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Assessment of Inorganic Chemical Composition of PM_{2.5} Aerosols in Tiaret-city, Algeria

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ABSTRACT

The objective of this work is to determine and quantify the inorganic compounds of PM_{2.5} aerosols; collected using a two-stage Dekati® PM10 impactor; in educational establishments in Tiaret-city "Algeria". It also aimed to follow the dispersion of these particles in sites near the forest, urban fabric and urban sites exposed to dense road traffic, as well as to determine the source of this type of pollution. 23 primary schools were chosen as sampling sites in Tiaret city. The collection of PM_{2.5} was carried out in the winter and summer period. The spectroscopic analysis carried out by FTIR-ATR shows that the sulfate ions recorded their maximum in the summer period than in the winter, as well as the nitrate and ammonium ions which do not show a large seasonal difference. The dominant species in urban areas are NO₃⁻, SO₄⁻² which due to the presence of a very important source of NOx and SO₂ which, by gas-particle conversion, transform into nitrate and sulfate ions respectively. These species present a lower proportion in sites close to the forest. However, the same order of abundance of these ions exists in the two sampling periods. The main sources of NOx in urban sites are combustion phenomena linked in particular to the transport system, which is characterized by very dense traffic in the city's agglomeration.

Keywords: Air pollution; PM_{2.5} Aerosols; Road traffic; FTIR-ATR spectroscopy; Schools; Tiaret city.

1. INTRODUCTION

In recent decades, air quality has given rise to serious concerns in the face of rapid economic and industrial development in many countries around the world. Currently, suspended particles (PM) are universally recognized as the main pollutant in the atmosphere [1].

In urban areas, PM mainly comes from human activities. The automotive transport sector is one of the main responsible for their emission into the air, the latter contributing to the respective emission of 13% of PM_{2.5}, with particles of aerodynamic diameter less than 2.5 μm [2]. Numerous studies on the toxicological effects of suspended particles have reported that exposure to fine particles (PM_{2.5}) constitutes a risk factor for human health.

Our work consisted in determining the inorganic chemical composition of PM_{2.5}, by estimating the different sources of emission of these particles in Tiaret city, to provide local authorities with better knowledge of the sources on which we can act to decrease particle levels.

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2. MATERIALS AND METHODS

2.1 Study Zone

The study was carried out in Tiaret city. It is located in the North-West of Algeria, between the Tellian chain in the North and the Atlassian chain in the South, at an altitude of 980m on average (Fig. 1). The climate is semi-arid Mediterranean with an average annual rainfall of 400 mm / year. The predominant winds come from the West and the North-West, their average speeds vary from 3 to 4 m/s.

This city had more than 200 km of the urban road network, its vehicle fleet are made up of 156,952 vehicles of all types, and 71.33% of gasoline cars, and 26% diesel. This park is very heterogeneous (private or a utility vehicle, gasoline or diesel, recent or old, etc.). In total, new cars (0 to 5 years old) represent 12% of the park. On the other hand, cars over 10 years old represent 75%. However, older vehicles are the most polluting [3].

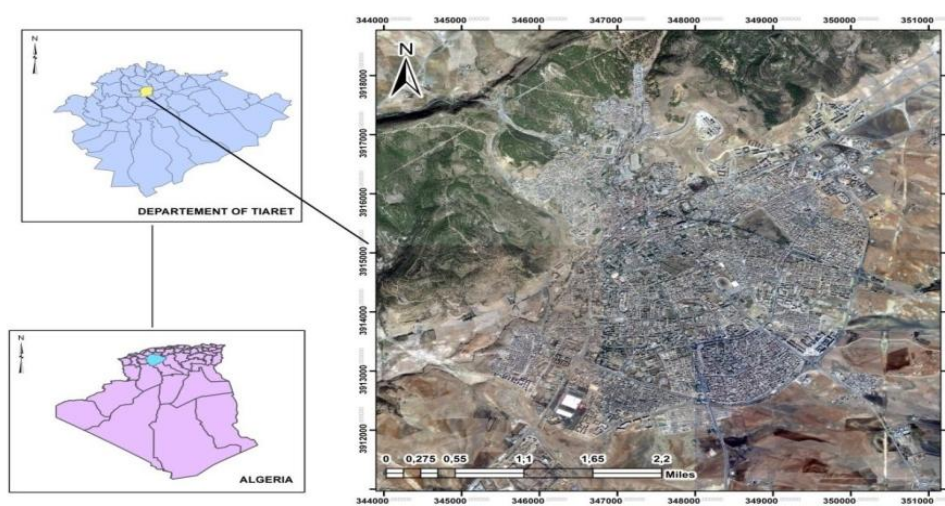


Fig. 1. Location of the study area [4]

