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March 29th, 2012

To whom it may concern

I would like to confirm that the paper "**Investigation of transport pathways and potential source regions of atmospheric aerosols in Belgrade: receptor modeling and LIDAR system**", authors: *Zoran Mijic, Darko Vasiljevic, Aleksander Kovacevic, Bratimir Panic, Milan Minic, Mirjana Tasic, Branislav Jelenkovic, Ilija Belic, Ana Vukovic* was an invited lecture of the 5th Workshop on Optoelectronic Techniques for Environmental Monitoring-OTEM 2011 which took place during 28th to 30th of September, 2011 at the "Romanian Atmospheric Observatory", Magurele, Romania.

Yours sincerely,
Director of the organizing committee
Dr. Doina Nicolae

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Investigation of transport pathways and potential source regions of atmospheric aerosols in Belgrade: receptor modeling and LIDAR system

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Abstract: The transport pathways and potential source regions of PM_{10} were investigated based on backward trajectories and PM_{10} concentrations measured in Belgrade from 2004 to 2008. A study of airflow characteristics was performed using cluster analysis of 48-hour backward trajectories of air masses arriving at six different vertical levels above Belgrade, Serbia. Airflow directions were grouped into five classes indicating typical origin of air masses. The results suggest that the highest PM_{10} concentration was related to the west southwest pathway. Potential source contribution function (PSCF) and concentration weighted trajectory (CWT) models were used for identification of source regions. Long – range transport from west and southwest pathway was evident and sporadically (mostly in spring and summer) associated with African dust outbreaks. Case study of elastic backscatter LIDAR observation and complementary prediction of vertical concentration profiles of Saharan dust aerosols by the DREAM model were also carried out.

Keywords: Aerosols, Transport pathway, Potential source contribution function (PSCF), Concentration Weighted Trajectory (CWT), LIDAR

1 Introduction

Atmospheric aerosols are of major scientific interest due to their confirmed role in climate change [1] and their effect on human health [2] and local visibility. In order to protect public health and the environment i.e. to control and reduce particulate matter (PM) levels, air quality standards were issued and target values for annual and daily mean PM_{10} (particles below 10 μm in diameter) mass concentrations were established. The studies of the transport and mobilization of trace metals up to now have attracted attention of many researchers. Within the European Program for Monitoring and Evaluation of the Long-Range Transmission of Air Pollutants (EMEP), measurements of PM_{10} and trace metals, as highly toxic species, have been introduced. Studies of spatial and temporal variation of atmospheric aerosol particles also gained in significance and resulted in an increased interest in the use of analytical techniques capable to measure the size, morphology and chemical composition of individual aerosol particles. Such data are essential for an understanding of particle formation, transport, transformation and deposition mechanisms, as well as the impact of particles inhaled by a respiratory system. One of the main difficulties in air pollution management is to determine the quantitative relationship between ambient air quality and pollutant sources. In the field of atmospheric sciences receptor models aim to re-construct the impacts of emissions from different sources of atmospheric pollutants based on ambient data measured at the monitoring sites [3]. These methods are called receptor-oriented or receptor models since they are focused on the behavior of the ambient environment at the point of impact, as opposed to the source-oriented dispersion models that focus on the transport, dilution and transformations that begins at the source and continue until the pollutants reach the sampling or receptor site. The problem is, using the data measured at the receptor site alone, to estimate the number of sources, to identify source composition and most importantly, from a regulatory point of view, to asses the source contributions to the total mass of each sample. The two main extremes of receptor models are chemical mass balance (CMB) and multivariate models.

One of the main differences between models is the degree of knowledge required about the pollution sources prior to the application of receptor models. For pollutant sources that are unknown, hybrid models that incorporate wind trajectories Potential Source Contribution Function (PSCF) [4] and Concentration Weighted Trajectory (CWT) [5] can be used to resolve source locations. In this paper these models were applied on originally PM_{10} data set from Belgrade in order to provide a map of source potential of geographical areas. Statistical analysis of air mass back trajectories combined with long-term ambient air pollution measurements are useful tools for source identification [6]. In addition, transport paths of air masses with similar history and origin were obtained by trajectory cluster analysis.

