the measured quantities of various chemical elements (Al), introducing a number of errors in the process. Last, the contamination of the samples from anthropogenic and even local sources can have a considerable effect on the process of evaluation by altering the levels of measured dust concentrations. However, the underestimation in many cases is small and is acceptable for dust-forecasting purposes.

As Figs. 9 and 10 illustrate, the coexistence of dust plumes with sulfates is a common pattern in the GMR. The case selected for demonstration is during August of 2001 when Saharan dust intrusions are not as frequent as during the transient seasons (spring and autumn). Despite this fact, the coexistence of the plumes from different origins (North Africa and Europe) and compositions (mineral dust and industrial sources) is clearly evident. The consequences in this case are mainly associated with air-quality degradation ("exceedances" of imposed air-quality limit values).

## 3. Concluding remarks

The scope of this work was to summarize the existing knowledge concerning the long-range transport and transformation patterns of PM that have anthropogenic and natural origin in the GMR, to discuss some problems related to this coexistence, to illustrate the modeling capabilities, and to recommend future work in the field of atmospheric and air-quality modeling. The paths and spatiotemporal scales of transport of PM have been described in detail from previous work and current research. The general trend is the transport of anthropogenic PM from Europe and the Middle East toward the south, over the Mediterranean Sea, North Africa, and toward the Atlantic Ocean and North America following the easterlies. The natural PM originates mainly from the Saharan desert. The transport paths for dust are from the south toward the Mediterranean Sea and Europe, as well as toward the Atlantic Ocean. The transport from distant areas, well known for producing heavy amounts of PM, toward the GMR seems to be limited. The most favorable period for PM transport, from both categories of sources, is the warm period of the year, because of the prevailing synoptic and mesoscale circulations and the limited wet-removal processes. The transient seasons are the most favorable for Saharan dust transport toward Europe.

Photochemical processes can produce high amounts of anthropogenic PM, such as particulate sulfates that are transported over long distances—mainly toward the eastern part of the GMR, the Middle East, and North Africa. Violations of air-quality standards resulting from high PM concentrations in southern European cities are associated with Saharan dust transport episodes for 30%–70% of the cases. This transport is an important issue in many European cities, because it is difficult to meet EU air-quality standards not only for ozone but also for PM.

The impacts of aerosols on air quality and climate from each source (natural and anthropogenic) can be described with existing methodologies and techniques. Advances in modeling techniques and observations have led to the capability of describing and predicting such impacts. Nevertheless, the current status of the existing models is considered to be advanced but not adequate to describe accurately the complicated physicochemical processes taking place along the transport path in the GMR. Such processes are related to several factors:

- Heterogeneous chemical reactions, in which anthropogenic and natural PM play an important role in air-pollutant by-products.
- Aqueous-phase chemistry and cloud-interaction processes are critical and are able to affect even the water budget in the region. There is limited knowledge related to the role of each source of PM on cloud formation and evolution (e.g., impact of sea salt, sulfates, or mineral dust). The combination or coexistence of these PM in the wet environment is not understood well (Andreae et al. 2004; Levin et al. 2005; Rosenfeld 2006; among others).
- Radiative-transfer impacts and interactions appear to be poorly described, especially for naturally produced aerosols.
- The influence of marine types of aerosols (e.g., DMS or sea salt) on air quality has not been clarified yet.

Climate and air-quality feedbacks are not well understood, and hence future work requires specialized surface and upper-air measurements to explore the validity of the various model elements. Phenomena focused primarily on air-quality degradation (heterogeneous chemical transformations) and distortion of cloud, precipitation, and water budget of the area (suppression of precipitation or enhancement of torrential systems) are considered to be essential for the GMR. The effects of such phenomena need advanced modeling tools and observations that include at least the basic components for describing the processes involved in order to be applied in an integrated way.

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