2.2 Selection of Sites

The purpose of this study is to collect $PM_{2.5}$ in primary schools in the city of Tiaret by a two-stage Dekati® PM_{10} impactor (ISO23210, www.dekati.com, 2017), and to determine its chemical inorganic composition. Overall, 23 primary schools were chosen in these three axes (sites located near the forest (green circle), urban sites (yellow circle) and urban sites exposed to dense road traffic (red circle)) (Fig. 2).

Collecting the $PM_{2.5}$ involves placing the Dekati impactor in the middle of the schoolyard with one meter high, in order to remove any source of contamination from the ground. The collection time is 30 minutes, with six samples/ a day/ a school. A total of 276 samples were collected, during eight months of sampling.

2.3 FTIR-ATR Spectroscopy Analysis

The particles sampled are detected by back scattered an electron image analysis (chemical contrast) after a calibration of the microscope. The Cartesian coordinates of each detected particle are stored and an X microanalysis is performed automatically for each particle. The counting time per a particle is of the order of 2 to 4s. The elements (Ammonium NH_4^+ , Phosphates PO_4^{3-} , and Nitrate NO_3^-) were also analyzed by UV-visible spectroscopy.



Fig. 2. Location of sampling sites (Google Earth, 2017)

3. RESULTS AND DISCUSSION

3.1 Spectroscopic Analysis Results

The results found by this analysis are illustrated in Fig. 3 which presents the FTIR-ATR spectra, of different samples taken

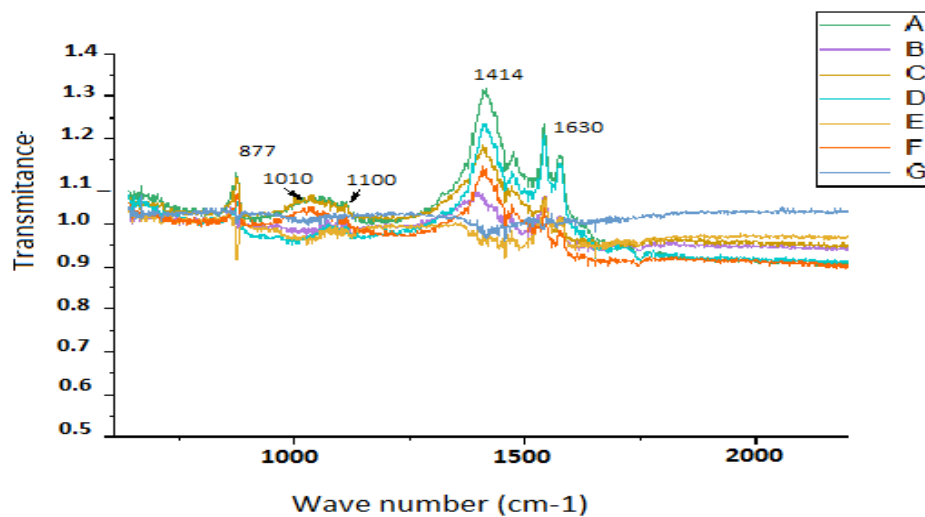


Fig. 3. Sample spectra of PM2.5 analyzed by FTiR spectroscopy

Légende:

A: Spectrum of sample taken from school N ° 04 (class 3), B: Spectrum of sample taken from school N ° 18 (class 1)

C: Spectrum of sample taken in school N ° 13 (class 2), D: Spectrum of sample taken in school N ° 01 (class 3)