The Mediterranean region and particularly the Balkan Peninsula have been under the influence of Saharan dust transport and deposition over millennia. Identification of the concentration, composition, origin, transport and geographical distribution of PM in Mediterranean atmosphere has been the subject of research activities since the last two decades as it is heavily affected by two contrasting sources; mineral dust (mainly from Sahara Desert) [7] and various anthropogenic (from industrialized/semi-industrialized countries) emissions. However, in the Mediterranean region relatively limited research has been carried out on the identification of potential source regions of aerosol components in particulate matter and a few comparisons among the various source apportionment techniques have been performed [8]. Within the EARLINET (European Aerosol Research LIdar NETwork) [9] particular attention is devoted to Saharan dust transport events monitoring. The first case study LIDAR measurement of Saharan dust events over Belgrade will be shown related to DREAM (Dust REgional Atmospheric Model) model prediction.

2 Methodology

Air masses back trajectories were computed by the HYSPLIT (HYbrid Single Particle Lagrangian Integrated Trajectory) model [10] through interactive READY system [11]. Daily 48-h back trajectories, started from the center of Belgrade ($44,804^\circ$ $20,478^\circ$) at 12:00 UTC each day, were evaluated for six different heights above the starting point at ground level (350, 500, 750, 1000, 2000 and 3000 m). In total, 10934 trajectories were generated throughout the study period of 2004-2008. The grid covers the area of interest with cells size of $0.5^\circ \times 0.5^\circ$ latitude and longitude. For air mass trajectory visualization and statistical analysis, a software application called TrajStat was used in which clustering, PSCF and CWT methods were included [12]. Demonstration of PSCF and CWT usage was presented on five years PM_{10} data set (2004-2008) continuously recorded by the Institute of Public Health of Belgrade.

2.1 Potential Source Contribution Function

Air parcel back trajectories, ending at the receptor site, are represented by segment endpoints. Each endpoint has two coordinates (latitude, longitude) representing the central location of an air parcel at a particular time. To calculate PSCF, the whole geographic region of interest is divided into an array of grid cells whose size is dependent on the geographical scale of the problem so that PSCF will be a function of locations as defined by the cell indices *i* and *j*. The construct of the potential source contribution function can be described as follows: if a trajectory end point lies at a cell of address (*i,j*), the trajectory is assumed to collect material emitted in the cell. Once aerosol is incorporated into the air parcel, it can be transported along the trajectory to the receptor site. The objective is to develop a probability field suggesting likely source locations of the material that results in high measured values at the receptor site.

Let *N* be the total number of trajectory segment endpoints during the whole study period. If segment trajectory endpoints fall into the *ij*-th cell (represented by n_{ij}) the probability of this event is given by

$$P[A_{ij}] = \frac{n_{ij}}{N} \tag{1}$$

where $P[A_{ij}]$ is a measure of the residence time of a randomly selected air parcel in the *ij*-th cell relative to the total time period. In the same *ij* cell there is a subset of m_{ij} segment endpoints for which the corresponding trajectories arrive at the receptor site at the time when the PM_{10} measured concentration are higher than a pre-specified criterion value. The choice of these criterion values have usually based on trial and error and in this paper the mean value of the measured concentration was used. In some publications the use of the 60-th and 75-th percentile criterion produced results that appeared to correspond better with known emission source locations. Thus, the probability of this high concentration event is given by

$$P[B_{ij}] = \frac{m_{ij}}{N} \tag{2}$$

where $P[B_{ij}]$ is subset probability related to the residence time of air parcel in the *ij*-th cell for the contaminated air parcel. Finally, the potential source contribution function is defined as

