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Backward trajectories and cluster analyses for study of PM_{10} concentration in Bulgaria during dust episodes

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Abstract: Air pollution with particulate matter (PM) is a serious problem in Bulgaria and although the measures are taken, the results are still unsatisfactory. The PM concentrations are highly related to the air masses arriving at the receptor site. For the forecasted by the Barcelona Dust Forecast Center (BDFC) dust episodes, Trajectory Statistical Methods (TSMs) are applied to back-trajectories, computed by HYSPLIT model (Hybrid Single-Particle Lagrangian Integrated Trajectory). Cluster analysis (CA), Potential Source Contribution Function (PSCF) and Concentration Weighted Trajectory (CWT) are used to identify the influence of the air mass origin on particulate matter levels in Bulgarian urban areas (Sofia and Plovdiv) and the near background areas (Kopitoto and Rozhen) for 2019 and 2020. The CA shows that the prevailing direction of air masses coming towards Bulgaria during dust episodes is east (38.1 – 53.2 % of the observed cases). One particular dust event is examined and the observed PM_{10} concentrations are discussed.

Keywords: dust episodes, PM_{10}, back-trajectories, TSM, cluster analysis, PSCF, CWT.

1. INTRODUCTION

The air pollution affects in a negative way the human health. Because of their easy inhalation into the respiratory tract, PM_{10} and $PM_{2.5}$ are widely recognized to cause inflammatory processes and diseases of the respiratory and cardiovascular systems (Lim et al., 2012; Straif et al, 2013; WHO, 2003; Querol et al., 2019). They can affect negatively the environment also (climate, visibility and biogeochemical cycles) (IPCC, 2013). Although there are many international actions and collaborations in order to control the particulate levels and effects (EEA, 2018), the decrease of the PM levels is still unsatisfactory. 78.6 % of the population in Bulgaria is exposed to the exceedance of the limit values of PM_{10} (MoEW Report, 2019).

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Particulate matters (PMs) are emitted in the atmosphere by natural and anthropogenic sources and they can be released at one location, but to have an impact on air quality at long distances away (Gangoiti et al, 2001; Kallos et al., 2006; Buseck and Adachi, 2008; Moran et al., 2014). Dust outbreaks might be due not only to desert dust but also to other anthropogenic and natural sources. They can be mixed with dust during transport towards the receptor station, because of the synoptic situation, causing extremely high PM levels locally. During the last years there is a tendency of increasing dust production and transportation. That is why several dust models have been developed for regional and global dust forecasting.

Computing air parcels trajectories, analysing air masses arriving at receptor site and applying trajectory statistical methods (TSM) can be very useful in order to investigate the source region of PM (Salvador et al., 2008, 2010; Molnar et al., 2017; Lopez et al, 2019; Meng et al., 2020). Back trajectory analysis helps to determine the origin of air masses and establish source-receptor relationships (Fleming et al., 2012). The backward approach allows tracing the motion of air parcels from receptor to source sites. Since the air parcel arriving at a site could have followed different trajectories, the use of multiple trajectories could reduce the uncertainty generated by interpolation of meteorological data, low temporal or spatial resolution of wind or an inappropriate selection of the starting heights, transport, turbulence and truncation processes. The TSMs are used in PM source apportionment studies for identifying potential source regions (Perrone et al., 2018; Gunchin et al., 2019; Belis C. et al. 2019). Applications of these methods for Bulgaria are rare. For the first time, researchers from NIMH (Neykova R. & Hristova E., 2020) tried to identify the influence of the air mass origin on particulate matter levels.

This study aims to present the application of those methods together with forecasted dust episodes by Barcelona Dust Forecast Center on daily levels of PM_{10} in two of the most populated Bulgarian cities (Sofia and Plovdiv) and the nearest background stations (Kopitoto and Rozhen) using TSMs as supplementary tools. This is done for the two year period from 01.01.2019 to 31.12.2020.

2. METHODOLOGY

2.1. The regions of study

In this work four sites (Sofia, Kopitoto, Plovdiv and Rozhen) were selected (Figure 1). Sofia and Plovdiv are both struggling with air quality problems - high levels of PMs and exceedance of the permitted number of days per year with daily averaged PM_{10} concentrations above 50 $\mu g.m^{-3}$.

In Sofia city there are five urban automatic monitoring stations (AMS), part of the National air quality network: AMS Nadezhda (42.732292° N, 23.310972° E), AMS Hipodruma (42.680558° N, 23.296786° E), AMS Pavlovo (42.669797° N, 23.268403° E), AMS Mladost (42.655488° N, 23.383271° E). There are two AMS in Plovdiv:

AMS Kamenitsa (42.142889° N, 24.765239° E) and AMS Trakia (42.141186° N, 24.787952° E).

When discussing cross-border pollution, it should be noted that it is first detected at the mountain stations. This is the reason for including AMS Kopitoto and AMS Rozhen in our study. Moreover, these are the only background stations in Bulgaria where PM concentrations in ambient air are measured. AMS Kopitoto (42.637192° N, 23.243864° E, 1321 m asl (above sea level)) is placed near to Sofia in the Vitosha Mountain. AMS Rozhen (41.694342° N, 24.738078° E, 1750 m asl) is located on the Rozhen peak in the Rhodope Mountains, at a distance of 50 km from the town of Plovdiv.