E: Spectrum of sample taken in school N ° 20 (class 1), F: Spectrum of sample taken in school N ° 23 (class 2)

G: Spectrum of sample taken from school No. 20 (class 1)

The peaks had found in the spectra of samples taken from sites exposed to heavy traffic and urban samples are similar to each other. This suggests that the structural and functional groups of PM2.5

samples show many similarities. The figure shows that ammonium and these derivatives show maximum concentrations during sampling at 1414cm^{-1} in the different samples. Most of the PM_{2.5} samples peak at 615cm^{-1} , which exhibits the element of SO_4^{2-} , revealing that the sulfate ion is an important component of the collected PM_{2.5} particles. The peak reported at 1450cm^{-1} , which characterizes solid samples, can be attributed to functional organic groups such as aliphatic carbon (CH-3). This finding shows a great consistency in the functional properties of PM_{2.5}. However, most cations and anions are secondary pollutants that formed in the atmosphere from primary pollutants. The conversion of secondary pollutants depends on the concentrations of the primary pollutants (precursors) and meteorological parameters, such as a temperature and a solar radiation. Therefore, the positive association probably shows that the meteorological parameters affect both ions (NO_3 and SO_2). Nitrates and ammonium show different sulfate profiles during the secondary pollution process, the formation of nitrates is more intense than that of sulfates shows a significant improvement during the rapidly forming a pollution episode [2].

We have observed the presence of SO_4^{2-} , NO_3^- , SiO_4^{2-} , CO_3^{2-} and NH_4^+ cations with higher concentrations in the collected particles. This also showed that dust storms are characterized by an enrichment in a SiO_4^{2-} and a CaCO_3 .

3.1.1 Winter period

The most abundant anions are sulfates, nitrates at 825 , 1356 and 1410cm^{-1} (NO_3^-) and chlorides. The most abundant cations are ammonium and these derivatives at 1414cm^{-1} (NH_4^+ , $(\text{NH}_4)_2\text{SO}_4$) and sulfate [5]. These ions mainly come from the automotive source. Most often, these compounds are very soluble in water (in the case of sulfate), and promote the capture of atmospheric water and therefore the formation of clouds. There are also insoluble or less soluble elements such as aluminosilicates, and transition metals such as iron, mainly from the terrigenous source (Fig.4)

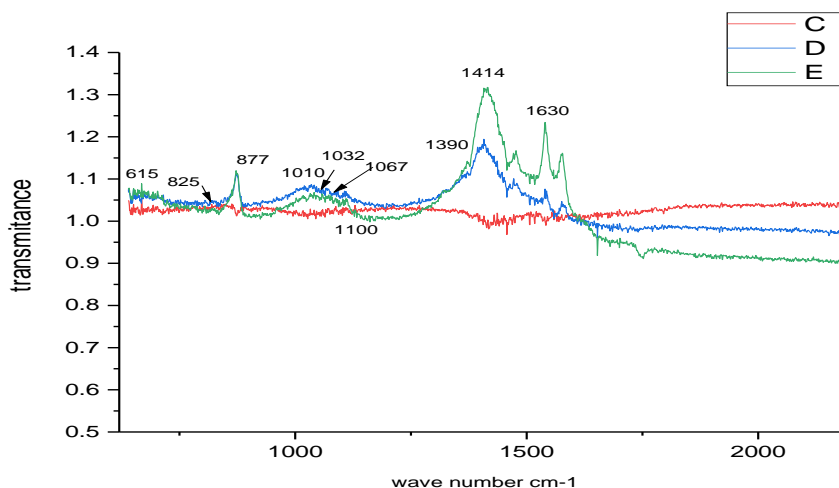


Fig. 4. Sample spectra of PM_{2.5} analyzed by FTIR spectroscopy during the winter period

Légende:

C: Spectrum of sample taken from school No. 20 (class 1)

D: Spectrum of sample taken from school N ° 12 (class 2)

E: Spectrum of sample taken from school N ° 2 (class 3)

The peak recorded at 1010 , 1114cm^{-1} is probably related to the Si-O group [6]. Thus, OH radicals can oxidize SO_2 and NO_2 , respectively into sulfuric acid (H_2SO_4) and nitric acid (HNO_3) [7]. These ions found recorded high values in the 3rd class which group together urban sites heavily polluted by road traffic than the other classes.