$$PSCF_{ij} = P[B_{ij} \mid A_{ij}] = \frac{m_{ij}}{n_{ij}} \tag{3}$$

where PSCF is the conditional probability that an air parcel which passed through the *ij*-th cell had a high concentration upon arrival at the receptor site. A sufficient number of endpoints should provide accurate estimates of the source location. Cells containing emission sources would be identified with conditional probability close to 1, if the trajectories that have crossed over the cells effectively transport the emitted contaminant to the receptor site. One can draw the conclusion that PSCF model provides a map of source potential of geographical areas, but it can not apportion the contribution of the identified source area to the measured concentration at the receptor site. Thus, the potential source contribution function can be interpreted as a conditional probability describing the spatial distribution of probable geographical source locations inferred by using trajectories arriving at the sampling site. Cells related to the high values of potential source contribution function are the potential source areas. However, the potential source contribution function maps do not provide an emission inventory of a pollutant but rather show those source areas whose emissions can be transported to the measurement site. To reduce the effect of small values of n_{ij}, an arbitrary weight function $W(n_{ij})$ is multiplied into the PSCF value to better reflect the uncertainty in the values for these cells.

2.2 Concentration Weighted Trajectory

In the current PSCF method, grid cells having the same PSCF values can result from samples slightly higher than the criterion concentrations or extremely high concentrations. As a result, larger sources can not be distinguished from moderate sources. According to this problem, a method of weighting trajectories with associated concentrations, CWT model was developed. In this procedure, each grid cell gets a weighted concentration obtained by averaging sample concentrations that have associated trajectories that crossed that grid cell as follows:

$$C_{ij} = \frac{1}{\sum_{l=1}^{M} \tau_{ijl}} \sum_{l=1}^{M} C_l \tau_{ijl} \tag{4}$$

C_{ij} is the average weighted concentration in the grid cell (i,j), C_l is the measured PM_{10} concentration observed on arrival of trajectory l, τ_{ijl} is the number of trajectory endpoints in the grid cell (i,j) associated with the C_l sample, and M is the total number of trajectories. Similar to PSCF model, a point filter is applied as the final step of CWT to eliminate grid cells with few endpoints. Weighted concentration fields show concentration gradients across potential sources. This method helps determine the relative significance of potential sources.

2.3 LIDAR measurement

A ground-based elastic LIDAR system has been operational in the Institute of Physics, Belgrade, since 2009. This biaxial LIDAR system is a single-wavelength backscatter system pointing vertically to the zenith. The light source used is a commercial Nd:YAG laser (LOTIS TII, model LS2131) operating at the second harmonic frequency, 532 nm, with repetition rate up to 20 Hz with the pulse duration around 10 ns. The beam divergence is typically about 0.5 mrad. As a light collector a Meade LX200 Schmidt-Cassegrain telescope with 12˝ diameter was used. At the present configuration the system has a maximum overlap beginning at 150 m allowing the system to perform up to 6-8 km. The backscattered laser radiation is detected by a low-noise PIN photodiode FD5N coupled to a 3 nm interference filter (Thorlabs FL532-3) to assure the reduction of solar background and improve the signal-to-noise ratio. The output signal is recorded by a National Instrument NI-5124 digitizer (12 bit, 200 MHz). Data were averaged every 1 min and summed up in blocks corresponding to 10 min. The spatial resolution applied is 15 m, which corresponds to 100 ns sampling time. The LIDAR system equation contains two unknown parameters the aerosol extinction and backscatter coefficients. In order to solve the equation for the aerosol backscatter coefficient, a relationship between the two quantities has to be assumed. This assumption introduces errors that may exceed 20%. In this paper Klett inversion technique was used to resolve aerosol backscatter and extinction profiles [13].

2.4 DREAM model

DREAM is a regional model designed to simulate and/or predict the atmospheric cycle of mineral dust aerosol [14]. The South East European Virtual Climate Change Center (SEVCCC) maintains dust forecast operations with DREAM and conducts modeling research and developments to shorten prediction. During model integration, calculation of the surface dust injection fluxes is made over the model grid points declared as deserts. Once injected into the air, dust aerosol is driven by the atmospheric model variables: by turbulent parameters in the early stage of the process when dust is lifted from the ground to the upper levels; by model winds in the later phases of the process when dust travels away from the sources; finally, by thermodynamic processes (atmospheric water phase changes producing clouds, rain and dust wet scavenging) of the atmospheric model and land cover features which provide wet and dry deposition of dust over the Earth surface.

