

Article

Saharan Dust Storm Aerosol Characterization of the Event (9 to 13 May 2020) over European AERONET Sites

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Abstract: This research was aimed at investigating the Saharan dust cloud recorded on 11 and 12 May 2020, by AERONET AOD stations in Italy, Austria, Slovakia, Poland, Ukraine, and Romania and determining whether it affected the area of the Republic of Moldova. During this period, the Chisinau AERONET monitoring site was not operational. The incentive for the investigation was the discovery of a high sediment load in rainwater collected on 12 May 2020 in Pelinia, a village in the Dochia district of the Republic of Moldova, in the southeastern part of Europe (47.8780 latitude, 27.8344 longitude), which could have originated from the Saharan dust storm. Backward trajectory analysis with NOAA's HYSPLIT model confirmed that the Saharan dust storm impacted the village of Pelinia. Scanning electron microscopy coupled with electron dispersive X-ray spectroscopy (SEM-EDS) and Fourier transform infrared spectroscopy (FTIR) analysis of Pelinia rainwater sediments confirmed the chemical composition and morphological structure of Saharan dust particles. The particle size of the sediments matched the measurements at the AOD stations at Timisoara and Magurele, supporting the suggestion that Saharan dust probably entered the Republic of Moldova from Romania. FTIR analysis identified chemical compounds such as carbon dioxide, carbonates, sulfates, ferrocyanides, and organics (amines, amides, polypeptides, imines, oximes, pyrroles, aldehydes, sulfoxides, sulfones, nitro-derivatives) that were adsorbed and/or absorbed from the atmosphere, consistent with Saharan dust aerosols. Bio-allergens such as pollen were detected in the SEM images, showing the role of Saharan dust in transporting and spreading this kind of biological material. This study highlights the risk of Saharan dust clouds to humans, animals, and plants, but also its potential benefits for agriculture when suitable conditions are met in this regard.

Keywords: Saharan aerosols; biological material; marine phytoplankton; agriculture and health



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1. Introduction

This paper presents research aimed at studying the trajectory and influence of Saharan dust over European countries using mixed methods of investigation to make up for the lack or temporary inactivity of specific measurement stations.

Due to the continuous and accelerated erosion of the soil, dust particles from the Sahara desert, by their characteristics, belong to the category of suspended particulate matter (PM) [1]. Periodically, atmospheric conditions favor the movement of Saharan dust clouds from North Africa to the Mediterranean area, reaching the central and eastern parts of Europe [2–5] and even the American continent [6–8]. Monitoring the trajectory of

Saharan dust is of interest to both researchers and the public due to its particularities and its influence on the climate, but also on agriculture and even human health based on its chemical composition, which has been identified [9]. Its biological content [10], including toxic biological allergens, can affect human health by causing respiratory and cardiological disorders and skin irritation [11], which is why it is of strong interest to monitor the route of dust clouds and identify the affected areas.

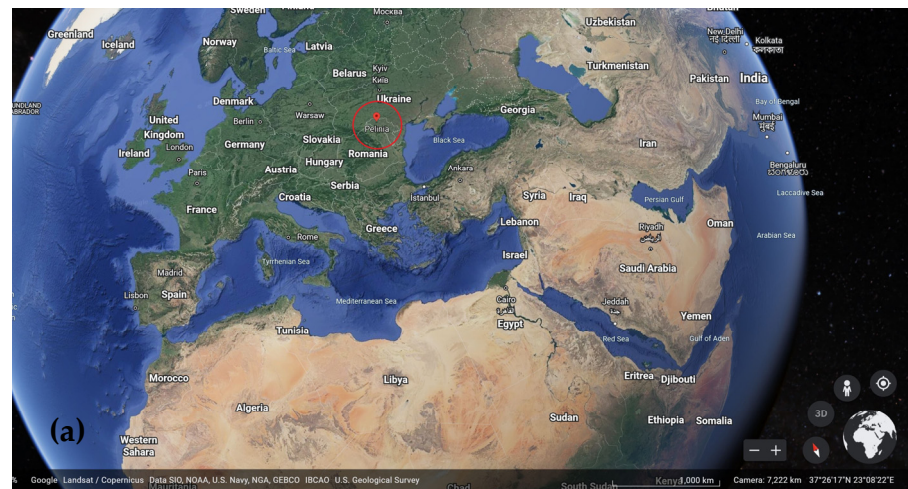
Saharan dust has also been reported to produce effects on marine life as a carrier of microorganisms, causing large-scale diseases such as coral sickness in the Caribbean [6–8]. Conversely, Saharan dust has also proven to be a supplier of nutrients for plants and marine phytoplankton in connection with the soluble iron derivatives it contains [1,12–17]. Further, the presence of Saharan dust particles, due to their optical properties, can cause physical and radiative changes in clouds, with possible influences on temperature and thus climate conditions [2,18–20]. In parallel with the study of the characteristics and influences of Saharan dust on the environment and quality of life, over time, measurements have been made in Europe, especially in western countries, both at ground level and at high altitudes in order to monitor its presence in various locations [21]. As a result, monitoring networks were established. The Aerosol Robotic Network (AERONET) system, founded by NASA together with the French National Center for Scientific Research (CNRS), is a group of such networks for measuring aerosols at ground level [22,23].

2. Materials and Methods

In the AERONET system, standardized and calibrated instruments that perform photometric measurements of solar radiation are used for data acquisition. The data are further processed and made available to researchers and the general public and consist of aerosol optical depth (AOD) and the Angstrom exponent, the latter providing information on the size of particulate matter in the atmosphere based on the optical depth measured at different wavelengths [24]. However, there are situations in which such measurements are missing, and these should be supplemented by other means and methods. In this regard, the novelty of the study consists of an investigation in which the passive optical methods of solar photometry underlying the measurements in the AERONET network are complemented by simulation and physico-chemical analyses. Simulations employing NOAA's hybrid single-particle Lagrangian integrated trajectory (HYSPLIT) model were used to extrapolate the trajectory of Saharan dust in areas where measurements are missing, coupled with spectroscopic methods for the analysis of particles brought by rainfall from the atmosphere to the ground in the area of interest. Scanning electron microscopy coupled with electron dispersive X-ray spectroscopy (SEM-EDS) and Fourier transform infrared spectroscopy (FTIR) analyses performed on rainwater sediments and filtrated material provide information on the chemical composition of particulate matter [25–27] and the influence of the atmosphere on ground level by rainfall through the components of the Saharan dust event.

The air circulation over Europe favored the movement of a Saharan dust cloud from North Africa to Europe between 11 and 12 May 2020. Central Europe was particularly affected, including Romania and the Republic of Moldova. The present study considers the analysis of meteorological data that led to the passage of air mass with Saharan dust content over Pelinia, a village in the Republic of Moldova, in the southeastern part of Europe (Figure 1a). Pelinia is located in Drochia district, in the northern part of the country, at a latitude of 47.8780, longitude of 27.8344, and altitude of 164 m above sea level. The village is located 16 km from the city of Drochia and 142 km from Chisinau, the capital of Republic of Moldova. According to the 2004 census, the population of Pelinia was 7538. Because the AOD station in Chisinau was not operational at the time, data on the trajectory of the Saharan dust particles to the Republic of Moldova were acquired from 7 other stations (one each in Italy, Austria, Slovakia, Poland, and Ukraine and two in Romania). As shown in Figure 1b, the Saharan dust particles reached the northern half of

the Republic of Moldova, including Pelinia village, on 12 May 2020. The analysis period under discussion is 9–13 May 2020.



Barcelona Dust Forecast Center – <https://dust.aemet.es>
 NMMB-MONARCH Res:0.1°x0.1° Dust Surface Conc. ($\mu\text{g}/\text{m}^3$)
 Run: 12h 11 MAY 2020 Valid: 09h 12 MAY 2020 (H+21)

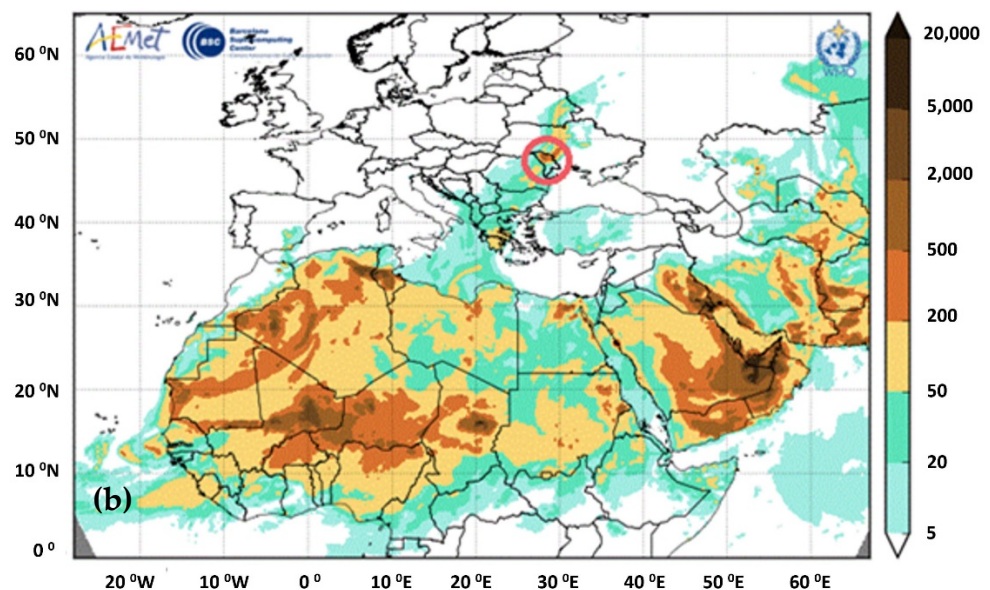


Figure 1. (a) Geographical position of Pelinia village, Republic of Moldova. (b) Presence of Saharan dust particles over Republic of Moldova (12 May 2020) [28].

