PRODUCTION AND TRANSFORMATION OF ANTHROPOGENIC AND NATURAL AEROSOLS IN THE MEDITERRANEAN REGION

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ABSTRACT

Various research and operational activities related to air quality analysis and forecasting showed that European air quality standards related to particulate matter are often violated not only from the anthropogenic activities but from the contribution of natural sources. The main contribution of natural sources in such air quality degradation in Europe is from the transport of Saharan dust. Air quality is affected significantly by increasing the particle concentrations in the atmosphere and reducing visibility. Such an effect is also attributed to the fine particulate sulphates in the atmosphere. Since the long-range transported dust particles are mainly of the size of PM2.5 the impacts on health are considerable but most of them are still unknown. In general, aerosols of fine mode such as aerosol sulphates and desert dust, affect human health and welfare of hundreds of millions people in Europe and North Africa. In order to examine the degradation of the air quality due to the synergetic effects of desert dust and sulphates, simulations were performed with the aid of advanced regional atmospheric and photochemical models. In addition, the estimation of the SO_2 to sulphate ratio provides with qualitative information on the paths and scales of sulphate production, transformation and chemical age in the Mediterranean Region. The integrated approach followed in this work can provide useful information concerning the air quality degradation in urban environments as well as the Greater Mediterranean Region.

INTRODUCTION

In the present work an attempt was made to analyze and study the air quality degradation in the Mediterranean Region, due to both natural and anthropogenic aerosol influence. Levels of atmospheric aerosol are monitored in ambient air quality networks because of their potential impact on human health, visibility and climate. Various countries in the Mediterranean Region are not capable of meeting the imposed air quality standards, due to the non-controllable effect of natural aerosols in the atmosphere of the region, as well as to the characteristic climatic and flow patterns, the multi-scale transport and transformation processes that dominate the area. As it is suggested in previous studies [1, 2], the synoptic/regional circulation during summer, favours the long-range transport of air pollutants released from Southern and Eastern Europe and Central Mediterranean towards the Eastern Mediterranean, North Africa and Middle East. The main objective of this work is to identify the spatiotemporal patterns and scales of anthropogenic and natural aerosols in the Mediterranean Region, with the aid of advanced modelling systems. This is achieved by focusing in two sections: Aerosols of fine mode (sulphates) as anthropogenic inputs and Saharan dust as natural input in the atmosphere of the Mediterranean region.

A large amount of anthropogenically-generated aerosols, such as sulphates, contribute to the increased concentrations of particles in the atmosphere and to the reduction of visibility. This is attributed mainly to fine particles, which are capable of long range transport, influencing the air quality in remote locations. In addition to that, natural aerosols, having as main source of origin the Sahara desert, contribute significantly in the increase of particle concentration in the atmosphere. Sahara is the largest world desert and Europe is frequently exposed to large amounts of dust generated in intense dust storms. Therefore dust particles affect the air quality of a specific region mainly as episodic phenomena.

Just a decade ago, only a limited amount of aerosol observations were available. Due to the increased scientific and public interest to improve understanding of the aerosol cycle in the atmosphere and the effects on human and climate, several mathematical models were developed. The models are used for simulation and/or prediction of the atmospheric aerosol processes and until today they have achieved a level providing a closer insight to the entire atmospheric aerosol cycle. In addition to the above, a number of studies have focused on the long range transport of Saharan air masses over Western and Eastern North Atlantic areas [3], South America and the Mediterranean [4, 5]. Several studies in Europe and other parts of the world suggest that fine desert particles of the size around 2.5μm are a considerable portion of the entire dust production and can travel thousands of kilometres affecting remote locations [3, 6].

The work presented here, is a part of a larger effort devoted to contribute to a better understanding of simultaneous interactions between anthropogenically produced air pollution, naturally produced particles (dust aerosol) and the tropospheric environment, focusing on the Eastern Mediterranean. It is focused on the paths and scales of transport and transformation of SO_2 to particulate sulphate ratios over Southern Europe and Mediterranean. In addition, we examine naturally produced aerosols (Saharan dust) convoluted to the above, with the aid of advanced atmospheric and photochemical models.

MODEL DESCRIPTION

A short description of the modelling systems used for performing simulations is provided below.

The SKIRON/ETA is a modelling system developed at the University of Athens from the Atmospheric Modelling and Weather Forecasting Group [7, 8]. It is based on the ETA model. It has enhanced capabilities with the unique one to simulate the dust cycle (uptake, transport, deposition). It has been applied to the Saharan dust transport towards the Mediterranean Basin and USA as well as in other areas of the Earth.