Sulfates are often associated with NH_4^+ cations (80%), as the ionic balance is in a favor of anions, there is a deficit of a positive charge, compensated by the contribution of H^+ ions. This phenomenon explains why the aerosol and therefore the rain or snow has formed around the aerosol after water vapor capture always has an acidic pH. The pH is often around 5. It can drop to 4 for certain types of snow.

3.1.2 Summertime

The notable peak at 1630cm^{-1} is attributed to the aromatic bond $\text{C} = \text{C}$. We record a vibration of a large peak at 1390cm^{-1} , which cannot be discerned in any of the potential source spectra (Fig. 5). It has been ignored by some studies, and by others [8]. It is attributed to compounds of NH_4NO_3 . It is on behalf of the secondary particle which could be produced in photochemical processes, and exhibits a high concentration of secondary particles. Indeed, these results led to the conclusion that the secondary aerosol concentrations are very high during fog events in winter. However, the intensity of this peak is very different from that of the summer $\text{PM}_{2.5}$ samples. Additionally, all $\text{PM}_{2.5}$ samples have a broad peak at 1080cm^{-1} . Some authors have linked this peak to vibrations only from the $\text{C}-\text{O}$ bond or 60 polysaccharides, but others believe that there is a strong overlap with the sulfate ion peak at 1090cm^{-1} .

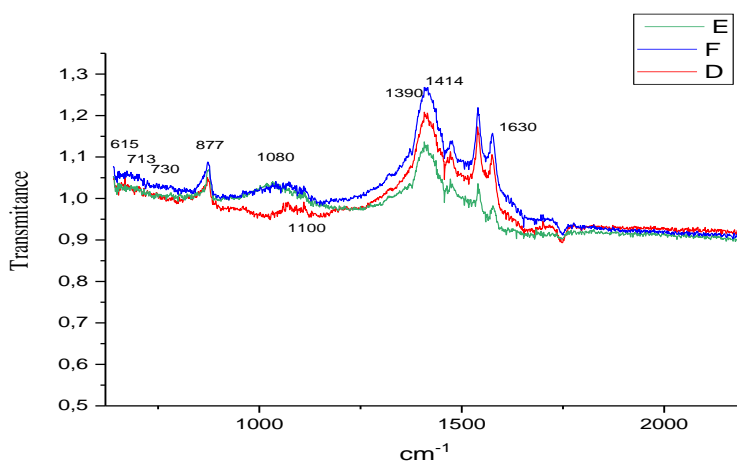


Fig. 5. Samples of $\text{PM}_{2.5}$ analyzed by FTiR-ATR during the summer period

Légende:

E: Spectrum of sample taken from school No. 20 (class 1);

D: Spectrum of sample taken from School No. 15 (class 2);

F: Spectrum of sample taken in school No. 2 (class 3).

In the $\text{PM}_{2.5}$ samples analyzed, we observed typical vibration frequencies of the ions at 1414cm^{-1} (NH_4^+); 825 and 1356cm^{-1} (NO_3^-); and at 713 , 730 and 877cm^{-1} (CO_3^{2-}).

The presence of the compounds of $(\text{NH}_4)_2\text{SO}_4$ and NH_4NO_3 is confirmed by the spectra also illustrated in Fig.5. Another peak was recorded at 615 and 1100cm^{-1} (SO_4^{2-} group) [8]. The peak of 1100cm^{-1} being attributed to the asymmetric stretching vibration of the sulfate ion. Ammonium, displayed a maximum concentration during the sampling days of this period, due to the high temperature and dry soil in the schoolyard. The main known reactions are photochemical oxidation by OH° and NO° radicals which lead to Nitro HAP3 [9]. These compounds are even more carcinogenic than the parent PAHs, but the concentrations generally do not exceed $0.1\text{ng}/\text{m}^3$, except during strong episodes of photochemical pollution.