The Saharan dust cloud moved from North Africa to Eastern Europe, crossing the Mediterranean Sea, Italy, Austria, Croatia, Bosnia and Herzegovina, Serbia, Hungary, Slovakia, Poland, Ukraine, Romania, and the Republic of Moldova, as shown in Figure 1b. The use of backward trajectory analysis by NOAA's HYSPLIT model was considered. This application is a complete system for measuring simple air parcel trajectories to determine the origins of air masses and establish source–receptor relationships. It is used in this study to calculate the trajectories of air masses for the period of interest and determine the direction of the atmospheric currents that favored the movement of the dust cloud over southern Europe to the village of Pelinia.

The trajectory obtained with the HYSPLIT model for the study presented herein is illustrated in Figure 2, with the areas of action being North Africa and central and eastern Europe. Figure 2 shows the route traveled by the Saharan dust cloud, starting from an

altitude of 750 m on 9 May 2020, and continuing its movement to different altitudes until 14 May 2020, when it passed the eastern European area, heading to the Middle East. The map shows that the trajectory obtained at an altitude of 1250 m (graphically represented by a green line) reached the northern part of the Republic of Moldova, confirming the information provided in Figure 1.

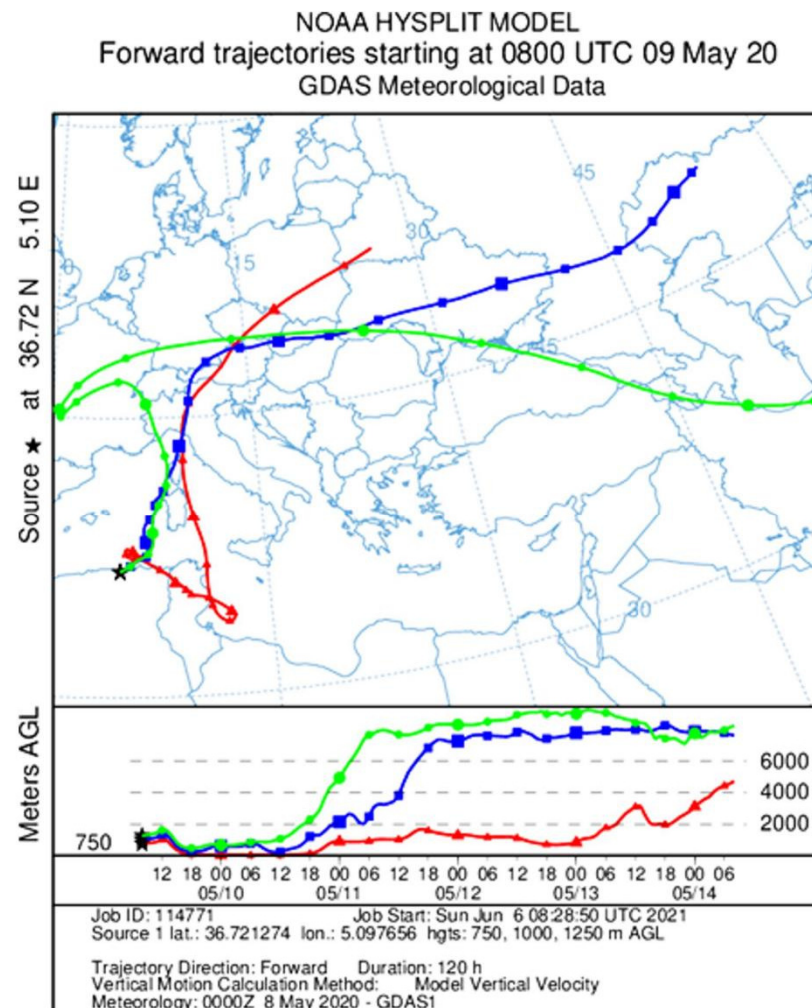


Figure 2. Trajectory of air currents showing the route traveled by the Saharan dust cloud, starting from an altitude of 750 m (red), 1000 m (blue) and 1250 m (green)/(9–13 May 2020).

As not all countries traversed by the Sahara dust cloud had measuring stations, AOD data from the main measuring stations located on the trajectory from the source (North Africa) to the study area (Pelinia, Moldova Republic) were taken for analysis: Rome La Sapienza (Italy), Vienna Univie (Austria), Poprad–Ganovce (Slovakia), Strzyzow (Poland), Kyiv–AO (Ukraine), Magurele (INOE), and Timisoara (Romania). The Republic of Moldova, where the target area is located, has only one AOD measuring station, in Chisinau, but no data were recorded there between November 2018 and June 2021. Therefore, the data for the period and the area of interest (Pelinia) are missing. The AOD measuring stations were selected based on the dust cloud's trajectory toward the northeast. A map with the locations of AOD measuring stations in southeastern Europe is presented in Figure 3. The map includes the position of every AOD station taken into account for this study, considering the dust cloud trajectory area. Other AERONET stations with AOD data were not relevant for this study due to their inadequate location on the trajectory or the unavailability of data for the period of interest at an acceptable quality level.

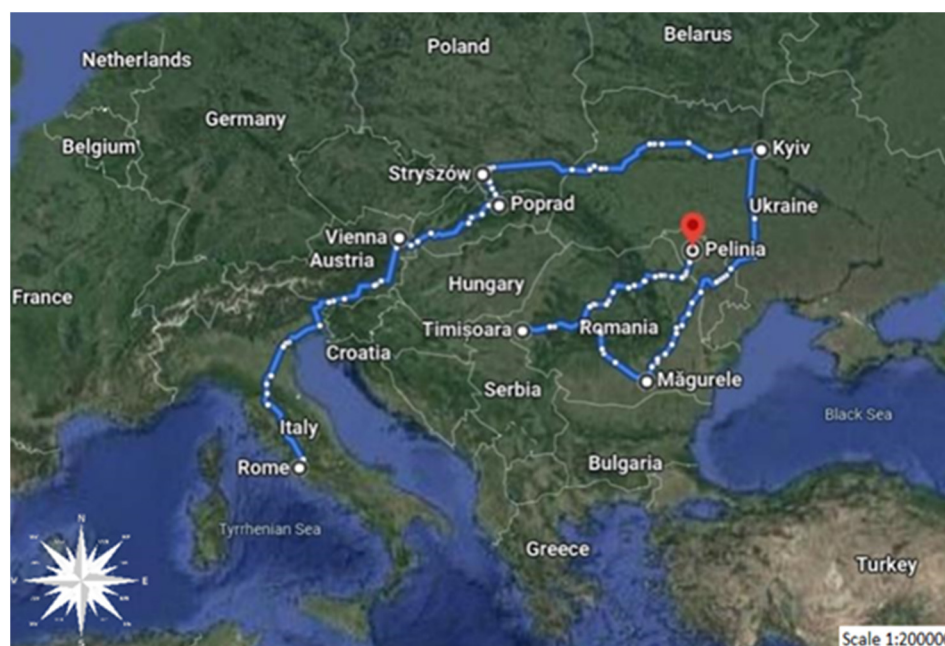


Figure 3. Southeastern Europe: locations of AOD measuring stations.

AERONET data with a quality level of 1.5 (<https://aeronet.gsfc.nasa.gov/>, accessed on 11 August 2021) for Austria, Ukraine, and Poland and quality level of 2.0 (<https://aeronet.gsfc.nasa.gov/>, accessed on 11 August 2021) for Italy, Slovakia, and Romania were acquired from the AERONET website [23] using a CIMEL Electronique CE318-T multiband sun photometer. This device has demonstrated an excellent capacity to monitor aerosols globally. The data were then processed and calibrated by NASA in order to be published at the highest quality. The AOD measurements were made at ground level, at wavelengths between 340 and 1640 nm and Ångström exponent in the ranges of 440–870, 380–500, 440–675, 500–870, 340–440, and 440–870 nm. The Ångström exponent, the second parameter taken into account besides AOD data, describes how the optical thickness of an aerosol typically depends on the wavelength of the light; in other words, it shows the size of the aerosol particles present in the atmosphere.

The AOD 550 nm values were used in this study and an analysis was performed for each geographical area on the trajectory of the Saharan dust storm in the order of traversal.

The analysis consisted of relating the AOD values at 550 nm to the Ångström exponent values at 440–870 nm, based on the uniform spectral dependence of these wavelengths. Where data were not available for the required wavelengths, data at the available wavelengths were analyzed as appropriate.

From a meteorological point of view, in the village of Pelinia, Dochia district, Republic of Moldova, in the first part of 12 May 2020, there was precipitation of 0.2 mm and the minimum temperature recorded was 6 °C. Following the Saharan dust episode on that day, samples of rainwater were collected in Pelinia in a vessel. Later, two samples of solid phase collected from the rainwater were analyzed: one sample was sediment from the bottom of the vessel (SEDIMENT) and the other sample resulted from water filtration (FILTRATE). Scanning electron microscopy coupled with energy-dispersive X-ray spectroscopy (SEM-EDS) was used to analyze the morphology and elemental composition of sediments, along with Fourier transform infrared spectroscopy, a spectroscopic method for detecting functional groups with covalent bonds based on their infrared absorption properties translated into transmittance intensity versus wavenumber (cm^{-1}) spectra.

3. Results and Discussion

3.1. AERONET AOD Data Analysis

The elaboration of this study took into account the collection of meteorological data based on the direction and speed of the wind on the date when the “Saharan smog” reached the Republic of Moldova.

Following data centralization, charts were obtained showing the direction and intensity of air currents that favored the movement of air masses containing dust particles of Saharan origin above Pelinia. Given that the Republic of Moldova is located in the northeastern part of Romania, beyond the eastern Carpathians, it can be determined that this favored a proportion of 45.9% prevailing wind from the northwest, specifically north-northwest (see dark red section in Figure 4c), with 41.7% moving at a rather significant speed of over 6 m/s (Figure 4a). This presumes the presence of Saharan dust in the northern half of the Republic of Moldova. The northwesterly direction remained predominant in terms of the degree of atmospheric stability. The highest proportion of 33.5% was identified at the level of class 2, with a proportion of 16.7% blowing from the southwest, but also from the south, southeast, east, and east-southeast. A proportion of almost 30% at stability class 5 blew from the northeast (see green section in Figure 4d). Regardless of the cardinal point from which the wind was blowing, the atmospheric stability was low in the village of Pelinia on 12 May 2020, because in over 50% of the wind directions it was identified as atmospheric stability class 1 to 3 (low level).