3 Results and Discussions

Since we were concerned on the directions of the trajectories, the angle distance between back trajectories [15] has been used as the cluster model. A cluster number can be deduced through visual inspection and comparison of the mean-trajectory maps. We chose five as for the number of cluster because this gives the best representation of airflow classifications and their representative trajectories are shown on Fig. 1. The main directions of air mass flows over Belgrade were therefore grouped into 5 classes named: (1) West Southwest (W-SW); (2) East (E); (3) West Northwest (W-NW); (4) North (N); (5) South Southwest (S-SW).

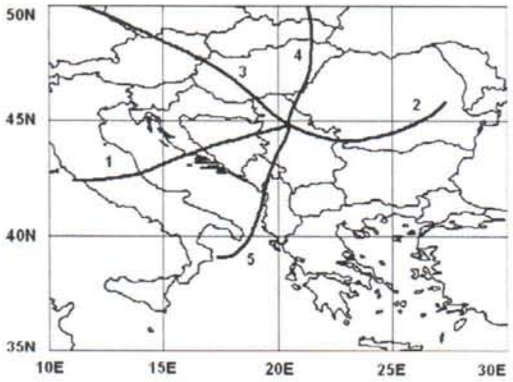

Fig. 1 Trajectories representing grouping of 48 hour backward trajectories of air masses over Belgrade into four classes for the period 2004-2008.

Based on the analysis of the whole trajectory data set, the most frequently arriving directions are W-NW (36%) and E (19%) thus suggesting the sampling site might be under influence of several source regions. Trajectories within different classes of airflow had distinct effects on the PM_{10} concentrations. The highest PM_{10} concentrations were associated with classes W-SW (61 μg m^{-3}) and S-SW (52 μg m^{-3}), while the lowest PM_{10} concentrations were found in class N (40 μg m^{-3}). Mean contribution of each class (including all trajectories and trajectories associated with PM_{10} measured at the receptor site greater then 50 μg m^{-3}) are shown on Fig. 2. High PM_{10} concentrations related to W-SW and S-SW classes would imply high polluted pathways. Along such pathways, the high emissions could be located, which would be further indicated by high PSCF values.

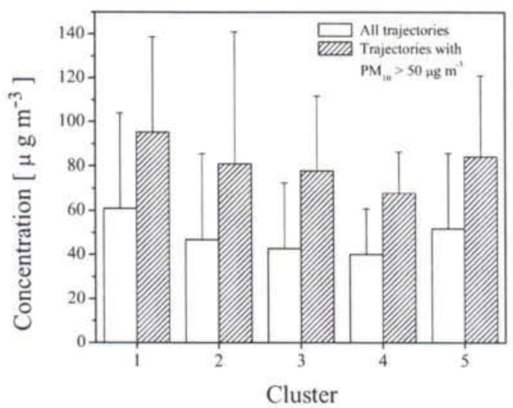

Fig. 2 Mean PM_{10} concentration and corresponding standard deviation within five classes

Additional insights into the nature of the PM_{10} sources are provided through a trajectory based evaluation of the upwind locations associated with high concentrations of these sources. Five year PM_{10} data set (2004-2008) has been used in PSCF and CWT modeling. Calculated PSCF values were subdivided into four categories: very weak (0.0–0.20), weak (0.20–0.40), intermediate (0.40–0.60) and strong (0.60–1.0). The results of PSCF are presented in Fig. 3 (above) and indicated that the strong potential probabilities are located in the west. In addition, higher PSCF values are observed from north east.

The CWT method evenly distributes concentration along the trajectories similar to PSCF, as presented in Fig. 3 (below). However, this method has an advantage over PSCF since CWT distinguishes major sources from moderate ones by calculating concentration gradients. PSCF shows probabilities of potential sources based on samples with concentrations higher than the criterion, which does not distinguish between moderate and major sources. The results suggest that the major contribution to atmospheric PM_{10} concentrations comes from local and regional sources. There is evident a long – range transport from western countries which is sporadically (mostly in spring and summer) associated with African dust outbreaks in levels of both PM_{10} and $PM_{2.5}$ [16]. Thus, PM_{10} data were separated for seasonal period and then divided into the two groups, greater and lower then average values for specific period. PSCF and CWT results for spring and autumn period are presented in Fig. 4. Based on the analysis of the seasonal trajectory data set, the most frequently arriving directions are west, north-west and south-west thus suggesting the sampling site might be under influence of several source regions. It can be seen that the highest PSCF and CWT values are from the west, south west and north during spring period. In addition, higher PSCF values are observed from west and south west during autumn period.