Fig. 1. Map of Bulgaria with study areas – Sofia and Plovdiv (red dots) and background stations Kopitoto and Rozhen (green dots)

2.2. PM_{10} data

The PM_{10} hourly concentrations for the period 01.01.2019 – 31.12.2020 are used in the analysis of the cases with forecasted by the Barcelona Dust Regional Center dust episodes over Bulgaria. This data is provided by the Executive Environment Agency in Bulgaria.

2.3. Back-trajectory analyses

The HYSPLIT 4.0 model was used to determine the region of origin of the air masses that affect the explored stations (Stein et al. 2015). Trajectories at several heights above the point of interest were run in order to better illustrate the vertical structure of the atmosphere. Three days back-trajectories ending at 04:00, 12:00 and 18:00 UTC and 500, 1500 and 2000 m above ground level (a.g.l.) were computed. The weekly archived data GDAS (Global Data Assimilation System) with resolution 1º was used as meteorological input information.

2.4. Trajectory statistical methods (TSM)

The trajectory statistical methods (TSM) were used as additional instrument for characterizing the synoptic situations of dust episodes over Bulgaria. Cluster analysis (CA), Potential Source Contribution Function (PSCF) and Concentration Weighted Trajectory (CWT) were applied as TSMs in our study. More detailed information for those methods is presented in Neykova and Hristova (2020). All those functions are applied to the HYSPLIT back-trajectories with the software package "Openair" in R (http://www.opair-project.org/PDF/OpenAir_NewsLetter_Issue14.pdf).

2.5. Barcelona Dust Regional Center

The Center was created in 2007 thanks to the formal agreement of two Spanish institutions - the Meteorological State Agency of Spain (AEMET) and the Barcelona Supercomputing Center (BSC). BDFC manages and coordinates the research activities and operations of the World Meteorological Organization (WMO) related to sand and dust storms. The Center provides access to available dust products and coordinates a network of collaborators in Northern Africa, the Middle East and Europe (Activity Report 2020). The NMMB/BSC-Dust (Pérez et al., 2011; Haustein et al., 2012) is an online multi-scale atmospheric dust model designed and developed at BSC-CNS in collaboration with NOAA/National Centers for Environmental Prediction (NCEP), NASA Goddard Institute for Space Studies and the International Research Institute for Climate and Society (IRI).

3. RESULTS AND DISCUSSION

3.1. Dust episodes

In order to identify dust episodes in Bulgaria the first step was to explore the archive data from Barcelona Dust Forecast Center – BDFC. The data for surface dust concentrations for the period 01.01.2019 – 31.12.2020 was examined. All cases with forecasted dust surface concentrations over Bulgaria were extracted and summarized in Table 1. There was no information for the periods: 22.01.2019 – 28.02.2019, 05.03.2019 – 31.03.2019, 03.04.2019 – 30.06.2019, 02.07.2019 – 21.07.2019 and 12.09.2020 – 15.09.2020. The analysis showed that forecasted dust events over Bulgaria were 41% of the study period. More cases with dust transport from North Africa and Middle East to Bulgaria were forecasted in 2020 than in 2019. The duration of those periods is longer compared to the other. Dust events are more frequent during the period August – October for both years.

Table 1. Cases with forecasted dust episodes over Bulgaria according to the BDFC in the period 01.01.2019 – 31.12.2020

Month	2019	2020
January	20, 21	14-15, 25-29
February		04-05, 08-11
March	01, 03	01-05, 07-10, 22, 25-31
April		02-05, 14-15, 18-20, 29-30
May		10-21
June		05-27
July	01-02, 22-23, 28-31	03-07, 24-25, 27-31
August	01-04, 14-17, 21-28, 31	04, 07-08, 14-16, 19, 25-26, 31
September	01, 07-15, 19, 23-24	01, 03-04, 06-10, 17-18, 23-27, 29
October	03-05, 10-14, 18-30	03-08, 10-11, 13, 16-17, 23-26, 28-29
November	04-21	13, 17, 29
December	06-07, 10, 13, 20-22	05-06, 09-11, 30-31

3.2. Particulate matter concentrations

The daily averaged PM_{10} concentrations for all days in Table 1 and for all studied stations are explored in this paragraph.

3.2.1. Plovdiv

There is a good relation in variations of the daily PM_{10} concentrations observed at AMS Trakia and AMS Kamenitsa in Plovdiv (Figure 2). The number of days with exceedance of the 24-h limit value (50 $\mu g.m^{-3}$) at AMS Trakia is 79 or 34.5% of the cases, while for AMS Kamenitsa it is 53 or 23.1 %. Some of those exceedances are due to dust transportation from North Africa and Middle East. Probably the differences in these numbers are due to local sources of air pollution. The daily PM_{10} concentrations for the study period are between 15.71 and 183.12 $\mu g.m^{-3}$ at AMS Trakia and from 11.38 to 149.32 $\mu g.m^{-3}$ at AMS Kamenitsa. The registered days with fog are 28 and 189 with rain. 67.7% of the cases with rain are with 0 mm amount. The daily averaged wind speed is ranged from 0.3 to 8.1 $m.s^{-1}$.