3.1.3 Seasonal variations

Seasonal variations in cation and anion concentrations show that the nitrate concentration was higher during summer sampling and halved during winter. Due to the low rate of photochemical oxidation of organic material to produce OH° ; a radical responsible for the formation of HNO_3 via (R12) [10]. The ion concentrations of the sulfates followed a pattern similar to that of the ions of the nitrates. The photochemical oxidation of SO_2 becomes active in summer (solar radiation and high temperatures) to produce SO_2-4 [11]. The concentration of ammonium ions which had the highest percentage in these fine particles showed relatively higher levels in summer. The variation of these ions is well correlated with the sulfate ions which recorded an increase in summer, due to the improvement in the rate of photo oxidation of SO_2 and components of the earth's crust SiO_2 (1010 , 1032cm^{-1}) [12]. These high

values could be explained by the widely developed use of sulfur fuel (diesel) in buses and trucks, the combustion of which leads to the formation of carbonaceous soot rich in sulfur, especially in the environment highly polluted urban area [13].

The seasonal cycles of Ammonium in fine particles PM_{2.5}, followed a pattern similar to that observed for the ions of the Sulfates with low rates in winter but probably strong in summer, which represents a favorable environment for the evaporation of this one [14]. This is because many absorption bands are likely to overlap. The nitrates (NO₃⁻), sulfates (SO₄⁻²) and ammonium (NH₄⁺) ions which are very abundant in the aerosol produce very wide and intense absorption bands liable to interfere with the measurements of the desired chemical functions [1,15]. Nitrogen aerosols are reported to play an important role in deteriorating the air quality.

3.2 Determination of Ammonium, Phosphates and Nitrates

To identify the sources of PM_{2.5} emissions, we have analyzed ammonium NH₄⁺, phosphates PO₄⁻³ and the nitrate NO₃⁻ by UV-Visible spectroscopy. The automatic classification of the mass concentrations found, appears 03 pollution classes.

3.2.1 Pollution classes

Table 1. Descriptive statistics of pollution classes by inorganic compounds (Ammonium, Nitrate and Phosphate)

Compounds (ug/m ³) ×10 ⁻⁵	Pollution class	E	N	Moy	Min	Max	Variance	Standard deviation
Ammonium (NH ₄)	1	34	2	0,24	0,23	0,25	0,0002	0,014
Ammonium (NH ₄)	2	20	2	0,31	0,30	0,4	0,0032	0,057
Ammonium (NH ₄)	3	18	2	0,35	0,31	0,39	0,0032	0,057
Nitrate (NO ₃ ⁻)	1	34	2	0,23	0,22	0,24	0,0002	0,014
Nitrate (NO ₃ ⁻)	2	20	2	0,4	0,36	0,44	0,0032	0,057
Nitrate (NO ₃ ⁻)	3	18	2	0,81	0,66	0,95	0,0421	0,205
Phosphate (PO ₄)	1	34	2	0,022	0,018	0,027	0,0000	0,006
Phosphate (PO ₄)	2	20	2	0,038	0,028	0,049	0,0002	0,015
Phosphate (PO ₄)	3	18	2	0,067	0,054	0,080	0,0003	0,019

E: number of samples involved in the class.

N: winter and summer period.

Table 1 clearly shows that the high concentrations of inorganic compounds were recorded in the third class with a maximum of 0.35 ± 0.057 ug / m³ × 10⁻⁵, 0.81 ± 0.205 ug / m³ × 10⁻⁵ and 0.067 ± 0.019 ug / m³ × 10⁻⁵ in Ammonium, Nitrates and Phosphates respectively. These values are excessively higher than those for class 1, which includes sites close to the drill. The second class characterizes urban sites.

Nitrates have the highest concentrations in all samples analyzed compared to phosphates which are slightly weak and superior to ammonium

These results are somewhat lower than the concentrations previously reported in the range of other studies [16-17] on the analysis of fine particles that are distributed from the atmospheric source to urban sites.

3.2.2 Evolution of ions (Nitrates, Ammonium and Phosphates) during sampling

We have performed an analysis of the variance between the analyzed ions and the sampling time, the result is shown in Fig. 6.