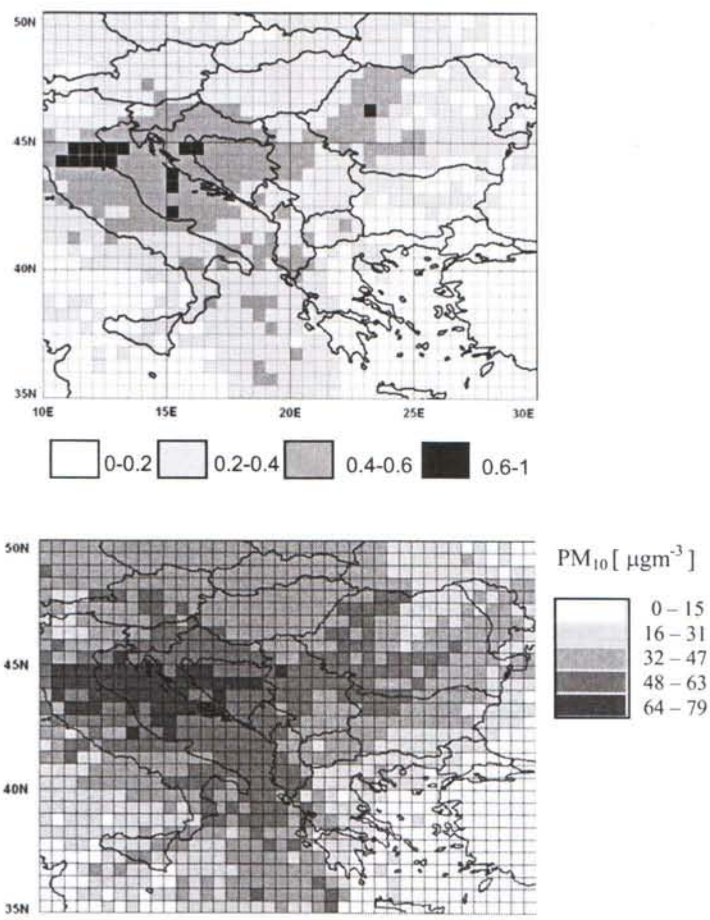

Fig. 3 Distribution of PSCF (above) and CWT (below) for the period 2004-2008.

Saharan dust particles can be transported at different altitude ranges affecting the local aerosol content when it reaches very low altitude and the planetary boundary layer. If lofted in the free troposphere, dust particles can be transported over long distances, penetrating deeply into Northern Europe. LIDAR measurements are very useful tool for investigation of dust transport thanks to the capability to provide high quality dust vertical profiles. The first elastic backscatter LIDAR measurements over Belgrade have been conducted during 2009. Saharan dust is rich with Fe, which in both soluble and suspended forms gives a characteristic red or yellow color to precipitation. Such event was observed on 27. March, 2010 over Belgrade and in Fig 5. dust load predicted by the DREAM model can be seen.