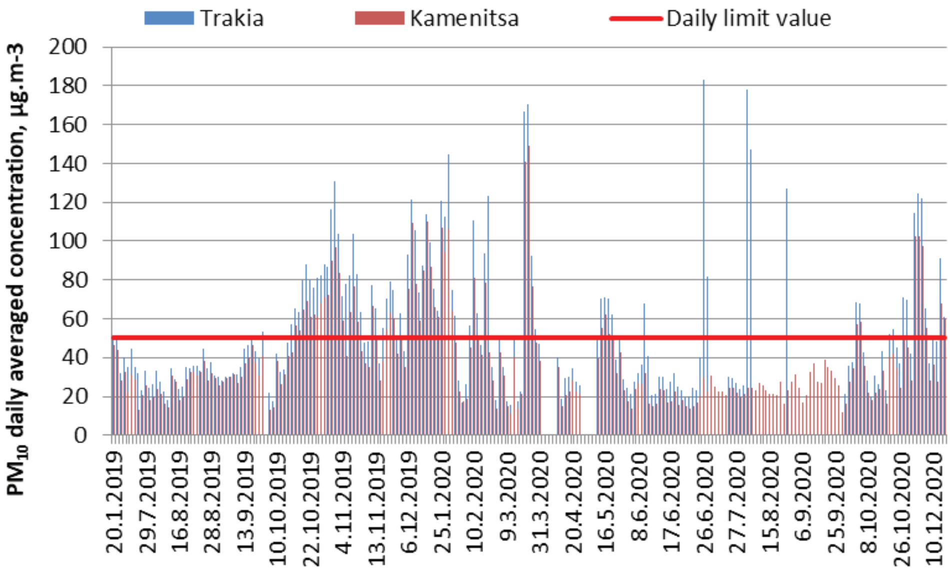


Fig. 2. Daily mean PM_{10} values at Plovdiv air quality stations - Trakia and Kamenitsa for the cases with forecasted dust episodes according to the BDFC for the period 01.01.2019 – 31.12.2020

3.2.2. Rozhen

Daily PM_{10} concentrations obtained at AMS Rozhen during the study period are presented in Figure 3. Only 2 days were observed with exceedance of the 24-h limit value (50 $\mu g.m^{-3}$): 26 March (195.72 $\mu g.m^{-3}$) and 26 October (196.36 $\mu g.m^{-3}$), both in 2020. The daily PM_{10} concentrations are between 0.33 and 196.36 $\mu g.m^{-3}$. The registered days with fog are 82 and 131 with rain. 37.4% of the cases with rain are with 0 mm amount. The daily averaged wind speed is ranged from 0.5 to 6 $m.s^{-1}$.

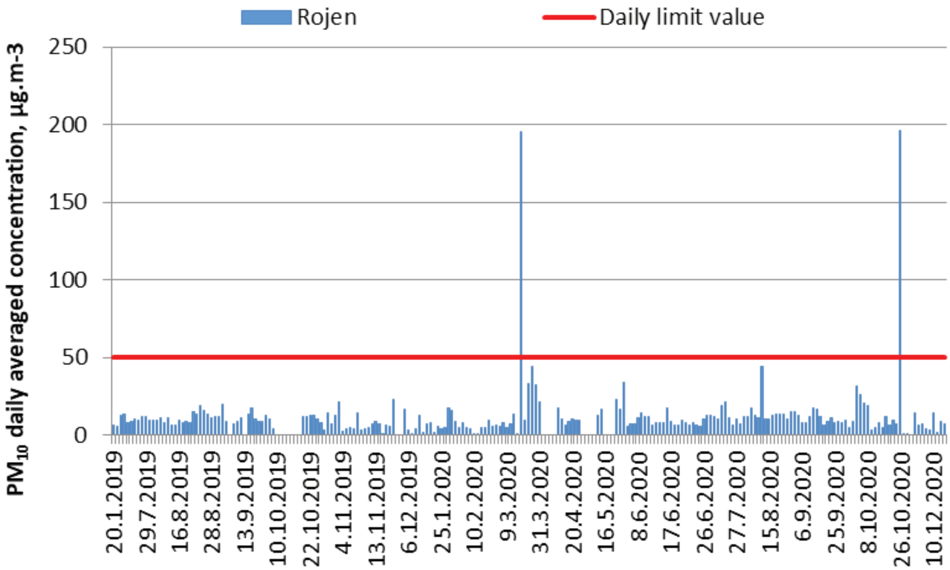
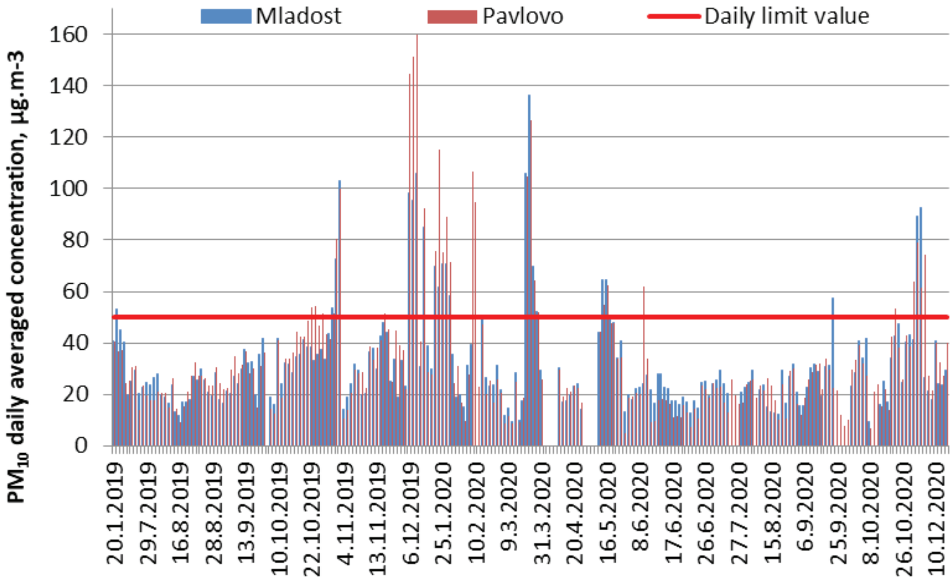