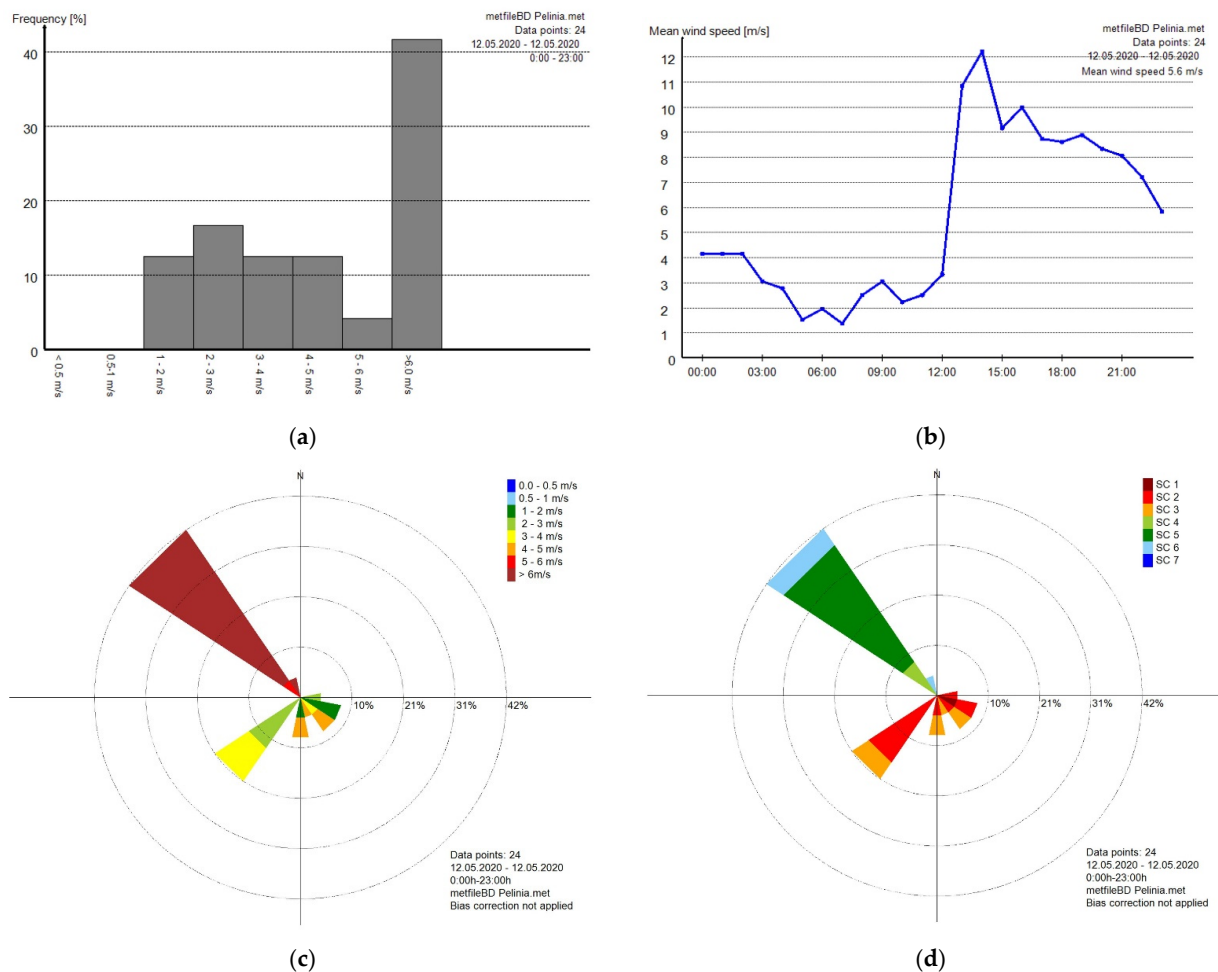


Figure 4. Meteorological parameters for Pelinia village, Republic of Moldova, on 12 May 2020: (a) frequency of wind intensity; (b) hourly frequency of wind intensity; (c) wind rose; (d) stability of wind rose.

This low stability produced an environment that favored the movement and deposition of Saharan dust in the area of Pelinia village. The wind blew at a speed over 6 m/s in a proportion of more than 40%, thus had high intensity, which also contributed to the movement and deposition of Saharan dust in those areas. In the first part of the day, the air circulation was predominantly from the southeast in the interval 07:00–13:00 at an approximate intensity of 1.5 to 3.3 m/s, while in the second part of the day the wind was predominantly from the northwest, starting from 12:00 until the end of the day (Figure 4b). Between 14:00 and 15:00, there was a slight change in the direction of the air mass, from northwest to north-northwest. It practically did not matter in the analysis of air mass direction, because it did not affect the main cardinal point of the wind, specifically the northeast. The intensity recorded in this direction was on average 8.9 m/s, with a maximum of around 12.22 m/s at 14:00 (Figure 4b).

For the AOD analysis, the data were downloaded from the AERONET website for the period 9–13 May 2020 and grouped according to the countries where the measurements were made at photometric stations, in order to follow the direction of movement of the Saharan dust cloud. The period chosen for the analysis includes the date of interest for the present study, 12 May 2020, when the presence of the Saharan dust cloud was detected above the village of Pelinia, Republic of Moldova. The countries on the trajectory of the dust storm were selected, meaning in the northeast direction from North Africa to eastern and southeastern Europe: Italy, Austria, Slovakia, Poland, Ukraine, and Romania.

Next, the tables and charts resulting from the centralization of AOD data and the Angstrom exponent for each of country are presented.

3.1.1. Italy–AOD Data Level 2.0

For Italy, the Rome–la Sapienza measuring station, with data processed at the highest quality level of 2.0, was taken into account. The information is presented in Figures 5 and 6a, which show the AOD values, the Angstrom exponent, and the AOD evolution according to the Angstrom exponent for the period 9–12 May 2020, respectively.

The average AOD values were recorded daily, and the Angstrom exponent was provided at wavelengths between 340 and 1640 nm. The highest values were recorded on 10 May 2020, when, according to data provided by the website <https://dust.aemet.es/forecast> (accessed on 11 August 2021), the Saharan smog was above Italy, continuing its route to Eastern Europe. The highest daily average values were recorded on this date regardless of the wavelength. The AOD value at 500 nm was 0.228148 and the Angstrom exponent value at 440–870 nm was 0.429631. It can be seen that on 10 May 2020, when the highest AOD value was registered, the Angstrom exponent was at its lowest value.

The decreasing trend of AOD at 500 nm from 9–12 May 2020 is shown in Figure 6a.

The decrease in AOD values at 500 nm for the same period indicates that the amount of dust particles in the atmosphere decreased, which explains the presence of the Saharan dust cloud above Italy at the beginning of the period. Its advance toward Eastern Europe causes the AOD values at 500 nm to drop, indicating a lack of dust particles in the atmosphere. With the decreased AOD values, from left to right on the chart, there is an increase in the value of the Angstrom exponent, which may be an indication of the presence of other light-absorbing components, such as mineral dust and organic aerosols. Taking into account the geographical position of Italy and its capital, Rome, where the Rome–La Sapienza measuring station selected for this study is located, the Saharan dust cloud was present in the area on 10 May 2020, the date on which the highest AOD values at 500 nm and the lowest Angstrom exponent values at 440–870 nm were recorded. The low average values of the Angstrom exponent indicate the presence of coarse particles in the atmosphere up to 10 μm in diameter (i.e., PM_{10}).

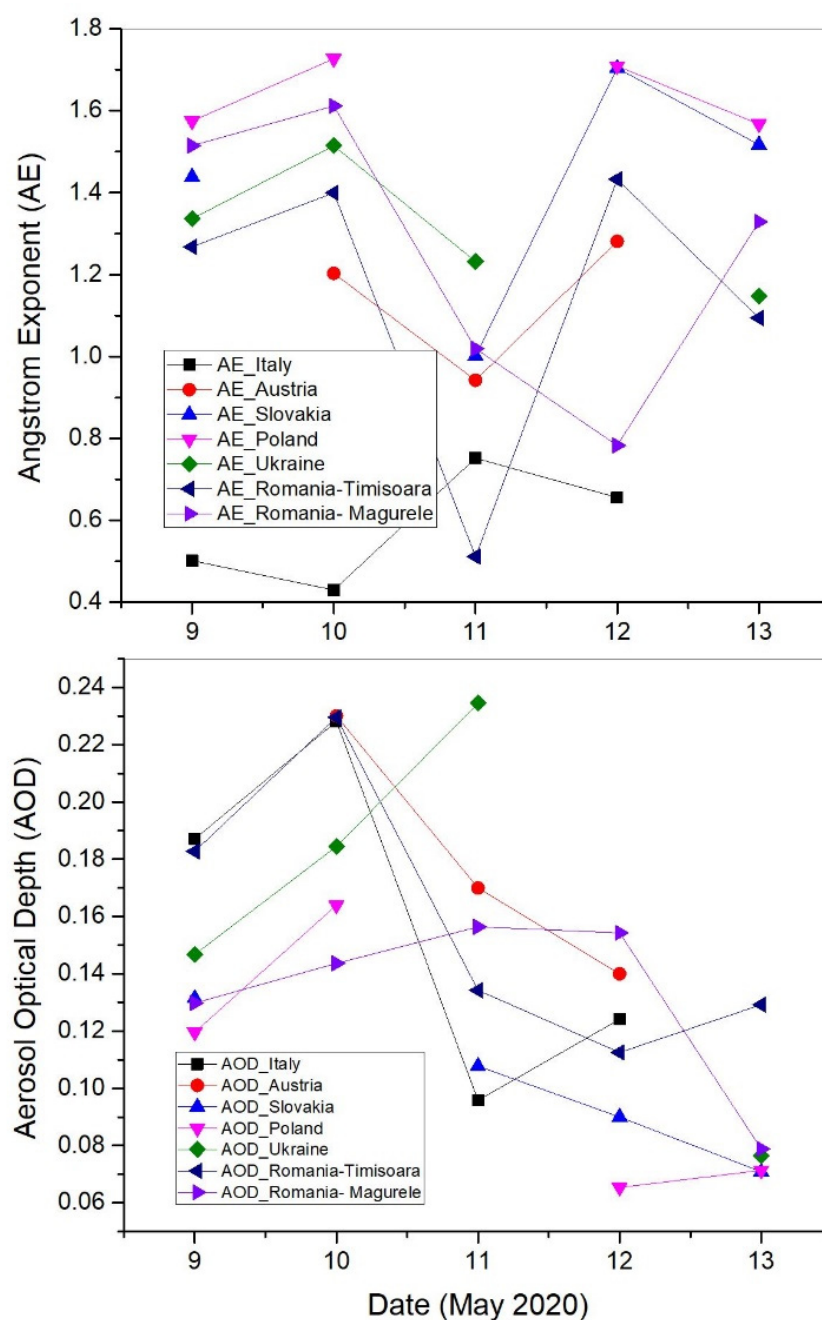


Figure 5. AOD and Angstrom exponent values at 440–870 nm, 9–13 May 2020, at Rome–La Sapienza (level 2.0), Austria–Vienna–Univie (level 1.5), Slovakia–Poprad–Ganovce (level 2.0), Poland–Strzyzow (level 1.5), Ukraine–Kiev (level 1.5), Romania–Timisoara (level 2.0), and Romania–Magurele (level 2.0).