RAMS (Regional Atmospheric Modelling System) is a highly versatile numerical code developed by scientists at Colorado State University and ASTER, Inc. for simulating and forecasting meteorological phenomena. It has been developed in order to simulate atmospheric phenomena with resolution ranging from tens of kilometres to a few meters. A general description of the model and its capabilities is given in [9].

The Comprehensive Air Quality Model with Extensions (CAMx) [10] is an Eulerian photochemical model that allows for integrated assessment of air-pollution over many scales ranging from urban to super-regional (http://www.camx.com). CAMx has also model structures for modelling aerosols, processes that are linked to the CB4 gas phase chemical mechanism. New science modules are introduced for aqueous chemistry (RADM-AQ) inorganic aerosol thermodynamics/partitioning (ISORROPIA) and secondary organic aerosol formation/partitioning (SOAP).

RESULTS AND DISCUSSION

Anthropogenic aerosols

The paths and scales of transport and transformation of air pollutants in the Mediterranean Region have been identified in previous projects [1, 2]. The results showed that the synoptic/regional circulation during summer, favours long-range transport of air pollutants released from Southern and Eastern Europe and Central Mediterranean towards the Eastern Mediterranean, North Africa and Middle East (Figure 1). PM2.5 particles (aerosols of fine mode) behave as gases in the atmosphere (weak gravitational settling). Therefore fine particles exhibit a long range transport pattern similar to that of gas pollutants such as ozone and NOx.

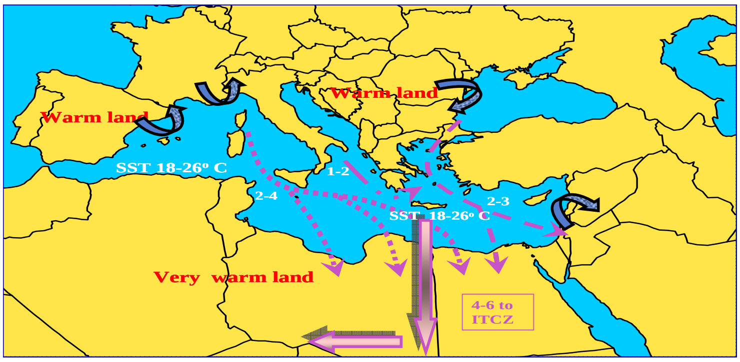

Figure 1: Characteristic paths and scales of transport of air masses in the Mediterranean Region.

The atmospheric/photochemical model simulations where performed with the aid of the RAMS and SKIRON/ETA modelling system in combination with CAMx photochemical model. The most recent version of CAMx (v4.03) was implemented specifically for the purpose of this work, including the newest modules for gas to particle and aqueous phase mechanisms (ISORROPIA).

In order to identify the paths and transformation of SO_2 to particulate sulphate, the sulphate ratio was calculated within the code of CAMx model. Sulphate ratio has been used in previous studies [11] in order to define the chemical age of an air mass, based on measurements of sulphur dioxide and particulate sulphate. Sulphate ratio is characterized as the ratio of sulphate concentration to total sulphur concentration (meaning both SO_2 and particulate sulphate), leading to a dimensionless value from zero to unity. According to Luria [11], the higher values for sulphate 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 its travel distance. CAMx model code was modified in order to calculate an average sulphate ratio for each hour of the simulation. The sensitivity tests followed in this work had a similar base case: zero values for every source of SO_2 emissions in the region, zero initial and boundary conditions. Only one point source is implemented in the domain (located near Athens, Greece), emitting SO_2 for only 1h. The preliminary results of this work are presented below.

The sensitivity tests are divided in two categories: a) Injection of SO_2 inside the Marine Boundary Layer (MBL), using a point source in a coastal area with releases near the surface (Figure 2). b) Injection of SO_2 in the free troposphere, using the same location for the point source (low velocity and temperature) (Figure 3). The release of SO_2 in the atmosphere, in both cases, occurs for 1h at 00UTC and as a second test, at 12UTC.