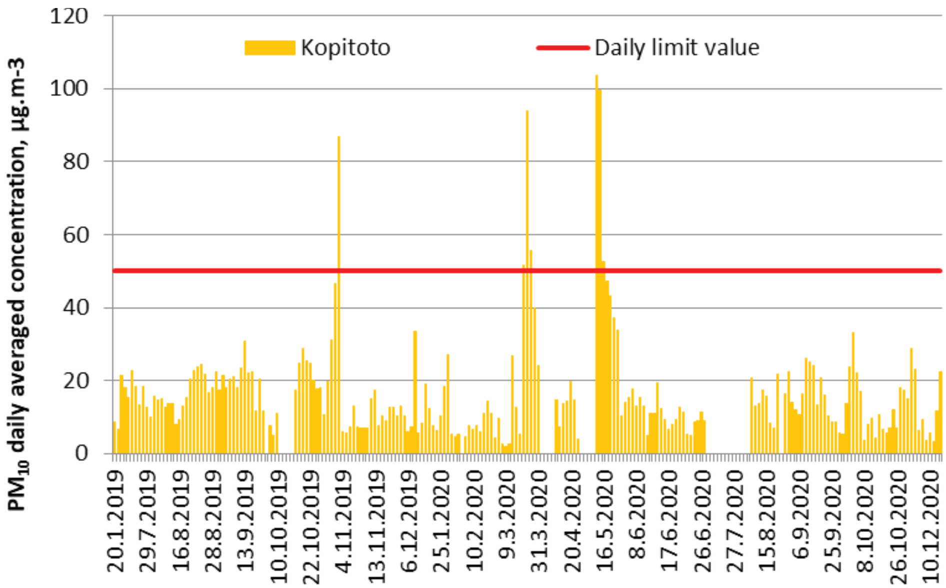
Fig. 3. Daily mean PM_{10} values at Rozhen air quality station for the cases with forecasted dust episodes according to the BDFC for the period 01.01.2019 – 31.12.2020

3.2.3. Sofia

Comparison of daily PM_{10} concentrations observed at AMS Mladost, AMS Pavlovo and AMS Kopitoto are presented in Figure 4. It can be seen that all cases with exceedance of the daily limit value at AMS Kopitoto coincide with those at the AMS Mladost and AMS Pavlovo. The number of days with exceedance of the 24-h limit value (50 $\mu g.m^{-3}$) at AMS Mladost is 23 or 10% of the study period, while at AMS Pavlovo it is 30 (13%) and only 7 (3.1%) at AMS Kopitoto. The AMS Kopitoto is a background station, situated outside the city in the Vitosha mountain. This is the reason for the lower observed PM_{10} concentrations and the less number of days with exceedances. Most of the cases of exceedances of PM_{10} concentration levels at stations Mladost and Pavlovo, are probably due to local pollutants. The daily PM_{10} concentrations are between 5 and 160.02 $\mu g.m^{-3}$ at AMS Pavlovo, and between 9.57 and 136.16 $\mu g.m^{-3}$ at AMS Mladost. At AMS Kopitoto the PM_{10} concentration varies in the range from 1.97 to 103.74 $\mu g.m^{-3}$. The registered days with fog are 10 and 135 with rain. 45.2% of the cases with rain are with 0 mm amount. The daily averaged wind speed is ranged from 0 to 4.5 $m.s^{-1}$.



(a)



(b)

Fig. 4. Daily mean PM_{10} values in Sofia air quality stations – a) Mladost and Pavlovo, b) Kopitoto for the cases with forecasted dust episodes according to the BDFC for the period 01.01.2019 – 31.12.2020

3.2. Results from TSM analysis

6183 backward trajectories, ending at 04:00, 12:00 and 18:00 UTC for each day of the study period and at 3 different heights (500, 1500, 2000 m agl) were computed for AMS Trakia (Plovdiv), AMS Rozhen and AMS Mladost (Sofia). The AMS Kopitoto is placed close to Sofia at 11 km to AMS Mladost and 5 km to AMS Pavlovo. The resolution of the meteorological data used in HYSPLIT is 1 degree latitude-longitude, so we present here only the back trajectories for AMS Mladost. The trajectories were grouped into five different air-flow patterns (clusters) using "Openair" package in R. The comparison of obtained clusters, PSCF and weighted mean PM_{10} concentrations for the 500 m agl level, is presented in Figure 5.

