# ГІДРОГЕОЛОГІЯ, ІНЖЕНЕРНА ТА ЕКОЛОГІЧНА ГЕОЛОГІЯ

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**Z. Mekti[1], Dr in Mining Engineering,**
**E-mail: mektizohir@yahoo.fr;**
**A. Boutemedjet[1], Dr in Mining Engineering,**
**E-mail: boutemedjetassia@yahoo.fr;**
**O. Sekiou[2], Dr in Immunology,**
**E-mail: o.sekiou@cre.dz;**
**S. Berdoudi[1], Dr in Mining Engineering,**
**E-mail: berdoudisaid@yahoo.fr;**
**A. Chaib[1], Dr in Mining Engineering,**
**E-mail: achaib_as@yahoo.fr;**
**F. Kouider[3], MSc in Chemical Engineering,**
**E-mail: kouiderfareskul016@gmail.com;**
**A. Rahmani[3], MSc in Chemical Engineering,**
**E-mail: Abdun81@gmail.com;**
**A. Benselhoub[2], PhD in Ecology and Environment Protection,**
**E-mail: benselhoub@yahoo.fr;**
**[1]Laboratory of Valorization of Mining Resources and Environment, Mining Department, Badji Mokhtar University, Annaba, Algeria;**
**[2]Environmental Research Center (C.R.E), Annaba, Algeria;**
**[3]Faculty of Engineering, Built Environment & IT, SEGi University, Malaysia**

## AIRBORNE PARTICULATE MATTER POLLUTION GENERATED BY PHOSPHATE EXPORT AT THE PORT OF ANNABA

***(Представлено членом редакційної колегії д-ром геол. наук О.І. Меньшовим)***

***For several decades, airborne particulate matter (PM) pollution has become one of the major concerns of policymakers across the globe, due to its adverse effects on climate, human health, environment and various socio-economic sectors. The process of phosphate export in bulk is one of the main source generating air pollutants, such as PM emissions of different shapes, concentrations and particle sizes, which can cause respiratory diseases. The objective of this work is to determine the dust level generated by the processes of unloading, storage and loading of phosphate in bulk to ships, by the company of the port installation at the port of Annaba (East of Algeria).***

***The PM measurement campaign was carried out according to the NF X 43-014. It was performed on 08 sampling points located at the port of Annaba and in a periphery with a radius of up to 2.3 km. The recorded results show very high levels of dust in 05 samples series largely exceeding the acceptable threshold (1000 mg/m²/day). The level of heavy metals in dust was evaluated by the analysis of Atomic Absorption Spectrometry. EDAX analysis showed that the phosphate PM have an angular morphology elongated particles and hook-shaped.***

***Keywords: Health risk, Heavy metals, Environment, Impacts, Northeast of Algeria.***

**Introduction.** Natural phosphates comprise more than 200 mineralogical species (*Fisher, Jerome, 1973*). However, the most abundant ones belong to the apatite family (*Raguin, 1961*). Phosphate ores are defined according to their $P_2O_5$ content or in terms of the percentage of tricalcium phosphate (TPL or BPL: Bone Phosphate Lime) which is really the basic phosphate product (1 TPL = 2.185 $P_2O_5$). It also presents a very varied field of use on an industrial scale, of which 80 to 90% of the production is used in fertilizers and in the manufacture of phosphoric acid (*Dumon, 1980*). In Algeria and in the Djebel-Onk region (Tebessa), the phosphate ore, is exploited by the phosphate mining company (SOMIPHOS), a subsidiary of the national company "FERPHOS Group" headquartered in Annaba. In 2018, the Somiphos Company exported 1.8 million tons of phosphate; it aims in the following years to produce and export 5 million tons.

The ore mined and processed in the Djebel-Onk factory is then shipped to the Annaba Port installation (IPA), either by rail (320 km) or by road transport. The entire production is exported as raw material to several countries around the world (*Office DED, 2019*). However, the production process of Phosphate generates environmental impacts such as gas emissions ($CO_2$, $NO_x$, $SO_2$), especially PM emissions at all levels of production, mainly in the port of Annaba, where the process is underway to export phosphate *(Benselhoub et al., 2020)*. PM generated by the phosphate export operation (Unloading, Storage, Loading) is the main air pollutant and contributes to respiratory illnesses for company employees as well as employees around the port installation. Several studies have been carried out previously concerning the impact of dust emitted during the extraction, processing, transport and export of phosphate (*Berman, 1969*) and *(Pan, 2002)*. Nevertheless, this work is the first in Algeria, which studied the PM emissions in the air, due to the activities of the Somiphos Company, in particular, exports of phosphate at the port of Annaba.