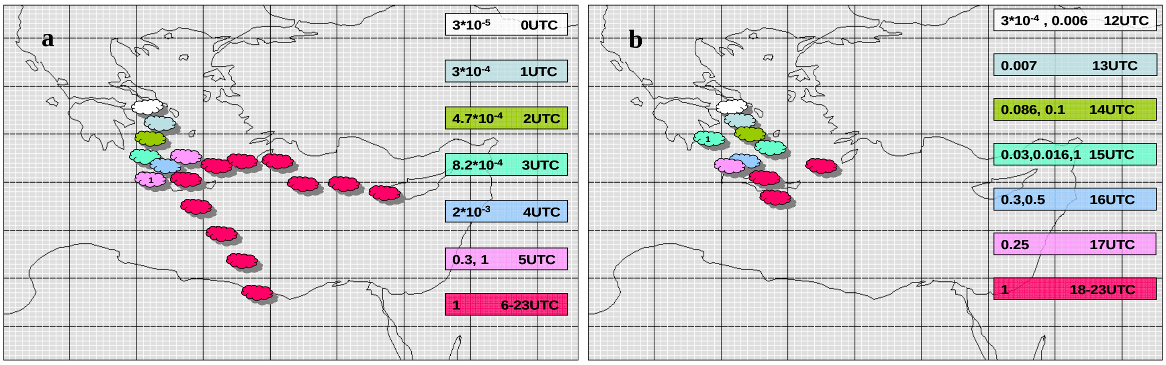

Figure 2: Sulphate Ratio for August 1, 2001; Layer 1: 0-50m. Point Source at Lavrio, located near the surface. a) SO2 emitted for 1h (00-01UTC). b) SO2 emitted for 1h (12-13UTC).

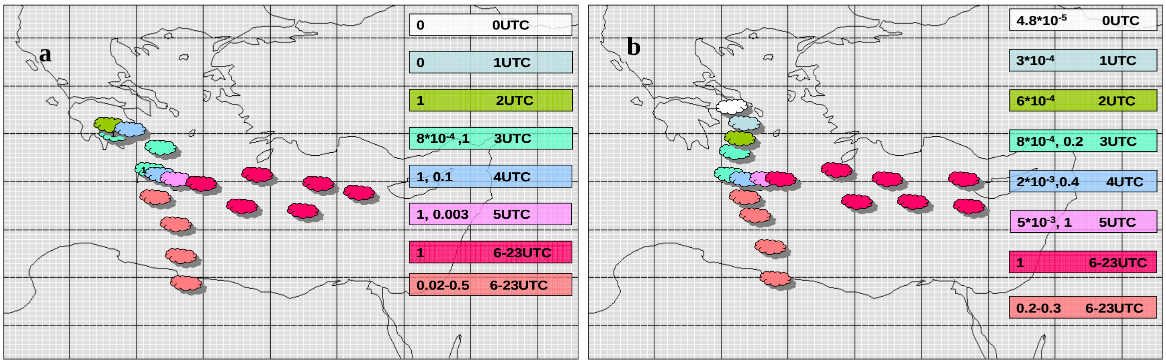

Figure 3: Sulphate Ratio for August 1, 2001; Point Source at Lavrio (emissions above the MBL). Release: 0-1UTC. a) Layer 1:0-50m. b) Layer 5: 300-450m.

The injection of hourly SO_2 emissions in the atmosphere of the Eastern Mediterranean Region showed some interesting results. When SO_2 is emitted inside the MBL, as in Figure 2, sulphate production is rather rapid due to the high moisture environment. When SO_2 is emitted in the MBL during night hours (Figure 2a), a major part of SO_2 converts into particulate sulphate within the first 5 hours (that is when sulphate ratio reaches unity). When the emission occurs during day hours (12UTC), a major part of SO_2 converts into sulphate within the first 3 hours (Figure 2b). When SO_2 is emitted in the free troposphere (Figure 3), the conversion of SO_2 to sulphate is also relatively fast. Looking at the layer where the injection of SO_2 in the atmosphere occurred (Figure 3b), the pattern is similar to that of the injection inside the MBL (Figure 2a), though in this case the air masses that reach the North African coast are considered less aged than before. Both SO_2 and particulate sulphate remain in the atmosphere for 2-3 days, until they have reached the northern coast of Africa and Eastern Mediterranean, where the modelling domain reaches to its end. In both sensitivity tests, the existence of islands in the area, contribute significantly in the perturbation and mixing of the air masses. By the time the air masses reaches Crete they are considered aged enough, and due to the long atmospheric lifetime of particulate sulphate the air masses continue their course towards the North African Coast and Middle East, according to the synoptic circulation of this particular time of the year.

This has been verified with measurements in Finokalia station, Crete. The intercomparison of model results with observations has not finished yet.