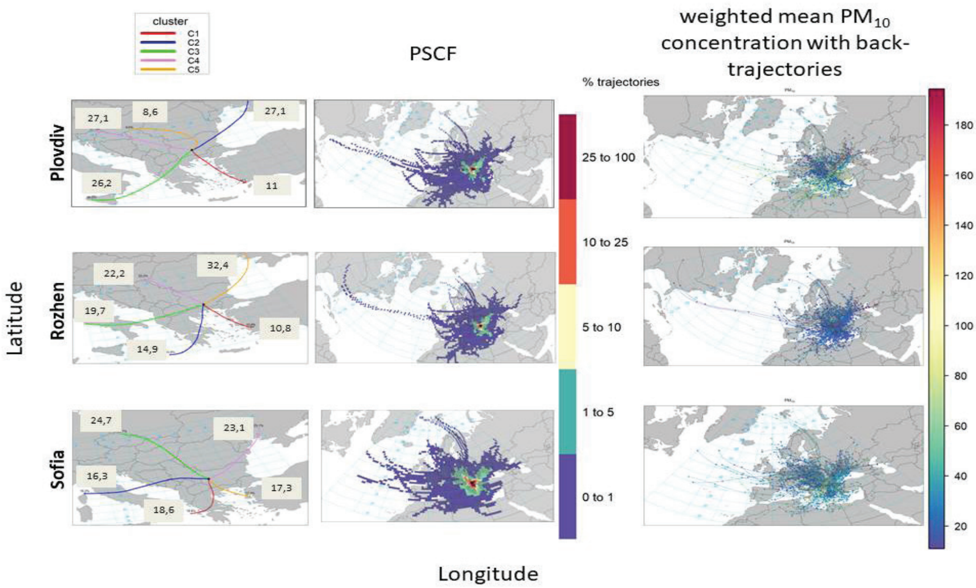


Fig. 5. Cluster analysis (CA) (left panels), PSCF (middle) and weighted mean PM_{10} concentrations with back-trajectories for level 1 (500 m agl) (right) for Plovdiv, Rozhen and Sofia, respectively

The comparison of the cluster analysis for the three starting locations show some similarities, but more differences. The differences are due to the predominant air masses, because of the location of the starting points. AMS Trakia (Plovdiv) is in the central part of southern Bulgaria, AMS Rozhen is located on a peak in the Rhodope Mountains near Plovdiv, while AMS Mladost (Sofia) is in the central western part of the country.

At all the three levels the predominant air masses are from west north-west (from Central Europe) in Plovdiv, while at AMS Rozhen at level 1 the predominant air masses come from north-east, turning to west south-west at levels 2 and level 3. For Sofia the west component is prevailing at all three levels.

PSCF frequency maps indicate the geographic origin of the air masses (transboundary) that reach the study site. The PSCF analysis for the 3 studied levels show the prevailing percentage of air masses in Plovdiv to be from west, west – south-west in Rozhen and south - south-west in Sofia.

The CWT analysis is made in order to have the PM_{10} concentrations for each one of the trajectories. The results are presented on geographical maps. For each cell of any map a weighted concentration of the PM was computed.

Results for the averaged concentrations of PM_{10} for all stations are presented on Table 2.

Table 2. Mean PM_{10} concentrations (μg.m^{-3}) by trajectory cluster at 3 different levels

	Plovdiv		
	Level 500	Level 1500	Level 2000
Cluster 1	59 (S)	58.8 (SW)	58.2 (SW)
Cluster 2	60.1 (NE)	56.5 (W)	49.1 (W)
Cluster 3	60.3 (SW)	46 (NW)	48.4 (NW)
Cluster 4	40.2 (SW)	53 (NE)	55.2 (NE)
Cluster 5	52.7 (SSW)	58.6 (S)	60.1 (S)
	Rozhen		
	Level 500	Level 1500	Level 2000
Cluster 1	9.3 (SE)	9.4 (SW)	10.3 (SW)
Cluster 2	8.6 (SW)	11.8 (W)	12.5 (W)
Cluster 3	11.4 (W)	12.1 (NW)	11.6 (NW)
Cluster 4	11.9 (N)	16.6 (NE)	15.3 (NE)
Cluster 5	15.8 (E)	12.1 (SE)	12 (SE)
	Sofia		
	Level 500	Level 1500	Level 2000
Cluster 1	35.8 (SW)	36.7 (SW)	36.3 (SW)
Cluster 2	35.2 (W)	33.7 (W)	30.9 (W)
Cluster 3	26.7 (NW)	26.4 (NW)	27.6 (NW)
Cluster 4	30.9 (NE)	29.7 (NE)	31.2 (NE)
Cluster 5	32.2 (SE)	31.5 (S)	30.6 (S)

The lowest concentrations of PM_{10} in Plovdiv are obtained during periods with air masses from south-west at level 1(C4) and from north-west (C3 at level 2 and level 3) and the highest – from south-west (C3 at level 1 and C1 at level 2) turning to south at level 3 (C5). The lowest concentrations in AMS Rozhen are obtained during air flows from south-west (C2 at level 1 and C1 at levels 2 and 3) and highest – from east (C5 at level 1), turning to north-east (C4 at levels 2 and 3). The air mases from north-west (C3 at levels 1, 2 and 3) contribute to the lowest measured concentrations of PM_{10} in AMS

Mladost (Sofia), while the highest are during flows from south-west (C1 at levels 1, 2 and 3).