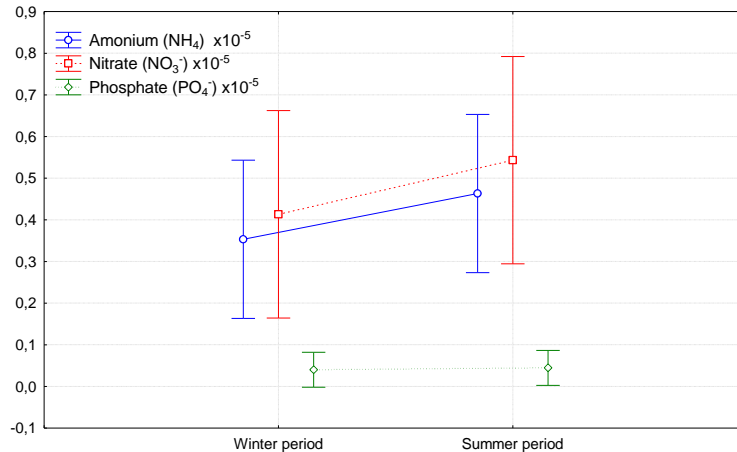


Fig. 6. Evolution of ions (Nitrate, Ammonium and Phosphate) during the sampling period

The examination of the Fig. 6 shows that the NO₃ series had relatively lower winter levels, while NH₄⁺ levels were slightly higher in summer-fall. These inverse seasonal trends are probably related to the summer oxidation conditions and the low thermal stability of NH₄NO₃ during the warm period. In addition, N levels can also explain the high formation of nitrates in the winter period. In addition, NH₄⁺ does not show a definite seasonal trend. Then, peaks of these elements occurred throughout the year and were strongly correlated with the appearance of Sahara dust reported during sampling.

NH₄⁺ concentrations have been found to establish the charge compensation, or the neutralization, with NO₃⁻ and SO₄⁻² ions. This phenomenon could be partly explained on one hand by the relatively high emission rates of ammonium precursors in the sampling region, and on the other hand due to the instability of the ammonium salts in a particulate form. In relatively hot atmospheres, as was encountered during sampling campaigns. In this case, the neutralization of the NO₃⁻ and SO₄⁻² species could be explained by their association with NH₄⁺, without excluding other transition metal ions [18]. The combination of these factors could explain the higher sulfate production and the volatilization of nitrate species in summer.

We find that fine, non-light absorbing inorganic aerosols such as sulfate can have a warming effect. Specifically, although the opacity of fine particles decreases at longer wavelengths, they can strongly absorb and re-emit the thermal radiation under resonant conditions at long wavelengths [19].

The higher levels of NO₃⁻ and SO₄⁻² in the urban site could be due to the presence of a very important source of NO_x and SO₂ which, by gas-particle conversion, are transformed into nitrate and sulfate ions respectively. The main sources of NO_x in the urban site are combustion phenomena, in particular by the transport system, which is characterized by very dense traffic in the city agglomeration. As for SO₂, the frequent use of sulfur fuels in heavy vehicles (buses and trucks) is the main source of SO₂ in the urban site [20-21].

According to [22] Spring dust storms had a strong impact on the pm_{2.5} Overall, organic carbon was the most abundant species, accounting for at least 30% of the total mass of PM_{2.5}. In these fine particles the ions: Ca⁺², SO₄⁻², NO₃⁻ are emitted by vehicles [23,8] NH₄⁺, K, by the diesel exhaust from buses. The presence of these elements with the NH₃ therefore indicates that these particles are secondary aerosols. the nitrate is derived from gas-to-particle conversion processes by NO_x oxidation products, which are mainly emitted in the vehicle exhaust gases.

3.2.3 Correlation of PM 2.5 concentration and elements

The examination of Fig. 7 shows that there is a highly significant difference between the three pollution classes ($p = 0.00$), with a dominance of PM_{2.5}, recorded in the 3rd class, which represents the schools located in the main streets of the city center, and exposed to heavy road traffic, coinciding with increased human activities in the city.