Natural Aerosols

In addition to the anthropogenic produced aerosols, such as sulphates, desert dust contributes significantly in the total amount of fine particles in the atmosphere, due to the episodic character of increased desert dust concentrations. The Saharan dust amounts over Mediterranean and Europe on seasonal timescale is examined with the aid of the most recent dust modelling system SKIRON. Since January 2000, the SKIRON/ETA model runs operationally covering the Mediterranean Region, providing 3-day forecasts of dust load and deposition (http://forecast.uoa.gr), among other meteorological parameters. Using the data available, a database of model-derived seasonal amounts of dust deposited on Mediterranean Sea and Europe has been created. The amount of Saharan dust deposited on the Mediterranean waters or over the European land exhibits significant seasonal and inter-annual variability [12], having a rather episodic character (Figure 4). The strength and the frequency of occurrence of the Saharan dust episodes define the annual deposition amounts and patterns of aerosols to a high degree, alternating the mean annual values. This leads to the fact that long-term modelling and measurement data are essential in understanding the synergetic effects of sulphates and desert dust in the atmosphere of the Mediterranean Region.

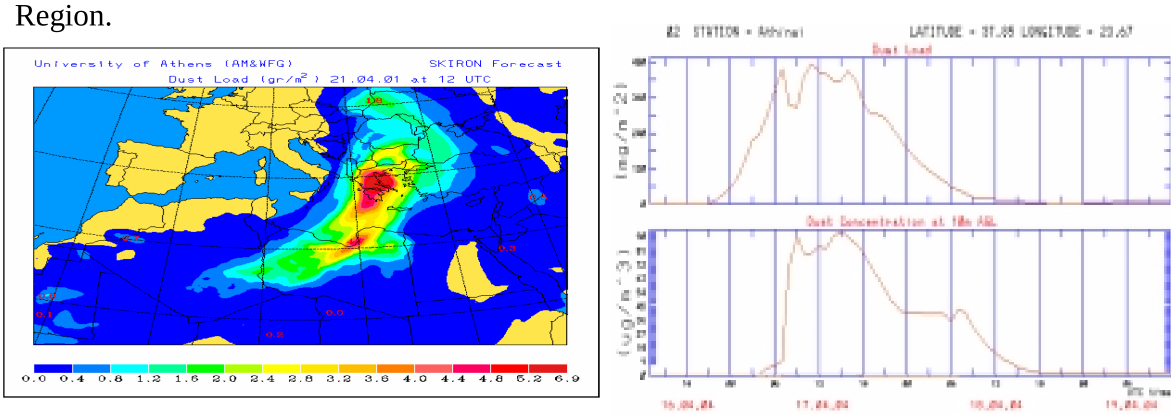

Figure 4: a) (left) Desert Dust Episode for April 21, 2001, at 12UTC, as simulated by SKIRON/ETA model. Dust Load (gr/m2), b) (right) Desert Dust episode for April 17, 2004, for the city of Athens.

CONCLUSIONS

Anthropogenic aerosol inputs as fine particulate sulphates play an important role in the air quality degradation. In this work an attempt was made to identify the paths and timescales of SO_2 to particulate sulphate conversion in the area of Eastern Mediterranean. This was accomplished by calculating sulphate ratio with the aid of a photochemical model. Sulphate ratio is considered an indicator of air mass age and chemical conversion efficiency, therefore the sensitivity tests performed for the purpose of this study showed some interesting results.

The conversion of SO_2 to particulate sulphate is less rapid and efficient when SO_2 is injected inside the MBL during the night hours, since the absence of sunlight for the first 5hours of the day slows down the oxidation of SO_2 to sulphuric acid and the nucleation of sulphuric acid with water. Sulphate ratio reaches higher values in a shorter timescale (3hours), when SO_2 is injected in the MBL during daytime (12UTC). During the day the photochemical processes enhance the oxidation of SO_2 to sulphuric acid and favour the formation of sulphuric acid vapour. In addition to the above, the high moisture environment (relative humidity reaches 90% in the area), leads SO_2 to rapid oxidation in the aqueous phase, formatting wet particulate sulphate. Low amounts of rainfall in the area during that day, might explain the high sulphate ratios due to the clearance of SO_2 from the atmosphere. When SO_2 is injected in the free troposphere during the night, the conversion to sulphate shows a similar pattern, though sulphate ratio reaches higher values faster (within 3hours). In both cases the air masses, consisting of SO_2 and particulate sulphate, continue their course towards the African coast and the Middle East for the next 2-3 days of the simulation.

The sensitivity tests performed in this work showed results similar to those of Luria [11], confirming the long range transport paths of sulphur towards the Middle East coast, during summer. In addition to the fine particulate sulphate paths and scales discussed above, desert dust episodes that govern the area of Eastern Mediterranean, contribute to the increase of fine particulate in the atmosphere, possibly increasing CCN (cloud condensation nuclei) and reducing visibility in the Region.

Acknowledgements

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