3.3. Back trajectories analysis for cases with dust events

The maximal, minimal and mean daily averaged PM_{10} concentrations in all AMS for the study period are presented in Table 3.

Table 3. Maximal, minimal and average daily PM_{10} concentrations ($\mu g.m^{-3}$) at the air quality stations for the study period

Station	min	max	mean
Trakia	15.71	183.12	54.42
Kamenitsa	11.38	149.32	38.98
Rozhen	0.33	196.36	12.35
Mladost	9.57	136.16	31.87
Pavlovo	5	160.02	32.88
Kopitoto	1.97	103.74	16.66

As the study period comprises all cases of forecasted dust episodes over Bulgaria, no matter if the forecasted dust surface concentration is in the study area, the mean values are lower than the daily limit value (except for AMS Trakia and it is probably due to local pollution).

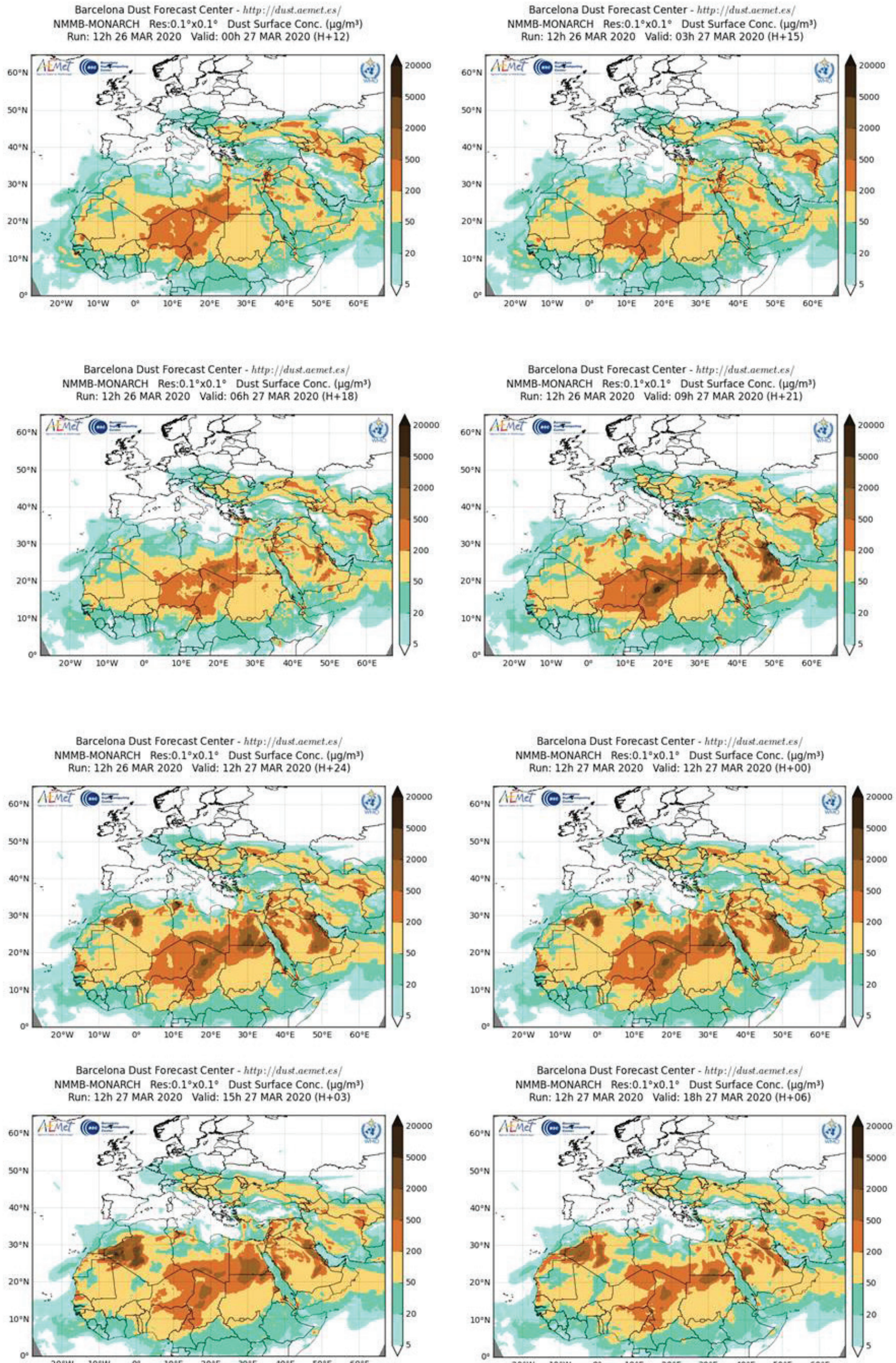
The cases with exceedance of the daily limit value in the background AMS stations Rozhen and Kopitoto are given in Table 4.