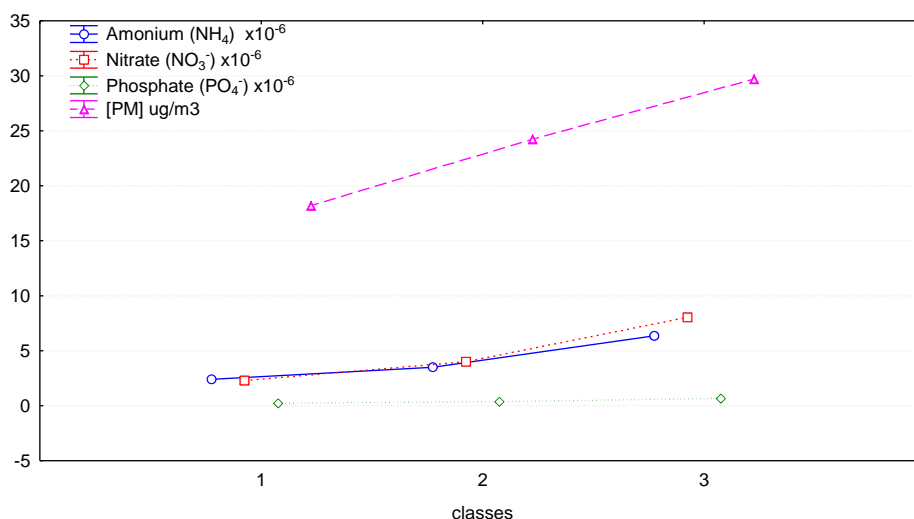


Fig. 7. Correlation analyzes between ammonium, nitrate, phosphate concentrations and mass concentrations of PM_{2.5}

The analysis in fig.7 also shows that there is a positive correlation between the mass concentrations of PM_{2.5} and the inorganic elements analyzed (ammonium, nitrate and phosphate).

3.3 Discussion

In general, this study shows that non-destructive methods are effective in tracing the sources of PM_{2.5}. They can be further improved and have a promising future in real-time PM_{2.5} monitoring. Thus, the generally anthropogenic spherical particles and those analyzed in this study exhibit a smooth surface and most of them are ammonium and nitrate which could come from the combustion of road traffic material [24]. On the other hand, given the fact that the particles emitted by road traffic can travel long distances up to 30 km before being deposited [25] these spherical particles could well be emitted in the main axes traffic in the city.

Sulfate and nitrate ions are the result of side reactions of aerosols, crustal dust with HNO₃ and H₂SO₄. Sulfate is the result of high levels of SO₂ from long distance transport; in winter [26, 27] corroborated by air mass during the same sampling days and by local exhaust emissions, thus diesel-powered buses run close to of sampling sites. Regarding the source of particulate nitrates, probably related to vehicle emissions. However, ammonia derived from vehicle emissions has recently attracted much attention [28-29]. The urban site is influenced by many direct and indirect sources of particulate emissions fines, such as the combustion of fuels which can be added to the resuspension of "soil dust" by the circulation of vehicles on the roads. All these factors can promote the formation of fine particles and could explain their higher concentrations in urban sites exposed to traffic compared to other sites.

The dominant species in urban areas are NO₃⁻, SO₄⁻². They have a lower proportion (fewer local sources of NO₃⁻ and SO₄⁻² precursors) in sites close to forests. However, the same order of abundance of these ions exists in the two periods.

According to [22], Spring dust storms had a strong impact on PM_{2.5} overall, organic carbon was the most abundant species, accounting for at least 30% of the total mass of PM_{2.5}, surtput in urban sites. In these fine particles, the ions: Ca₂⁺, SO₄⁻², NO₃⁻ are emitted by vehicles [30,22] [31] NH₄⁺, K, by diesel exhaust from buses. The presence of these elements with NH₃ therefore indicates that these particles are secondary aerosols. Nitrate derives from gas-to-particle conversion processes by NOx oxidation products, which are mainly emitted in vehicle exhaust gases [31].