According to the legal definition, dust is a solid particle with an aerodynamic diameter of less than 100 micrometers or whose falling speed is less than 0.25 meters per second (Article R. 4222-3 of the Labor Code). Dust suspended in the air presents serious health risks, especially if this dust contains a certain amount of heavy metals (Zn, Pb, Cu, As, Cr, U, Ni ...) *(Benselhoub et al., 2015)*. The particles size largely determines the extent of the hazard. Particles larger than 10 µm cannot be inhaled and therefore only affect external organs, they are mainly responsible for skin and eye irritation, conjunctivitis and eye infections *(Romdhane, 2017; Giannadaki et al., 2014; Merlen, 2015).* Particles less than 10 microns can be inhaled but are generally stopped in the nose, mouth and upper respiratory tract; they then trigger conditions such as asthma, tracheitis, pneumonia, allergic rhinitis and silicosis. However, when the fine particles enter the lower respiratory tract and reach the bloodstream, they can affect all internal organs and cause cardiovascular disorders. According to an assessment conducted in 2014 using a global model, exposure to dust particles would have



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caused 400,000 premature deaths due to cardiopulmonary diseases within the population aged over 30 years (*Giannadaki et al., 2014*).

The main objective of this study is to measure the level of dust generated by the phosphate export process using the Owen Gauge method, and then to characterize the dust, such as size and shape of particles, chemical composition, the presence of heavy metals, the impact on the environment, and even the effect of weather conditions.

**Material and Methods.** Emission of dust into the atmosphere is generally a discontinuous phenomenon in terms of flow. Depending on the size, granulometry and nature of the particles and weather conditions (wind, temperature, and humidity), the particles have a more or less long residence time in the atmosphere and will be eliminated according to two processes of separate atmospheric deposits *(Kharytonov et al., 2016)*:

- Dry deposition (gravity, by Brownian motion or by impaction and interception);
- Wet deposition (leaching during rainfall or trapping by cloudy water drops).

The collection in this study is carried out by means of precipitation collectors of the Owen gauge type, this method is of standard NF X 43-014 (*Merlen, 2015*). Owen gauge type collectors (Fig. 1) consist of a funnel and a 25 liters polyethylene collection bottle.

The whole system is inserted into a tripod serving as a support, the collection height (head of the funnel) is located 1.5 meters from the ground, and sometimes the plastic clips are used to fix the bottles to the vertical metal frames of the installation and the phosphate loading gantries.

**Monitoring program.** The monitoring network consists of eight (08) sampling stations arranged to cover the entire field of the port installation and its periphery (Fig. 2a and b); namely the unloading hall of the wagons (Station S1), the unloading hangar for phosphate storage trucks (Station S2). Small Ship Loading Gantry, dock 15 (Station S3), Technical Services (Station S4), Small Ship Loading Gantry, dock 17 (Station S5), Train Station outside the harbour at 700 m (Station S6), Ibn-Sina Hospital at 1200 m (Station S7), and finally the Church of Saint Valentine at 2.3 km (Station S8). The phosphate dust-monitoring network at the company is a permanent network. In the present study and for the sampling and quantification of sedimentable dust, we favoured the period between the ends of March 2019 until the beginning of July 2019, the period is divided into three measurement compaigns, and each compaign lasts 30 days of exposure. This period was chosen for two reasons: the first reason that the weather is dry in this month, which facilitates the dust flight, the second reason that in this period the company exported more than 230.000T of phosphate and about 250.000T between unloading and storage.