Table 4. Dates of exceedances of the daily limit value and the origin of the air masses

Station	Period	Origin of the air masses
Rozhen	26.03.2020	east
Rozhen	26.10.2020	west
Kopitoto	31.10.2019	west
Kopitoto	27, 28 and 29.03.2020	east turning to north
Kopitoto	14, 15 and 16.05.2020	south-west turning to north at level 1

One specific event (26-29.03.2020) detected in both background AMS is presented here. The analyses of the PM_{10} data show that the exceedance detected at the AMS Rozhen is a day before that at the AMS Kopitoto. This is due to the prevailing air masses at the AMS Rozhen station. The forecasts from the Barcelona Dust Forecast Center and the HYSPLIT back-trajectories are presented in Figures 6 and 7, respectively.

Figure 6 gives a forecast development for 27.03.2020 in 3 hours. The first run of the model is on 26.03.2020 12h and is valid for 27.03.2020 00h and the last run of the model is on 27.03.2020 12h, valid for 28.03.2020 00h.


Barcelona Dust Forecast Center - http://dust.aemet.es/
NMMB-MONARCH Res:0.1°x0.1° Dust Surface Conc. (µg/m³)
Run: 12h 26 MAR 2020 Valid: 00h 27 MAR 2020 (H+12)
Barcelona Dust Forecast Center - http://dust.aemet.es/
NMMB-MONARCH Res:0.1°x0.1° Dust Surface Conc. (µg/m³)
Run: 12h 26 MAR 2020 Valid: 03h 27 MAR 2020 (H+15)
Barcelona Dust Forecast Center - http://dust.aemet.es/
NMMB-MONARCH Res:0.1°x0.1° Dust Surface Conc. (µg/m³)
Run: 12h 26 MAR 2020 Valid: 06h 27 MAR 2020 (H+18)
Barcelona Dust Forecast Center - http://dust.aemet.es/
NMMB-MONARCH Res:0.1°x0.1° Dust Surface Conc. (µg/m³)
Run: 12h 26 MAR 2020 Valid: 09h 27 MAR 2020 (H+21)
Barcelona Dust Forecast Center - http://dust.aemet.es/
NMMB-MONARCH Res:0.1°x0.1° Dust Surface Conc. (µg/m³)
Run: 12h 26 MAR 2020 Valid: 12h 27 MAR 2020 (H+24)
Barcelona Dust Forecast Center - http://dust.aemet.es/
NMMB-MONARCH Res:0.1°x0.1° Dust Surface Conc. (µg/m³)
Run: 12h 27 MAR 2020 Valid: 12h 27 MAR 2020 (H+00)
Barcelona Dust Forecast Center - http://dust.aemet.es/
NMMB-MONARCH Res:0.1°x0.1° Dust Surface Conc. (µg/m³)
Run: 12h 27 MAR 2020 Valid: 15h 27 MAR 2020 (H+03)
Barcelona Dust Forecast Center - http://dust.aemet.es/
NMMB-MONARCH Res:0.1°x0.1° Dust Surface Conc. (µg/m³)
Run: 12h 27 MAR 2020 Valid: 18h 27 MAR 2020 (H+06)