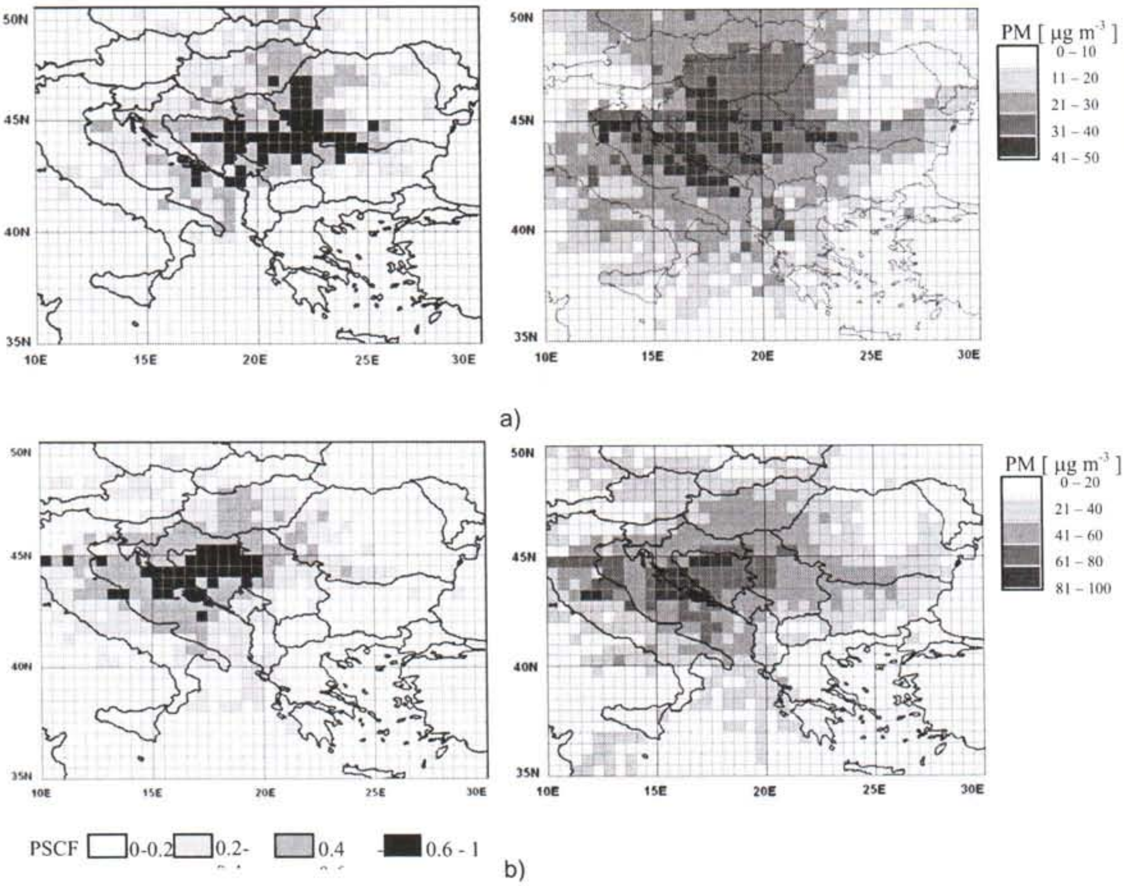

Fig. 4 Distribution of PSCF (left) and CWT (right) for PM_{10} during a) spring and b) autumn period 2004-2008.

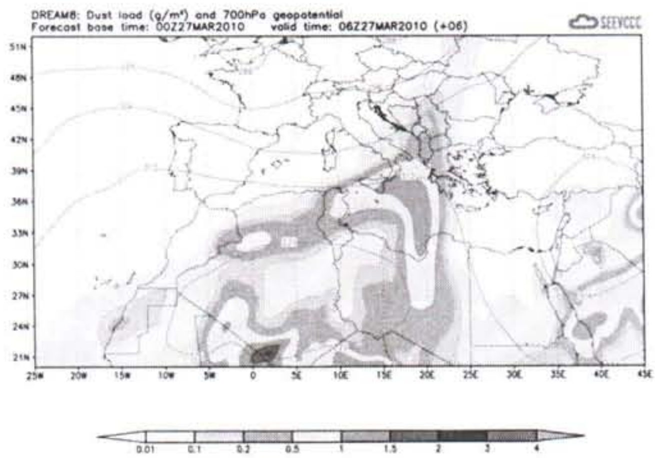

Fig. 5 Dust load over South Europe, on 27. March, 2010 estimated by the DREAM model

Vertical profiles and temporal variation of dust concentration predicted by the DREAM model over Belgrade, as well as averaged backscatter coefficient profile derived from LIDAR measurements are shown in Fig 6. It can be seen that the dust episode lasted about 12h and there is considerable agreement between predicted and measured variables.