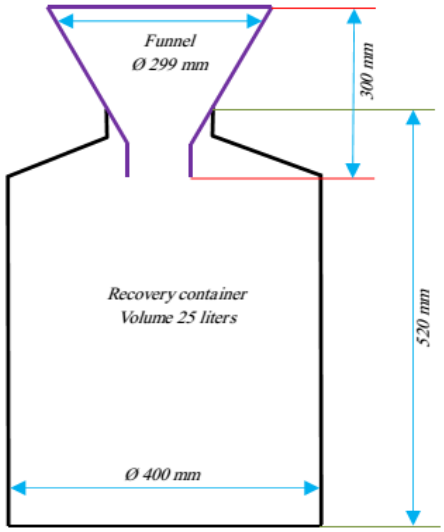


**Fig. 1. Presentation of the measuring device, the precipitation collectors, Owen gauge**

**Measurements process.** The schedule of environmental monitoring conducted during the period mentioned before is presented in Table 1.

***Table 1***

**The sampling follow-up schedule**

| **Years** | **2019** | | | | | |
|---|---|---|---|---|---|---|
| **Months** | March | April | May | June | July | |
| **Company** | 01 | | 02 | | 03 | |
| **Process** | 30 March installation | 29 April removal | 01 May installation | 30 May removal | 02 June installation | 01 July removal |
| **N$^{br}$ of days** | 30 days | | 30 days | | 30 days | |

**Analysis and expression of results.** The analysis of sedimentable dust is carried out in accordance with standard NF X 43-014. The amount of water collected in the gauges during the measurement period is filtered, the filtrate is dried at 105°C for a given time, and then weighed in order to determine the mass and assess the amount of dust contained in each gauge. The results are expressed in milligrams of dust deposited per square meter and per day (mg/m²/d).

The wind roses recorded over the three months of measurements (Fig. 3) generally have the same characteristics. The winds was considered as non-measurable or zero (<5 m/s), measured during the three measurement campaigns are relatively homogeneous, varying from 20.8% to 25.8% of the observations made over the period and in this case, the fallout is not influenced. The prevailing winds (10 to 38 m/s) are predominant and come mainly from the southwest quarter. Winds from the northeast (0° to 60°), are very poorly represented during periods of exposure. Therefore, the measurement stations, especially S6, S7 and S8, are less subject to the phenomena of atmospheric deposition of PM due to their location.

**Results and Discussion.** Once the weight of the dust from each sample has been determined, it is necessary to calculate the major dust content in mg/m²/day, according to the following relationship, given by the AFNOR standard.

$$P = \frac{m}{10^3} \text{x} \frac{10^4}{S} x \frac{730}{t} = 7300 \frac{m}{St}$$ *(Henni-Chebra et al., 2011),*

P – major dust content (mg/m²/day); t – duration of exposure in hours (with the average duration of one month = 730 hours); m – mass in milligrams of PM; S – exposure surface, the bottom of the Owen gauge in cm².

Table 2 presents the results of atmospheric deposition of dust, measured by means of precipitation collectors on the Annaba's port installations and their peripheries, and Fig. 4 shows the data visually.

*Table 2*

PM fallout in mg/m²/day

| Mouths | S1 | S2 | S3 | S4 | S5 | S6 | S7 | S8 |
|---|---|---|---|---|---|---|---|---|
| April | 23215 | 19740 | 14802 | 6102 | 15200 | 305 | 195 | 81 |
| May | 22102 | 19304 | 12300 | 7204 | 18321 | 297 | 245 | 62 |
| June | 19900 | 18810 | 12200 | 7600 | 17287 | 220 | 452 | 54 |


a


b

**Fig. 2. (a) - Location of measurement stations. (b) - Location of the study area and sites**
**Wind conditions data. Low wind speed will be associated with low dispersion of particles in the entourage**