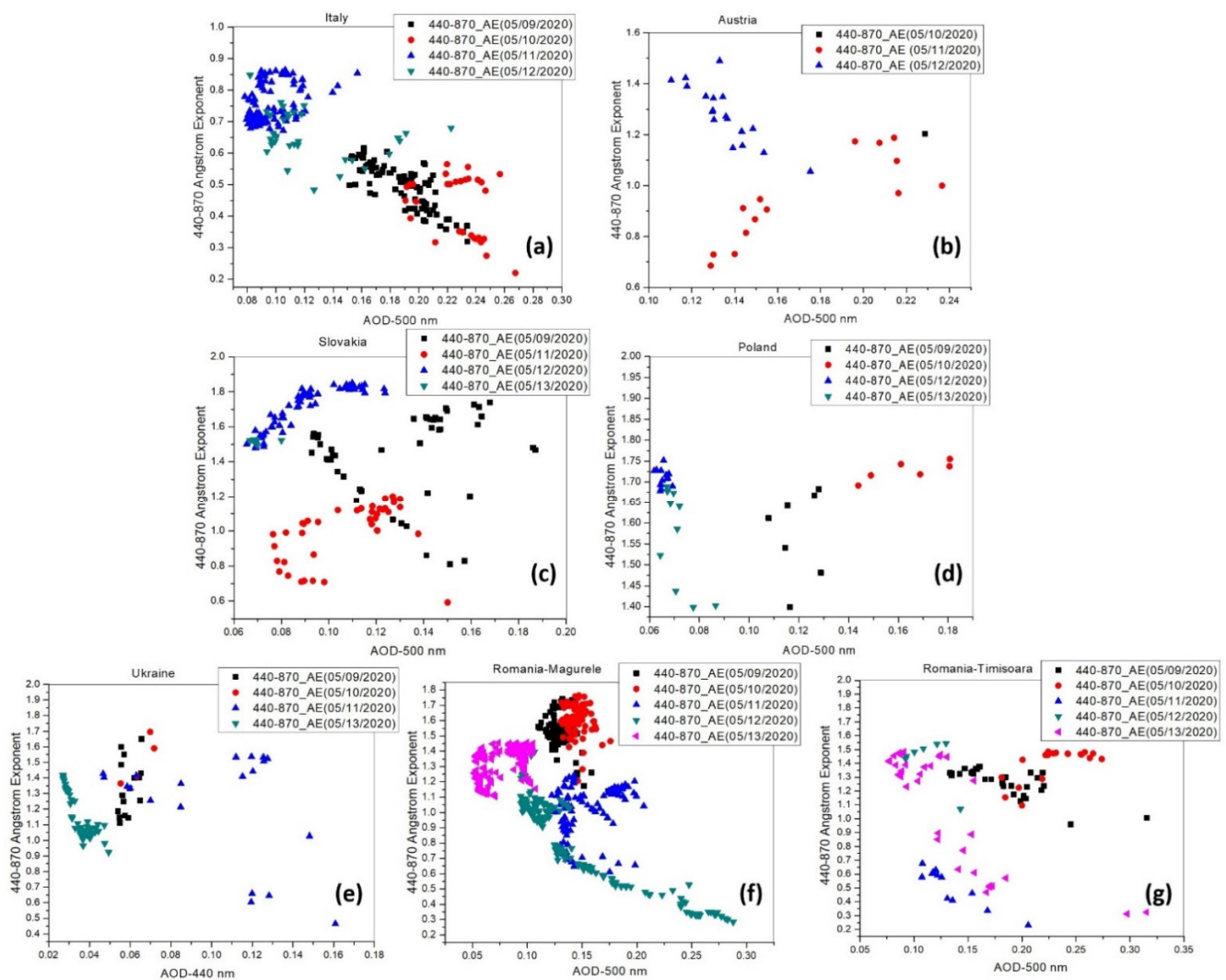


Figure 6. Evolution of AOD at 500 nm depending on Angstrom exponent at 440–870 nm (9–13 May 2020): (a) Rome–La Sapienza (level 2.0), (b) Austria–Vienna–Univie (level 1.5), (c) Slovakia–Poprad–Ganovce (level 2.0), (d) Poland–Strzyzow (level 1.5), (e) Ukraine–Kiev (level 1.5), (f) Romania–Magurele (level 2.0), and (g) Romania–Timisoara (level 2.0).

3.1.2. Austria: AOD Data Level 1.5

In Austria, data were collected from a single measuring station, Vienna–UNIVIE. The data quality was inferior compared to Italy, falling to 1.5. Here, were recorded the highest average daily AOD levels, regardless of wavelength, on the same date, 10 May 2020, which supports that on that day the Saharan dust cloud crossed both Italy and Austria, with suitable weather conditions for travel speed. The AOD value at 500 nm was 0.228671, higher than the values recorded at the Italian stations, and the Angstrom exponent value at 440–870 nm was 1.20295, indicating the presence of fine particles with a diameter between 0.1 and 1.0 μm (accumulation type).

Figure 6b shows a decreasing trend of AOD at 500 nm starting from 10 to 12 May 2020, the same as for the data obtained at the station in Italy. This decrease in respective values from one day to the next indicates that the amount of dust particles in the atmosphere was reduced, and the Saharan dust cloud left the airspace of Austria on 11 May 2020. As the AOD value at 500 nm dropped from left to right, an increase in the Angstrom exponent value is observed. This also shows the presence of mineral dust and organic aerosols in the atmosphere.

On the reference date, 10 May, the highest AOD levels, the values the Angstrom exponent did not register the lowest values, as for Italy.

3.1.3. Slovakia: AOD Data Level 2.0

In Slovakia, as in Austria, data were collected and analyzed from a single station, Poprad-Ganovce. The Saharan dust cloud reached above Slovakia on 11 May 2020, which became the reference date for the measured data. On that date, the average daily AOD levels were lower than the levels registered in Italy and Austria, with AOD at 500 nm equal to 0.107785, while the Angstrom exponent at 440–870 nm reached the lowest value of 1.001314.

On the reference date, AOD registered the lowest values compared to the data of the next period, 9–13 May 2020; for the Angstrom exponent, the lowest values are registered for 11 May compared to the remaining data of the period of interest.

Figure 6c shows the same decreasing trend of AOD values at 500 nm from 9–13 May 2020. In the atmosphere above Slovakia, the presence of the Saharan dust cloud was noted by the detection of mineral dust and organic aerosols. From 11 May until 13 May, AOD values decreased constantly, indicating a decrease in the abundance of dust particles in the atmosphere; in other words, the air became cleaner with the passage of the dust cloud. The average value of the Angstrom exponent at 440–870 nm was above 1, indicating fine particles in the atmosphere.

3.1.4. Poland: AOD Data Level 1.5

In Poland, data from the Strzyzow station, at a quality level of 1.5, were analyzed for the period 9–13 May 2020. Following the trajectory analysis of the Saharan dust storm shown in Figure 1, it can be observed that it crossed the surface of Poland on 11 May.

Taking into account that the dataset downloaded from the AERONET website does not contain information for the date of interest, 11 May, the observation refers to the previous days. Therefore, Figure 5 shows that the highest AOD value at 500 nm (0.164001) was recorded on 10 May, continuing a positive trend from the previous day.

The Angstrom exponent value at 440–870 nm was equal to 1.72702. Figure 6d graphically represents the AOD 500 nm data in correlation with the Angstrom exponent 440–870 nm data, highlighting a constant trend, thus indicating a small amount of dust particles in the atmosphere. This can be explained by the fact that the area of Poland was on the periphery of the Saharan dust storm trajectory, and the dust cloud moved with higher intensity to the more southern areas in the north of Romania and the Republic of Moldova. In correlation with AOD values at 500 nm, the recorded Angstrom exponent values at 440–870 nm were above 1, indicating that there were fine dust particles in the atmosphere.

3.1.5. Ukraine: AOD Data Level 1.5

The next area chosen for AOD data collection was Ukraine, through its only station in Kiev. As shown in Figure 5a,b and Figure 6e, the period with available data was 9–13 May 2020. Considering that Ukraine is located to the north of the Republic of Moldova, we can assume that the dust cloud entered this country through northern Romania and southern Ukraine, which is why the data available at Kiev station were chosen for analysis. Aeronet data are not available for AOD at 500 nm, the parameter analyzed at the rest of the stations on the trajectory of Saharan smog. Therefore, the AOD value at 440 nm was taken into account, the increase of which can be seen in the chart presented in Figure 5, due to the multitude of dust particles existing in the atmosphere over the examined area on 10 May. At the same time, the Angstrom exponent also has a high value, explaining the presence of fine dust particles in the atmosphere.

The constant and slightly decreasing trend observable in Figure 6e, which shows the AOD values at 440 nm correlated with the Angstrom exponent values at 440–870 nm, explains and confirms that at the beginning and middle of the period of interest, 9–13 May 2020,

the Saharan dust storm was present in the Ukraine area with an abundance of fine dust particles, and headed to the Republic of Moldova on 12 May.