4. SUSTAINABLE PLANNING AND INSTITUTIONAL DEVELOPMENT FOR COMBATING AIR POLLUTION

In terms of urban atmospheric pollution, that generated by automobile traffic in toxic emissions of gases and particles, as well as the formation of carbon monoxide, partially oxidized compounds and sulfur or sulfur dioxide (SO₂ and SO₃), which disperse, under the effect of the wind, in all directions and manifest itself as a difficulty in breathing, tingling of the throat and eyes and various irritations, high enough to cause adverse health effects, in susceptible individuals (children, elderly or sick).

The public authorities must, more particularly, ensure the adaptation and the strict application of the regulations in force. The fight to reduce toxic emissions in urban areas must also be the subject of the same rigor, given its effects on public health

4.1 Nature Based Solution towards Sustainable Planning in Algeria

The Algerian state must select alert thresholds for the danger of pollution to improve the air quality as well as atmospheric monitoring devices to assess the real degree of particulate pollution and implement a policy to protect human health. Pollution peaks and periods when thresholds are exceeded are detected and forecasts are made to plan and implement the measures necessary to improve air quality. Despite the difficulty of establishing environmental and energy strategies; Algeria, devotes specific provisions to the fight against the air pollution, by Law No. 43 of July 20, 2003, relating to the protection of the environment within the framework of sustainable development (www.Joradp.dz)

4.2 Management and Sustainability Approaches to Combat PM_{2.5} Aerosols in Tiaret, Algeria

Numerous scientific works have carried out by Algerian researchers confirm the need to reduce polluting emissions from road traffic by renewing the vehicle fleet, and improving the regulation of the engine combustion through the use of less polluting fuels.

The programs of the conservation of the vegetation cover and the planting of trees remain quite rational and reasonable, if one wants to rapidly increases the forest cover not only in the cities, but also in these schools, by favoring programs of the plantation of various types. This can include removing competing the vegetation, reducing the runoff, improving soil conditions, suppressing fires, and preparing the soil. However, it is recommended to:

- Rebuild forests and encourage the tree planting in new neighborhoods.
- Maintenance of the biological diversity and ecological balance;
- The use of bicycles instead of vehicles, minimizing the emission of the PM_{2.5}

4.3 Future Perspective

This study is a first initiative to quantify and assess PM_{2.5} pollution in the Tiaret-city and give us a preliminary idea of the composition and sources of this pollution.

To manage the factors of the degradation of public health, it is recommended to carry out an epidemiological study to see the prevalence between the human exposure to fine particles and respiratory diseases.

- Enhancement of the city's air by creating the air quality monitoring stations, and ensuring citizen awareness campaigns

In our future work, we will aim to take the advantage of the easy and rapid nature of spectroscopic methods to chemically characterize the formation of primary and secondary aerosols, emitted by divergent sources such as the combustion of the biomass and vehicles during different episodes of pollution.

5. CONCLUSION

This study shows that non-destructive methods are effective in tracing the sources of PM_{2.5}. They can still be improved and have a bright future in real-time PM_{2.5} monitoring.

The inorganic analysis had carried out by the FTIR-ATR spectroscopy, shows that the sulfate ions recorded their maximum in the summer period than in the winter, as well as the nitrate and ammonium ions which do not show a great seasonal deference.

The dominant species in urban areas are NO₃⁻, SO₄⁻²; However, the same order of abundance of these ions exists in the two sampling periods. The main source of NO_x in the urban site is the phenomenon of the combustion linked to the transport which characterizes very dense traffic in the metropolitan area of the city.

These sites have been influenced by direct and indirect sources, such as the fuel combustion, which can add to the re-suspension of "soil dust" from vehicle traffic on the roads. All these factors promote the formation of fine particles and explain their abundance in urban sites exposed to the trafficking compared to sites close to forests.

The presence of SiO₄⁻² and CaCO₃ ions, in the sites which are in low places between the slopes, also showed that the dust storms are characterized by an enrichment in these elements, especially in winter season which has been characterized by the dominance of the sand wind by contribution to the summer season.

In conclusion, PM show significant variations in their chemical composition, linked in particular to the proximity of sources and to temporal variability (daily or seasonal), leading to a poor understanding of their effects on health. Indeed, the precise composition of the particulate aerosol, in particular its chemical composition, its evolution over time but also its concentration in the air remains difficult to understand.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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