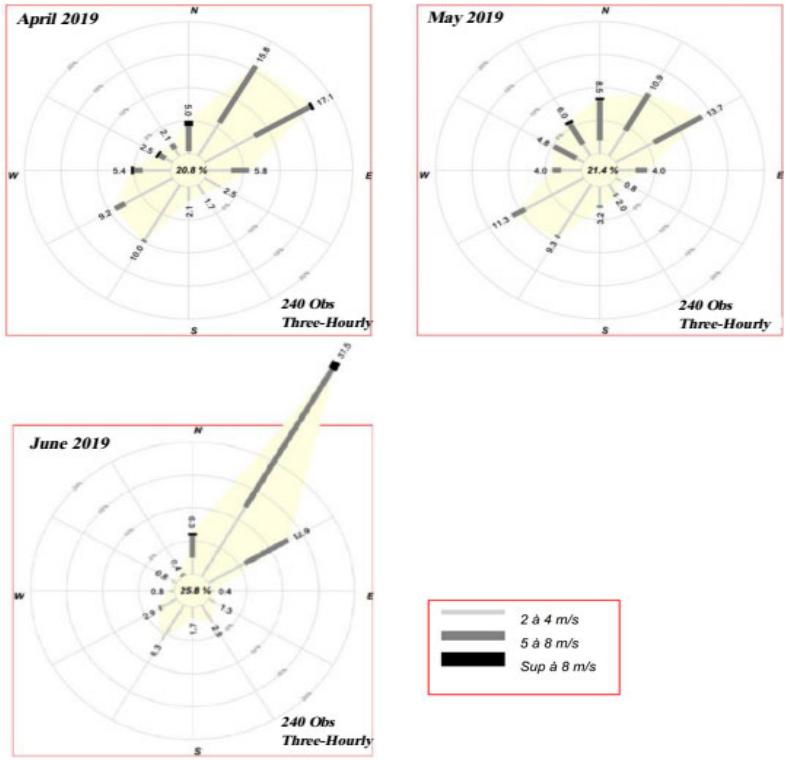


**Fig. 3. Wind regimes recorded during the periods of exposure of the gauges**

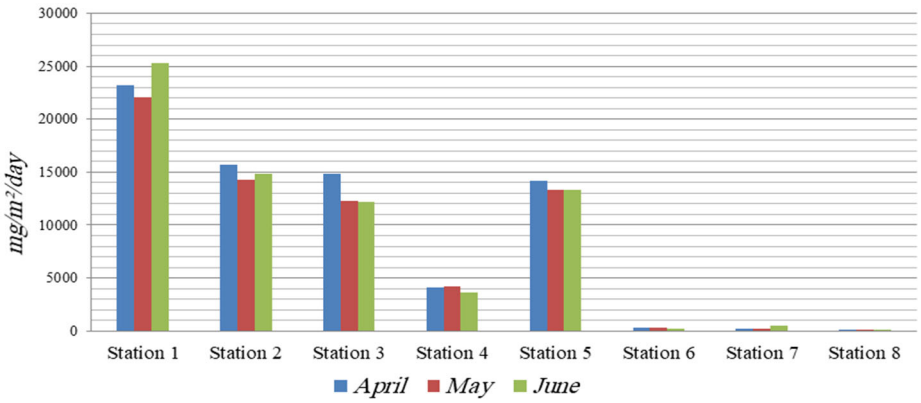


**Fig. 4. Dust level evolution**

The results obtained are illustrated in Fig. 5, for the evolution of the dust content of the eight series, where it is found that station 1 records the highest dust content. Sampling is well above the acceptable threshold set by the AFNOR standard (1000 mg/m$^2$/day), with the exception of stations S6, S7 and S8, which have low rates. The high rate at the other stations is mainly due to the location of the gauges in heavily dusted areas. The wagon hall (station S1) receives more than 4,500 tons of phosphate every day and without stopping, while storage is carried out periodically in a stock. Loading of phosphate into ships at the two docks (15 and 17); is occasional, which explains the low level of dust accumulation at station S1. The quantities of dust collected in stations S6, S7 and S8 are lower than the reference value set by the AFNOR standard, this result is justified by the distance of the stations from areas producing dust. Despite the location of station S7; on the axis of the prevailing north-easterly winds, according to data provided by the meteorological station of Annaba; but the rate does not exceed 452 mg/m$^2$/day, this is due to its presence at a high level above sea level, the propagation of dust has not been affected by winds throughout the period of this study, especially stations S6, S7 and S8. The table below shows the limited dust level of 350 mg/m$^2$/day, according to TA Luft.

**PM chemical composition.** To determine the origin of the PM collected in the eight (08) monitoring points, we carried out chemical analysis of the samples by the XRF; all the samples are mixed; homogenized and analyzed. The result of the chemical composition of the dust collected on the Owen gauge, are compared with that given by the laboratory of the Djebel Onk phosphate complex (Table 3).