3.1.6. Romania: AOD Data Level 2.0

In Romania, the AOD and Angstrom exponent data from two stations, Magurele and Timisoara, at the highest quality level of 2.0, were collected and centralized. The reference period was 9–13 May 2020, and the wavelength ranges were 340–1640 nm for AOD and 340–440 and 500–870 nm for the Angstrom exponent. The data obtained are displayed in Figures 5 and 6f for Magurele and Figure 6g for Timisoara.

The Saharan dust cloud was located above Romania on 12 May, entering through the western part and crossing the country through the northern part, later entering the Republic of Moldova. As can be seen in the charts in Figures 5 and 6g, according to the data recorded at Magurele, on 12 May, the AOD value at 500 nm was 0.154303, while the recorded Angstrom exponent value at 440–870 nm was 0.78643. Because these values are less than 1, it can be inferred that the dust particles in the atmosphere were coarse, with a diameter up to μm (PM_{10}).

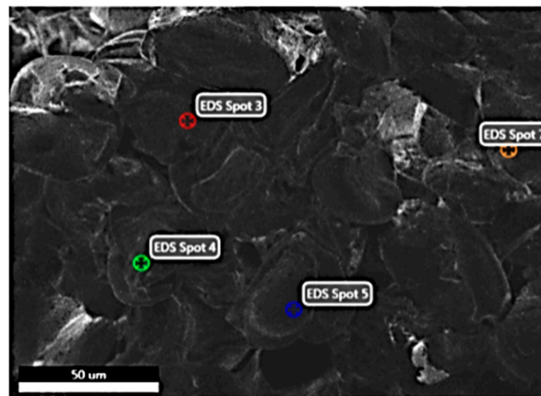
Figure 6f shows the AOD values at 500 nm in correlation with the Angstrom exponent values at 440–870 nm, showing a marked decreasing trend, explaining the existence of dust particles in the atmosphere during the period of interest. The actual decreasing trend emphasizes the direct proportionality between AOD and the Angstrom exponent, the latter also registering lower values around 12 May 2020, explaining the presence in the atmosphere of coarse particles (Figure 6g). Regarding the data collected from the Timisoara station, we have the following statistics: on 12 May, the AOD value at 500 nm was 0.112543, and the Angstrom exponent value at 440–870 nm was 1.432633. Along with the same decreasing trend for the correlation between AOD at 500 nm and the Angstrom exponent at 440–870 nm, the following can be stated: in the Timisoara area, the Saharan smog was present on 12 May 2020, characterized by an abundance of mineral dust and atmospheric aerosols in the atmosphere. Values of the Angstrom exponent above 1 confirm the existence in the atmosphere of fine particles of Saharan dust.

In conclusion, due to the geographic position of the Timisoara area farther north of the Magurele station, it was on the trajectory of the Saharan dust cloud and, implicitly, closer to the area where it passed on its way through the northern part of Romania to the northern part of the Republic of Moldova, where Pelinia is located. The size of particles arriving to the Republic of Moldova was more likely the influence of the dust storm passing through Timisoara. This means that we would expect to find fine particles in Pelinia. Yet, the influence of coarse particles heading through Magurele (Romania) to the Republic of Moldova would be expected as well. The Republic of Moldova was on the trajectory of the Saharan dust cloud, according to the weather data presented herein, but without AERONET data on AOD values, investigations by electron microscopy and EDX and FTIR spectroscopy must fill in the information on extended areas under the effect of the episode.

3.2. Rainwater Analysis after the Saharan Dust Episode in Pelinia

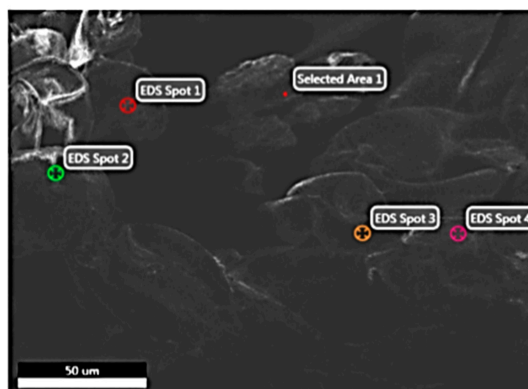
Based on the SEM-EDS analyses (Figures 7–10) of different areas and spots of the SEDIMENT and FILTRATE samples, a non-uniform distribution is noticed in terms of both morphology and elemental composition. A high carbon content (over 40%, and even as much as 70%) in spots of R1A1, R1A2, R1A3, R3A1, and R4A1 may denote vegetal or other biological material, also observed in the SEM images. Based on the morphology and elemental composition, the analyzed spots of areas R2A1 and R2A2 with carbon content between 20 and 50% may be assigned to carbonaceous crystalline structures, including carbonates and carboxylates. Based on the granular morphology, the spots in areas R2A3, R3A2, R3A3, and R3A4 with low carbon but high oxygen content with somewhat uniform dispersion of elements may be assigned to oxides, sulfates, and carbonates, as well as hydroxide compounds of calcium, sodium, aluminum, potassium, magnesium, and iron. Consistent quantities of sand particles (SiO_2) and silicates are indicated by these results.

Finally, a category with average carbon content between 5 and 20% in spots that exhibit aggregated structures is noted in areas R4A3 and R4A2. The aggregates may be assigned to a flocculation/coagulation process due to sodium aluminate [25], as noticed by the increased sodium and aluminum content in the elemental composition.



R1A1	Spot 3		Spot 4		Spot 5		Spot 7	
	Weight (%)	Atomic (%)	Weight (%)	Atomic (%)	Weight (%)	Atomic (%)	Weight (%)	Atomic (%)
C	52.91	60.71	60.47	67.76	62.97	70.14	55.37	67.99
O	43.28	37.28	36.35	30.57	33.53	28.04	21.89	20.18
Na	1.27	0.76	1.28	0.75	1.46	0.85		
Mg	0.21	0.12	0.27	0.15	0.13	0.07		
Al			0.09	0.05	0.10	0.05		
Si	2.30	1.13	1.46	0.70	1.70	0.81	22.02	11.56
Ca	0.03	0.01	0.07	0.02	0.11	0.04	0.73	0.27

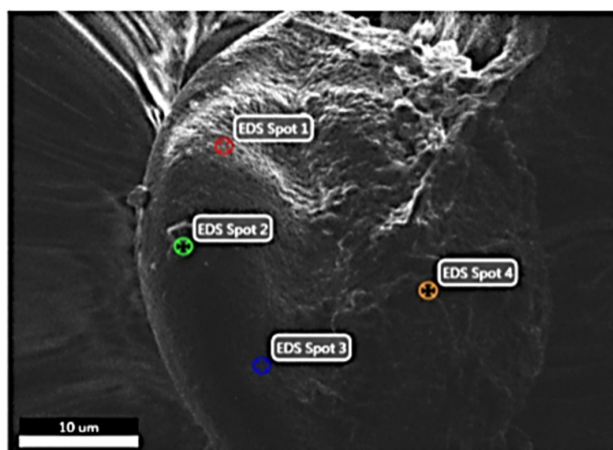
(a)



R1A2	Spot 1		Spot 2		Selected		Spot 3		Spot 4	
	Weight [%]	Atomic [%]	Weight [%]	Atomic [%]	Weight [%]	Atomic [%]	Weight [%]	Atomic [%]	Weight [%]	Atomic [%]
C	41.71	47.76	32.98	40.48	23.36	31.54	65.27	72.55	43.19	49.10
N	21.54	21.16	8.52	8.96					25.59	22.02
O	35.40	30.43	49.86	45.93	55.51	56.26	30.54	25.48	33.35	28.46
Na	0.57	0.34	0.82	0.53	0.74	0.52	0.65	0.38	0.38	0.23
Mg			0.58	0.35	0.54	0.36	0.30	0.16		
Al	0.09	0.05	2.54	1.39	6.10	3.66	0.52	0.26		
Si	0.22	0.11	4.01	2.11	12.06	6.97	1.35	0.64	0.16	0.08
P	0.07	0.03					0.25	0.11	0.05	0.02
S	0.05	0.02	0.13	0.06			0.27	0.11		
K			0.29	0.11	1.61	0.67				
Cl	0.07	0.03					0.50	0.19	0.11	0.04
Ca	0.08	0.03	0.13	0.05			0.37	0.12	0.10	0.04
Fe	0.18	0.04	0.14	0.04	0.09	0.03			0.06	0.01
Ni	0.02	0.00							0.00	0.00

(b)

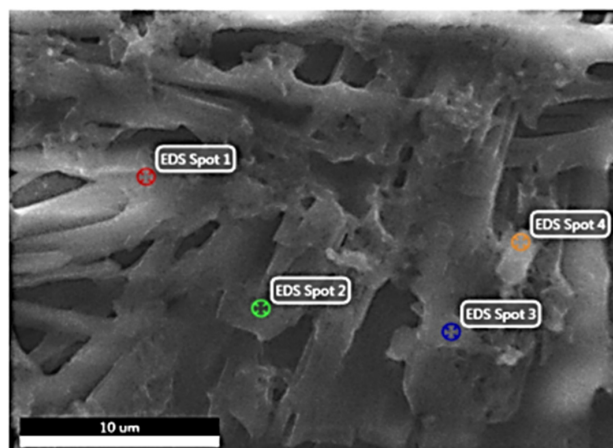
Figure 7. Cont.