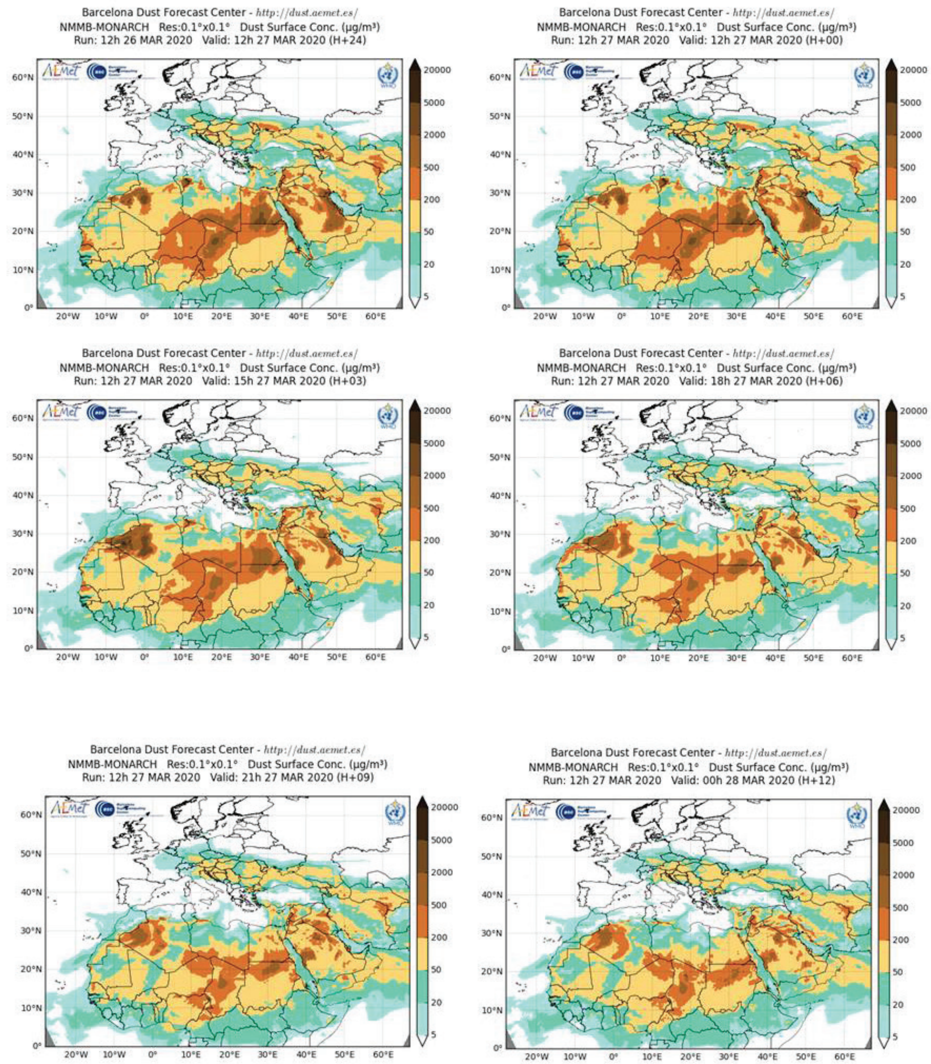


Fig. 6. Forecast for 26-27.03.2020 according to the Barcelona Dust Forecast Center

As it is seen this is a strong dust event, with origin from the Sahara desert. The prevailing air masses originate from North Africa, go through the Arabian Peninsula and making an U-turn, enter Bulgaria from the east, changing again direction to the north-west (Central Europe). The daily PM_{10} concentrations observed at the air quality stations are given in Table 5.

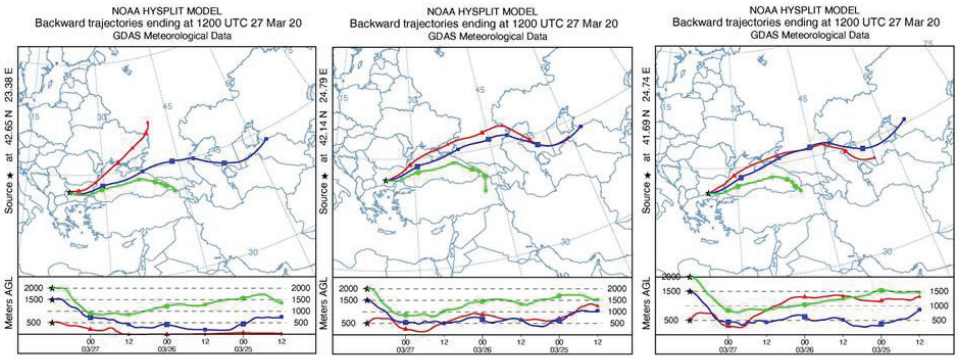


Fig. 7. HYSPLIT back trajectories for 27.03.2020 ending at Sofia, Plovdiv and Rozhen

Table 5. Daily PM_{10} ($\mu g.m^{-3}$) concentrations for 26-27.03.2020

Station	26.03.2020	27.03.2020
Trakia	22.38	166.61
Kamenitsa	21.06	140.98
Rozhen	195.72	9.67
Mladost	17.39	105.83
Pavlovo	18.73	104.61
Kopitoto	5.4	51.5

Confirming the expectations, this dust event is first detected at the highest situated station (AMS Rozhen), as seen from the observational data. The long range dust transportation is on higher levels of the atmosphere and needs time to reach the ground surface where it can be detected by the ground based stations discussed in this study.

4. CONCLUSIONS

This study presents an application of back trajectories and TSMs for forecasted dust episodes according to the Barcelona Dust Forecast Center for 2 years (2019 and 2020). The PM_{10} concentration data from ground based automatic measurement stations Plovdiv, Sofia and the nearest background stations – Rozhen and Kopitoto were examined. Air mass back-trajectories were grouped into 5 clusters, representing a typical meteorological scenario during dust episodes in Bulgaria. The results from CA, PSCF and CWT analyses present cross-border natural and anthropogenic dust transportation mainly from the east. The possible regions of origin are North Africa or the Middle East. According to the CA the air masses coming from east direction (Middle East) are between 38.1 – 53.2 % of the observed cases. Between 15 to 26 % of the air masses come from the south-west (North Africa). The observed dust event in the period 26-29.03.2020 shows that big amount of dust is lifted from the Sahara desert and transported towards the Middle East, going through Bulgaria from the east causing exceedance of the daily limit value in PM_{10} concentrations in the selected air quality stations.

More studies are needed about the seasonal trend of dust events. Combining these methods with analyses of the PM chemical composition would give more detailed information about the origin of detected dust in particulate matters, observed in Bulgaria.

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