***Table 2***

**Dust reference value (mg/m$^2$/d) (*Kharytonov et al., 2017*)**

| Class | Value in mg/m$^2$/d |
|---|---|
| Low dustiness | 0-200 |
| Medium dustiness | 200-300 |
| High dustiness | >350 |

***Table 3***

**Chemical composition of PM phosphate samples**

| Elements | $P_2O_5$ | $CO_2$ | $SO_3$ | CaO | MgO | $Fe_2O_3$ | $Al_2O_3$ | $Na_2O$ | $K_2O$ | $SiO_2$ | F | Cl (ppm) |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| PM of phosphate | 29.31 | 6.93 | 2.80 | 50.8 | 1.67 | 0.32 | 0.33 | 1.27 | 0.071 | 2.16 | 3.56 | 553 |
| Djebel-Onk phosphate | 30.22 | 6.86 | 3.00 | 52.3 | 0.83 | 0.12 | 0.50 | 1.35 | 0.078 | 3.21 | 4.02 | 449 |

The comparison of the values of the composition of PM of phosphate; shows a correlation with the chemical composition of Djebel-Onk phosphate. Stations S1, S2, S3, S4 and S5 are located in the unloading, storage and loading areas, the dust even comes from truck traffic, especially at station S4. The quantity collected at stations S6, S7 and S8, is so negligible that it does not affect the chemical composition of the aggregate sample. These results clearly prove that the PM collected in the installations is generated by the process of unloading, storage and unloading of phosphate.

**Determination of particle size distributions.** The particle size distribution of the phosphate dust sample, is obtained from the analysis by laser diffraction (Fig. 5), the result of the analysis showed that, more than 90% of the particles are with a smaller diameter at 118 µm; 50% of the particles are with a diameter less than 13.3 µm, and 10% of the particles have a diameter of less than 5.5 µm.

These results favour deep pulmonary deposition [8]. PM10 (particles with an aerodynamic diameter of less than 10 µm) are of major concern today, as they are small enough to penetrate deep into the lungs (WHO 2013). Particle size can behave in the human body as follows:

- Chest dust: mass fraction of inhaled particles enter the larynx (<30 µm);
- Alveolar dust: mass fraction of inhaled particles penetrate the non-ciliated airways (<15 µm). These fractions are included in the ISO 7708 standard and in the Afnor X43-100 standard *(Betelli et al., 2012)*.

**Phosphate PM Morphology.** The shape of PM in general can promote flight and decrease the sedimentation rate of a particle: a flat particle behaves like a leaf during its fall and will therefore settle more slowly than a spherical particle *(Imen Bel Hadj, 2013)*. The fall speed is a function of the aerodynamic diameter. PM of phosphate captured at the port of Annaba have different morphology and irregular shape (Fig. 6).

The angular shape of the particle surface is mainly due to the mechanical preparation of phosphates at the Djebel-Onk plant, unlike natural particles, where the surface is smooth and the grains are spherical in shape. For particles of irregular shape, relative movement becomes difficult due to the presence of more points of contact between them. If they are elongated and hook-shaped particles, it will be more complicated because they tend to form bridges by interlocking particles *(Gil et al., 2013)*.