R1A3	Spot 1		Spot 2		Spot 3		Spot 4	
	Weight [%]	Atomic [%]	Weight [%]	Atomic [%]	Weight [%]	Atomic [%]	Weight [%]	Atomic [%]
C	66.62	73.15	34.88	44.34	69.38	75.67	31.11	38.03
N							7.36	7.71
O	32.49	26.78	48.09	45.91	28.24	23.12	55.56	50.99
W	0.88	0.06						
F			1.59	1.27				
Na					0.66	0.37	0.72	0.46
Mg			0.91	0.57	0.21	0.11	0.52	0.31
Al			3.45	1.95	0.48	0.23	2.01	1.10
Si			10.59	5.76	1.04	0.49	2.62	1.37
K			0.25	0.10				
Ca			0.25	0.09			0.00	0.00
Fe							0.10	0.03

(c)

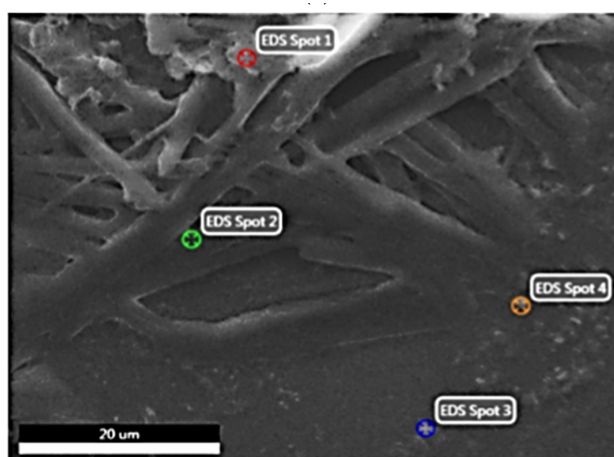
Figure 7. Report 1 of SEM-EDS results of analyzed areas of dry material decanted from rainwater: (a) R1A1; (b) R1A2; (c) R1A3.



R2A1	Spot 1		Spot 2		Spot 3		Spot 4	
	Weight [%]	Atomic [%]	Weight [%]	Atomic [%]	Weight [%]	Atomic [%]	Weight [%]	Atomic [%]
C	24.39	33.09	26.34	35.34	29.49	38.82	20.38	27.28
O	43.78	44.59	43.04	43.35	42.95	42.46	58.74	59.03
Na	30.08	21.32	29.66	20.79	26.10	17.96	15.37	10.75
Mg	0.40	0.27	0.30	0.20	0.26	0.17	0.77	0.51
Si	1.02	0.59	0.34	0.19	0.68	0.38	3.08	1.76
Cl					0.09	0.04		
K					0.08	0.03		
Ca	0.34	0.14	0.33	0.13	0.35	0.14	1.66	0.67

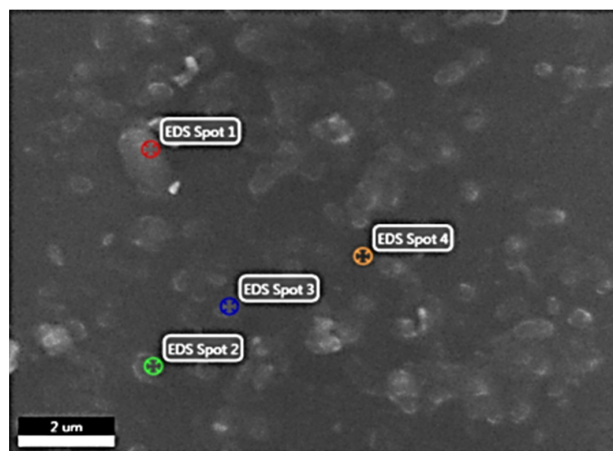
(a)

Figure 8. Cont.



R2A2	Spot 1		Spot 2		Spot 3		Spot 4	
	Weight [%]	Atomic [%]	Weight [%]	Atomic [%]	Weight [%]	Atomic [%]	Weight [%]	Atomic [%]
C	26.21	34.08	21.88	28.38	48.56	57.42	27.88	36.08
O	54.31	53.02	64.78	63.07	40.59	36.03	53.48	51.96
Na	16.47	11.19	8.85	5.99	9.41	5.82	14.40	9.73
Mg	0.45	0.29	0.74	0.47	0.23	0.13	0.83	0.53
Al			0.19	0.11			0.37	0.21
Si	2.56	1.43	3.56	1.98	1.05	0.53	1.74	0.96
S					0.07	0.03		
Cl					0.10	0.04	0.22	0.09
K							0.14	0.05
Ca							0.96	0.37

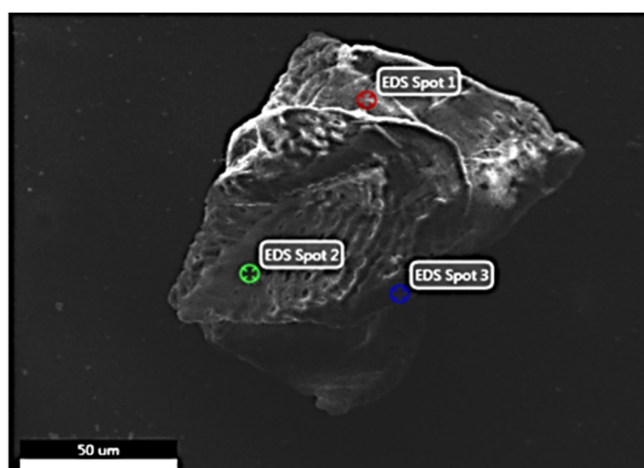
(b)



R2A3	Spot 1		Spot 2		Spot 3		Spot 4	
	Weight [%]	Atomic [%]	Weight [%]	Atomic [%]	Weight [%]	Atomic [%]	Weight [%]	Atomic [%]
C	6.33	9.55			4.00	6.06	3.29	4.98
O	53.23	60.33	61.69	72.35	60.15	68.44	62.62	71.19
F	4.94	4.72						
Na	16.75	13.21	12.90	10.53	14.81	11.73	11.40	9.02
Mg	2.22	1.66	3.02	2.33	2.50	1.87	2.59	1.94
Al	0.26	0.18	0.39	0.27	0.29	0.20	0.29	0.20
Si	15.38	9.93	21.09	14.09	17.55	11.37	19.04	12.33
S	0.23	0.13						
Ca	0.66	0.30	0.91	0.43	0.70	0.32	0.76	0.34

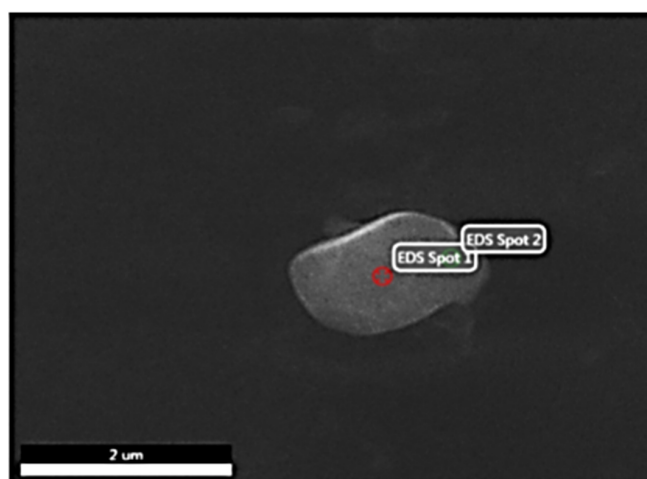
(c)

Figure 8. Report 2 of SEM-EDS results of analyzed areas of dry material decanted from rainwater: (a) R2A1; (b) R2A2; (c) R2A3.



R3A1	Spot 1		Spot 2		Spot 3	
	Weight [%]	Atomic [%]	Weight [%]	Atomic [%]	Weight [%]	Atomic [%]
C	53.16	60.85	57.07	64.33	53.44	61.26
O	43.26	37.18	40.68	34.42	42.23	36.34
Na	1.95	1.17	1.47	0.86	2.60	1.56
Si	1.63	0.80	0.76	0.38	1.73	0.85

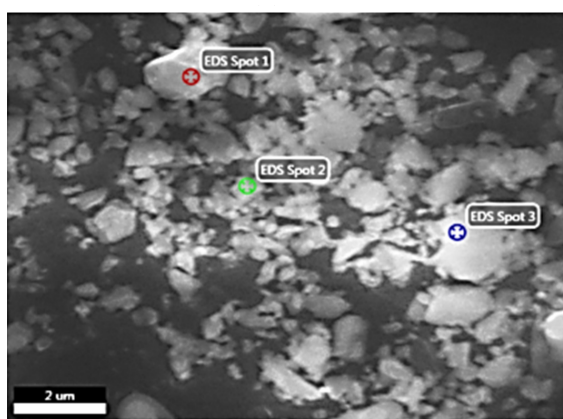
(a)



R3A2	Spot 1		Spot 2	
	Weight [%]	Atomic [%]	Weight [%]	Atomic [%]
C			7.64	12.85
O	45.75	59.03	35.16	44.38
Na	13.16	11.82	24.18	21.24
Mg	3.07	2.61	2.30	1.91
Al	0.96	0.73	0.86	0.64
Si	29.08	21.37	14.04	10.09
S	2.55	1.64	1.69	1.06
Cl			10.62	6.05
K	0.41	0.22	0.29	0.15
Ca	5.01	2.58	3.22	1.62

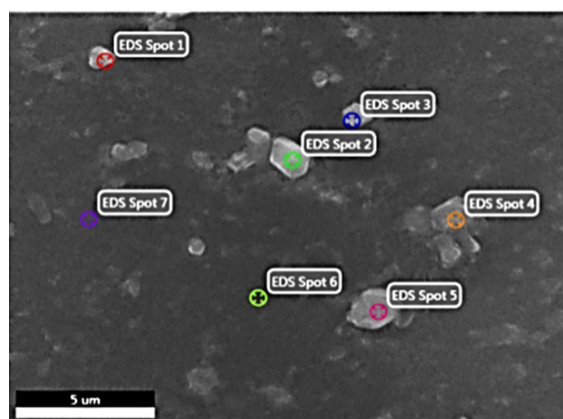
(b)

Figure 9. Cont.