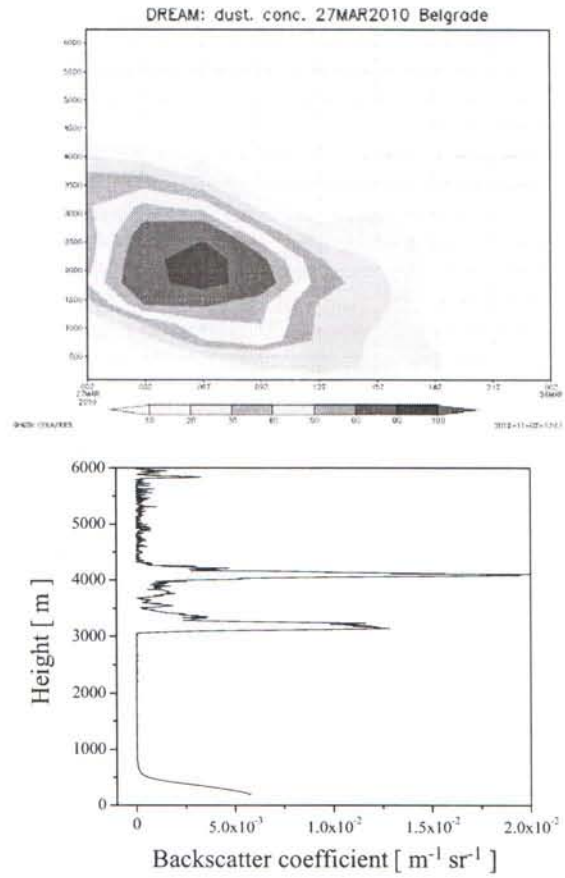

Fig. 6 Dust concentration predicted by the DREAM model (above) and observed backscatter coefficient over Belgrade, based on LIDAR measurement (below), on 27. March, 2010, 03:00 h

4 Conclusions

In this study, trajectory clustering, PSCF and CWT methods were used to investigate the transport pathways and potential source regions of PM_{10} in Belgrade based on 2004-2008 data set. Based on the cluster analysis of the 48 hour backward trajectories five classes of backward trajectories were generated: West Southwest, East, West Northwest, North and South Southwest. The highest PM_{10} concentration was found along the West Southwest and South Southwest pathway. Along such pathways, the high emissions could probably located, which were indicated by high PSCF and CWT values. For the case study analysis of regional Saharan dust impact elastic backscatter LIDAR measurements and DREAM model were used. The results show good temporal and spatial correlation between observed and forecast dust events.

Acknowledgment

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June, 9 2011

LETTER OF INVITATION

Dear Darko Vasiljevic,

It gives us a great pleasure to invite you to participate and give your valuable contribution as a speaker to the 5th Workshop on Optoelectronic Techniques for Environmental Monitoring-OTEM 2011 which will take place during 28th to 30th of September at the "Romanian Atmospheric Observatory", Magurele, Romania. It will be an honor to have you as an invited lecturer.

The aim of this meeting is to bring together experts from countries around the world to discuss laser remote sensing techniques for environmental monitoring issues and to provide a better understanding of this scientific field.

The Workshop will cover four topics:

- Sensors and instrumentations(in situ, laboratory, remote sensing)
- Satellite imagery
- Modelling and analysis tools
- Hazard and risk assessment

Also you are invited to participate to the following special events:

- September 28: ***Official opening*** of the Romanian Atmospheric Observatory-RADO
- September 29: ***Final conference*** of the DELICE project (FP7-REGPOT-2008-1, 229907) - Environmental Remote Sensing Conference
- September 30: ***Exhibition*** on optoelectronic equipments for environmental monitoring
- September 30: ***Info hour*** on Innovative clustering policy

We will be able to cover your travel and accommodation expenses. Please let us know your wishes regarding the exact period for traveling to Romania, so that we can make arrangements for airline tickets and accommodation.

We look forward to welcome you!

Yours sincerely,
Dr. Doina Nicolae