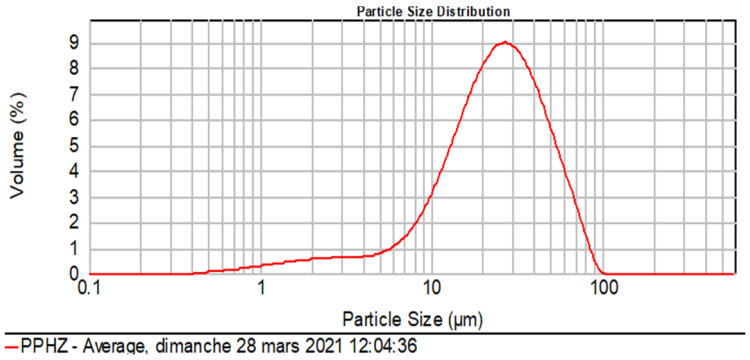


**Fig. 5. PM particle size by laser diffraction**

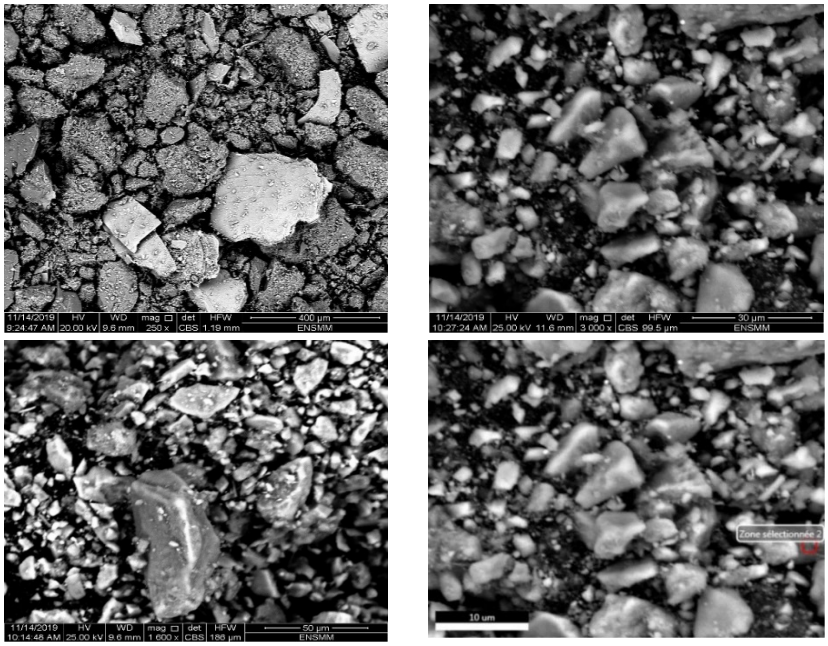

**Fig. 6. Phosphate dust morphology, by X-ray microanalysis coupled with a scanning electron microscope (SEM / EDX)**

**Heavy metals content in phosphate dust.** From a purely scientific and technical point of view, heavy metals can be defined as: Any metal with a density greater than 5 g/cm$^3$. Any metal with a high atomic number, generally higher than that of Sodium (Z=11), presenting a danger to the environment and to humans. Any metal that can be toxic to biological systems. In environmental sciences, the heavy metals associated with the concepts of pollution and toxicity are generally: arsenic (As), cadmium (Cd), chromium (Cr), copper (Cu), mercury (Hg), manganese (Mn), nickel (Ni), lead (Pb), tin (Sn), and zinc (Zn). In our study, the composition of heavy metals and some trace metallic elements was determined by atomic absorption spectrometry (Table 4).

The results of the metal concentrations measured for the eight stations present different values, those, which are higher than the German limit values, and those that are lower. We observe that the guide values for the metals of Cadmium (Cd) and mercury (Hg) have been exceeded. The limit value of Cd is 2 µg/m$^2$/d, on the other hand, the recorded values varied between 8.7 and 1.2 µg/m$^2$/d.

***Table 4***

**Daily and surface concentration of metals detected (µg/m$^2$/d) - German TA Luft 2002 (*Henni-Chebra et al., 2015*)**

| Element | Cd | Zn | Pb | Cu | Cr | Ni | Mn | Hg | Co | Sb |
|---|---|---|---|---|---|---|---|---|---|---|
| Limit µg/m$^2$/d | 2 | 400 | 100 | 100 | 250 | 15 | 15 | 1 | 15 | 100 |
| S1 | 8.3 | 113.2 | 15.8 | 23.4 | 122.2 | 13.5 | 11.3 | 3.2 | 2.2 | 12.4 |
| S2 | 8.7 | 111.3 | 16.7 | 25.1 | 124.8 | 14.5 | 10.9 | 2.2 | 1.4 | 13.8 |
| S3 | 7.6 | 102.2 | 10.2 | 20.1 | 121.1 | 5.4 | 12.3 | 3.4 | 3.3 | 14.3 |
| S4 | 5.3 | 72.5 | 7.5 | 14.2 | 113.4 | 8.8 | 8.4 | 4.1 | 2.9 | 10.7 |
| S5 | 5.6 | 80.4 | 9.6 | 10.0 | 115.7 | 9.1 | -- | -- | 2.8 | 8.2 |
| S6 | 3.2 | 20.1 | -- | -- | 100.4 | -- | -- | 2.1 | 2.1 | 4.2 |
| S7 | 1.2 | -- | -- | 2.4 | 5.2 | -- | 3.4 | 0.1 | 1.4 | 3.2 |
| S8 | -- | 13 | -- | -- | 1.2 | -- | -- | 0.3 | 1.2 | -- |