R3A3	Spot 1		Spot 2		Spot 3	
	Weight [%]	Atomic [%]	Weight [%]	Atomic [%]	Weight [%]	Atomic [%]
C	13.54	18.58	23.55	30.19	12.15	17.59
O	66.51	68.51	65.23	62.78	59.65	64.86
Na	9.07	6.51	6.61	4.43	8.25	6.24
Mg	1.21	0.82	0.71	0.45	1.29	0.92
Al	0.59	0.36	0.57	0.33	0.46	0.29
Si	8.45	4.96	3.33	1.83	10.13	6.27
S	0.13	0.07			2.77	1.50
Cl					0.10	0.05
K					0.17	0.07
Ca	0.51	0.21			5.04	2.19

(c)

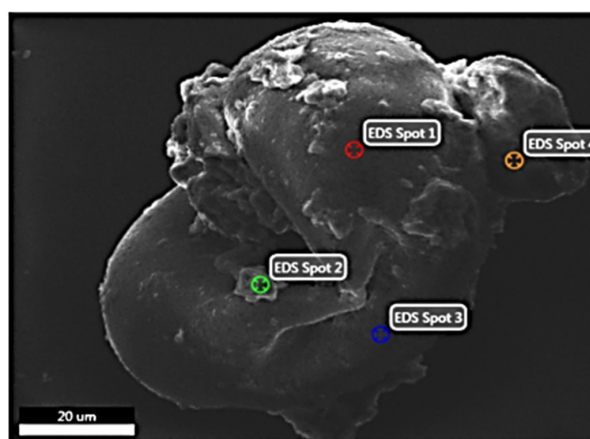


R3A4	Spot 1		Spot 2		Spot 3		Spot 4	
	Weight [%]	Atomic [%]	Weight [%]	Atomic [%]	Weight [%]	Atomic [%]	Weight [%]	Atomic [%]
C	4.13	6.53	4.19	6.79	5.14	8.11	3.51	5.65
O	60.93	72.32	52.31	63.68	56.17	66.51	54.90	66.28
Na	5.46	4.51	11.77	9.98	9.70	7.99	12.06	10.13
Mg	0.86	0.68	1.20	0.96	1.45	1.13	1.34	1.07
Al	0.35	0.25	0.29	0.21	0.63	0.44	0.27	0.20
Si	5.38	3.64	13.11	9.09	11.13	7.51	11.23	7.72
S	10.38	6.14	6.82	4.14	6.86	4.05	6.68	4.02
Cl	0.18	0.09	2.13	1.17	0.65	0.35	1.69	0.92
K			0.24	0.12	0.18	0.09	0.20	0.10
Ca	12.34	5.84	7.96	3.87	8.09	3.82	8.12	3.91

R3A4	Spot 5		Spot 6		Spot 7	
	Weight [%]	Atomic [%]	Weight [%]	Atomic [%]	Weight [%]	Atomic [%]
C	3.34	5.33	3.09	4.89		
O	61.32	73.48	65.35	77.53	21.30	34.44
Na	5.28	4.41	2.12	1.75	3.96	4.46
Mg	0.42	0.33	0.18	0.14	1.36	1.44
Al			0.13	0.09	0.92	0.88
Si	4.36	2.98	1.58	1.07	34.94	32.18
S	11.21	6.70	12.41	7.35	14.63	11.81
Cl	0.64	0.35	0.18	0.09		
K					0.84	0.56
Ca	13.43	6.43	14.96	7.09	22.05	14.23

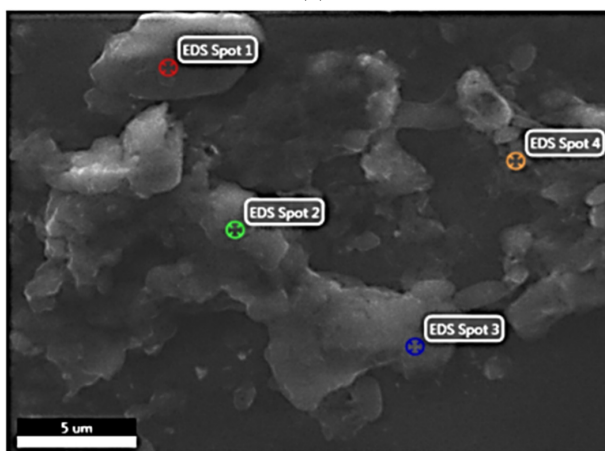
(d)

Figure 9. Report 3 of SEM-EDS results of analyzed areas of dry material decanted from rainwater: (a) R3A1; (b) R3A2; (c) R3A3; (d) R3A4.



R4A1	Spot 1		Spot 2		Spot 3		Spot 4	
	Weight [%]	Atomic [%]	Weight [%]	Atomic [%]	Weight [%]	Atomic [%]	Weight [%]	Atomic [%]
C	64.47	74.48	39.76	51.34	38.91	48.68	43.20	57.42
O			36.88	35.76	44.53	41.81	32.63	30.45
F			1.13	0.92			2.60	2.04
Na	17.70	14.23	2.71	1.83	6.57	4.29	2.14	1.39
Mg	1.68	1.28	0.51	0.32	0.97	0.60	0.32	0.19
Al	0.94	0.64	4.08	2.34	0.35	0.19	0.68	0.37
Si	11.49	7.56	10.17	5.62	7.11	3.81	15.01	7.98
P			0.07	0.03				
S			0.29	0.14	0.15	0.07		
Cl	1.48	0.77	1.21	0.53	0.36	0.15	0.11	0.05
K			1.31	0.52	0.09	0.04		
Ca	2.24	1.03	1.06	0.41	0.97	0.36	0.31	0.12
Fe			0.83	0.23				

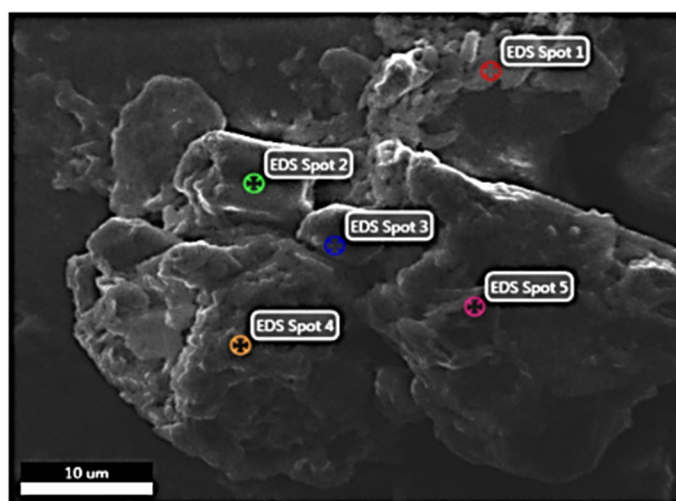
(a)



R4A2	Spot 1		Spot 2		Spot 3		Spot 4	
	Weight [%]	Atomic [%]	Weight [%]	Atomic [%]	Weight [%]	Atomic [%]	Weight [%]	Atomic [%]
C	21.19	27.66	12.27	17.91	10.05	13.85	18.15	24.45
N	9.43	10.55						
O	52.73	51.66	57.03	62.51	71.78	74.26	62.63	63.36
Na	6.36	4.34	5.69	4.34	7.90	5.69	8.16	5.74
Mg	1.39	0.90	2.41	1.74	1.66	1.13	1.48	0.99
Al	2.69	1.56	6.50	4.23	0.26	0.16	0.33	0.20
Si	5.36	2.99	12.21	7.62	8.35	4.92	8.80	5.07
S	0.09	0.04	0.67	0.36			0.12	0.06
Cl	0.29	0.13	0.29	0.14				
K	0.13	0.05	0.73	0.33				
Ca	0.27	0.10	0.97	0.43			0.33	0.13
Fe	0.09	0.02	1.23	0.39				

(b)

Figure 10. Cont.



R4A3	Spot 1		Spot 2		Spot 3		Spot 4		Spot 5	
	Weight [%]	Atomic [%]	Weight [%]	Atomic [%]	Weight [%]	Atomic [%]	Weight [%]	Atomic [%]	Weight [%]	Atomic [%]
C	11.41	16.72	5.52	8.38	22.65	30.81	22.47	30.28	12.54	18.16
O	58.59	64.46	57.89	65.89	54.28	55.42	56.91	57.57	59.47	64.67
Na	1.65	1.27	12.58	9.97	5.42	3.85	3.50	2.46	1.86	1.41
Mg	1.40	1.01			0.79	0.53	1.12	0.75	1.65	1.18
Al	4.28	2.79	7.33	4.95	4.95	3.00	4.69	2.81	7.12	4.59
Si	20.70	12.97	16.68	10.81	8.99	5.23	9.25	5.33	14.04	8.70
S					0.14	0.07	0.09	0.05		
Cl	0.06	0.03			0.32	0.15	0.24	0.11	0.21	0.10
K	0.85	0.38			0.74	0.31	0.67	0.28	1.27	0.57
Ca	0.18	0.08			1.16	0.47	0.50	0.20	0.42	0.18
Fe	0.87	0.27			0.56	0.16	0.55	0.16	1.43	0.45

(c)

Figure 10. Report 1 of SEM-EDS results of analyzed areas of dry material decanted from rainwater: (a) R1A1; (b) R1A2; (c) R1A3.

Sodium aluminate is a binder or flocculent agent for organic compounds, and for inorganic compounds that contain functional groups with covalent bonds (sulfates, carbonates, etc.).

Particle size measured on SEM images with ToupView software showed values between 5.83 and 11.9 μm for R1A1, 13.59 μm for R1A2, between 0.67 and 0.97 μm for R1A3, 0.17 μm for R2A3, 0.87 μm for R3A2, between 0.25 and 0.83 μm for R3A3, between 0.33 and 0.58 μm for R3A4, and between 2.5 and 8.3 μm for R4A3. With the exception of particle sizes related to R4A3 of 2.5 and 8.3 μm , which were the result of agglomeration phenomena (by flocculation or coagulation processes) that took place in the presence of water, all other measured particles showed the influence of the Saharan dust cloud heading to Pelinia via both Timisoara (fine particles, submicrometric) and Magurele (coarse particles, up to 10 μm).

The FTIR spectra for the solid material from the rainwater sample (filtrate and sediment) collected in Pelinia during the Saharan dust episode are almost identical (Figure 11). The small variations are mainly due to the uneven distribution of compounds in the mass of solid material collected.

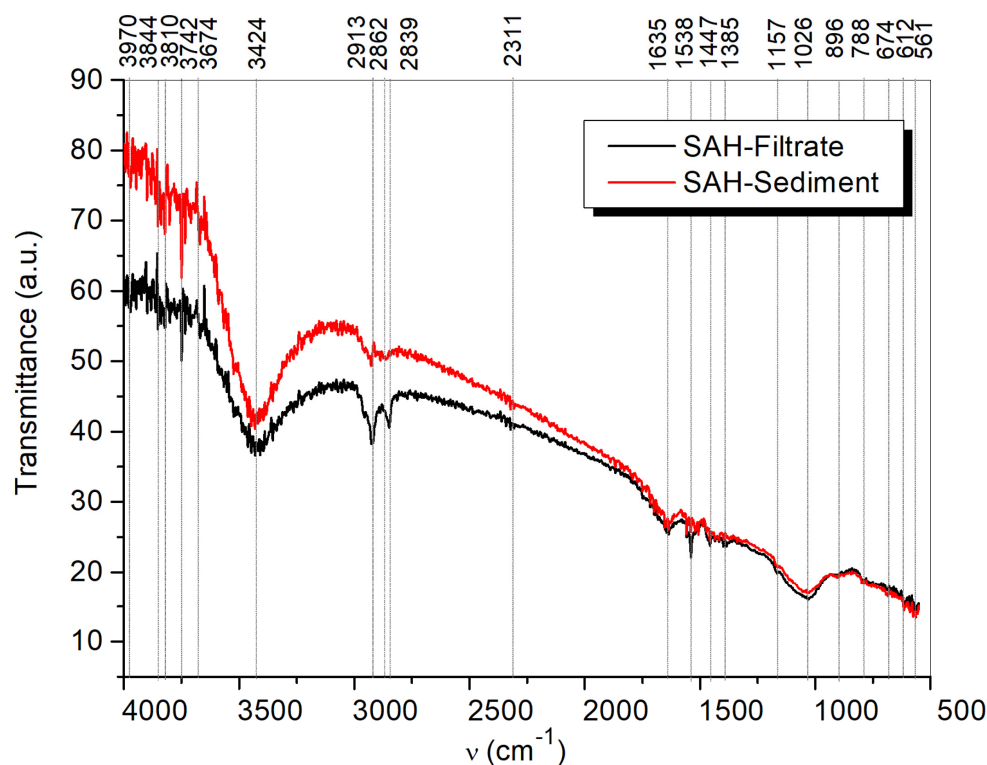


Figure 11. FTIR spectra of decanted solid phase from rainwater after Saharan dust episode.

Vibration modes denote functional groups that, in conjunction with EDS elementary chemical analysis, provide information about the chemical structures in the solid sediment and filtrate material resulting from the rainwater in Pelinia during the Saharan dust episode.

The band at 3970 cm^{-1} is assigned to alkali (mainly Na, K bond to OH groups).

Sodium aluminate in its different forms (NaAlO_2 , $\text{Na}_2\text{O}\cdot\text{Al}_2\text{O}_3$, or $\text{Na}_2\text{Al}_2\text{O}_4$ or alkali form in solution, $\text{NaAl}(\text{OH})_4$) is denoted by the bands at 3844 cm^{-1} (Al-OH stretching) [25,29,30] and 612 cm^{-1} (Al-O stretching) [25,29–31]. Magnesium oxide and hydroxide are evidenced by the bands at 3674 cm^{-1} (OH stretching in Mg-OH or Mg-O) [29,30,32,33], 2862 cm^{-1} [29,30,32,33], 2839 and 2311 cm^{-1} (Mg-O) [29,30,32,33], and 1447 , 1026 , 896 , and 561 cm^{-1} , which are also in the Mg $(\text{OH})_2$ spectrum [29,30,32,33]. The presence of these components is also confirmed by the rainwater's pH value of 8. In addition, carbonates, such as calcium carbonate and sodium or potassium carbonate, are evidenced by the vibrations exhibited in the sediment and filtrate spectra (Figure 11) at 1447 cm^{-1} for carbonate ions, CO_3^{2-} [29–31]; this peak also denotes a crystalline structure [34–36]. The band at 3810 cm^{-1} is assigned to OH free stretching due to adsorbed water, together with the band at 1635 cm^{-1} . Silanol groups bonded to terminal polymeric chains are denoted by the 3742 cm^{-1} band assigned to Si-OH stretching [25,31,36] and 2311 cm^{-1} assigned to Si-H stretching [36–38].

The band at 2311 cm^{-1} also indicates $\text{O}=\text{C}=\text{O}$ stretching in carbon dioxide (CO_2). Vibration modes of sulfates (FeSO_4 ; $\text{Al}_2(\text{SO}_4)_3$, $\text{CaSO}_4\cdot\text{H}_2\text{O}$) and ferrocyanides [30] overlap on the band at 1635 cm^{-1} . The two spectra in Figure 11 also exhibit organic components such as amines, amides, polypeptides, imines, oximes, pyrroles, aldehydes, sulfoxides, sulfones, and nitro compounds in a mixture that reflects the interaction of the Saharan dust with the organic matter in the atmosphere, including vegetal material such as pollen particles, or other biological material. In this sense, the vibration modes for the organic functional groups in the spectra of Figure 11 are described as follows:

The band at 3424 cm^{-1} is assigned to N-H stretching in aliphatic primary amines, and in heteroaromatic compounds such as pyrrole, pyridine, imines, and oximes [25,26,29–31,39–41], which may result from the degradation of leaves under sunlight interaction and/or enzy-

matic action. The vibration mode at 2913 cm^{-1} is for N–H stretching in amine salt, in the same range with aliphatic C–H stretching [25,30–40]. Aliphatic C–H stretching is also denoted by a band at 2862 cm^{-1} [29,30] while 2839 cm^{-1} is assigned to C–H stretching in aldehydes [30]. The band at 1635 cm^{-1} is for C=N stretching in imines and oxime [25,26,29–32], in the same range with C=O stretching in tertiary amide [25,29–31], as well as with C=C stretching in disubstituted (cis) alkenes and/or conjugated alkenes [25,29,32] and N–H bending amine [25,29–32,39]. The vibration mode of N–O stretching in nitro groups [25,29–32] denoted by the band at 1538 cm^{-1} is in the same range as skeletal vibrations of aromatic bonds C=C [25,30,31], amides II NHC=O free and H-bonded, and polypeptides [25,30]. The band at 1447 cm^{-1} is assigned to C–H bending in the alkane methyl group and the band at 1385 cm^{-1} is assigned to a number of functional groups with vibration modes in the same range: O–H bending in phenol [25,29,30], C–H bending in aldehyde [25,30], C–H bending in alkane gemdimethyl [25,31], S=O stretching in sulfates [25,30], and sulfonyl chloride [25,30]. The band at 1157 cm^{-1} of C–O stretching in secondary alcohol [25,30] is followed by the 1026 cm^{-1} band of C–N stretching in amine [25,30], and the 896 cm^{-1} band is assigned to C=C bending in alkene vinylidene [25,30] and to S–O stretching in sulfoxides and sulfones [25,30]. The 788 cm^{-1} band is for C=C bending in alkene trisubstituted [25,30], in the same range as the S–O stretching in sulfoxides and sulfones [25,30]. The vibration at 674 cm^{-1} denotes C–Cl halogen compounds [30] and the vibration at 561 cm^{-1} denotes C–S stretching in sulfones and sulfoxides [25,30].

Saharan dust is a provider of nutrients for plants and marine phytoplankton. The bioavailability of dust-derived iron requires the soluble form of iron. This implies a complexity of factors that contribute to the promotion of photochemical transformations resulting in the production of the soluble form of iron [12–17]. Acidic pH and increased temperature favor this process; also, the smaller the particles, the higher the solubility, by increasing the total contact surface with water.

Heteroatoms such as oxygen (O), sulfur (S), and nitrogen (N) can increase an organic molecule's iron-binding capacity; the presence of organic matter containing O and/or S and/or N can increase the solubility of iron contained in aerosols [40]. Carboxyl groups in particular have been noted for increasing the ligand-like activity of organic matter in aerosols [40,41]. Other functional groups known to contribute aerosols containing more of these ligands have higher percentages of soluble iron than aerosols that have fewer or no ligands. Saharan dust aerosols contain smaller amounts of these ligands, which contributes to the low solubility of iron from Saharan dust [40,41]. Aerosol organic matter of Saharan dust tends to contain more carbohydrate-like material, which generally does not have strong ligand activity [41]. Thus, based on the spectroscopic analyses performed, there are indications that the episode of Saharan dust that affected Pelinia would not have been beneficial for agriculture. This is supported by the alkaline nature of the analyzed rainwater (pH 8), which contradicts the hypothesis that only acidity would have contributed to the solubilization of iron ions. On the other hand, according to studies that show that other functional groups including ethers, esters, and amines can contribute to ligand-like properties in aerosols [40,41] and based on the fact that such groups are found in the spectral analysis of Saharan dust separated from the rainwater sample of Pelinia, there may have been some beneficial influence on agriculture.

On the other hand, organic sulfur compounds such as sulfoxides, sulfones, and sulfides, known as irritants, may induce negative effects on human health. The neurotoxic effect induced by the presence of imines cannot be neglected either [42,43]. Another aspect is allergens such as pollen transported with Saharan dust particles. Pollen bags were highlighted by electron microscopy performed on the sediment collected from rainwater related to the Saharan dust episode of 12 May 2020, in Pelinia (Figure 12).



Figure 12. Pollen bags detected in sediment of rainwater collected on 12 May 2020, in Pelinia.

4. Conclusions

Based on meteorological parameters, data from solar photometry measurements provided by the AERONET network, and trajectory calculation using the HYSPLIT model, the influence of the Saharan dust storm of 12 May 2020, in the northern part of the Republic of Moldova, above Pelinia village in Dochia district, could be confirmed. Precipitation brought the chemical components of the aerosols to the ground, and they could be identified in sediments and filtered material from rainwater samples. This investigation showed how the lack of AOD data could be remedied by using extrapolation methods with simulation and spectroscopic analysis of the chemical composition of particles in the atmosphere. At the same time, these analyses highlight the importance of monitoring not only the trajectory of Saharan dust, but also its chemical composition, because irritating chemical compounds and possible bioallergens in the form of pollen bags were detected in sediment from the rainwater collected in Pelinia. These analyses show the role of Saharan dust as a “carrier” of various substances and biocomponents, with potential impacts on agriculture [44], humans [11], animals, and plants.

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