The limit value of Hg according to the German standard, is 1 µg/m$^2$/d, but the results of the latter's analyzes vary between 3.2 and 0.3 µg/m$^2$/d in the eight measuring stations. The values of Zn, Pb, Cu, Cr, Ni, Mn, Co and Sb are below the limit of quantification. The points most exposed to fallout from the site (station S1, S2 and S3) have the highest levels of heavy metals.

The toxicity of mercury mainly affects cerebral functions (mercury is neurotoxic), renal functions (nephrotoxic), the endocrine system; mercury is suspected to be an endocrine disruptor and cell life disruptor. It was recently shown (2007) that even at low doses, it has a cytotoxic effect on central nervous system stem cells (as well as low doses of lead or parquât) *(Li et al., 2007)*. It can affect cell division; mercury is suspected of being able to induce or co-induce certain cancers (For example, cases of leukemia have been observed in Japan concomitantly in Minamata). In animal models, it can induce alterations in the blood, apoptosis (*Sharma et al., 2005; Waalkes et al., 2000.; Nagashima et al., 1997)*, and chromosomal aberrations. Cadmium poisoning can occur either acutely or chronically, with lesions mainly in the lungs, bones and kidneys. Cadmium has no physiological role in the human body *(Nishijo et al., 2004.: Nordberg et al., 2002.; Benselhoub et al., 2015)*. The metal itself and its compounds are extremely toxic, even at low concentrations, and tend to accumulate in living organisms and ecosystems *(Orlowski et al.,1998)*.

Usually to control dust emissions; several methods can be applied. For phosphate dust the method is special, because the product must not be touched by humidity, otherwise it will be contaminated, therefore it is strictly forbidden to spray the dust with water especially at the unloading points of trucks and wagons, at the level of the loading gantries of the ships. The most effective methods for reducing phosphate dust are:

- Complete cover of the product conveyor belts;
- Unloading of trucks and wagons in well closed and airtight sheds, to prevent dust from escaping outside the installation;
- Installation of baghouse dust collectors at ship loading gantries;
- Spraying the road with water;
- Complete cover of the product conveyor belts;
- Unloading of trucks and wagons in well closed and airtight sheds, to prevent dust from escaping outside the installation;
- Installation of baghouse dust collectors at ship loading gantries;
- Spraying the road with water to prevent dust from flying away during the passage of trucks.

**Conclusions.** Undoubtedly, the process of exporting phosphate in the port of Annaba, leads to a rate of dust emissions that exceed all international standards. What is certain is that the effects and consequences of these emissions on humans and the environment are disastrous. It is likely in the future, there will be a thorough study, dealing with the effects of these emissions on man, the environment, fauna and flora.

Based on the results obtained in this study, the following conclusions can be listed:

- The sedimentable PM measurement campaign gave very variable levels of dust during the eight sampling stations. Stations S1, S2, S3, S4 and S5 greatly exceed the acceptable threshold (1000 mg/m²/day) defined by the AFNOR standard, with a very low dust level for station S6, S7 and S8. This difference in dust level is mainly due to the distance between the collection points and the source of the dust.
- The chemical composition of the PM determined by the XRF shows a correlation between the chemical composition of the PM samples from the stations and the chemical composition of the phosphate from the Djebel-Onk plant located in Tebessa.
- The particle size distribution of PM, determined by laser diffraction, shows that more than 90% of particles with a diameter less than 118 µm, 50% of particles with a diameter less than 13.3 µm and 10% of particles have a diameter less than 5.5 µm. These results promote deep lung deposition upon inhalation of PM by humans.
- PM of phosphate captured at the port of Annaba have other morphology and irregular shape of different size. The angular shape of the particle surface is mainly due to the mechanical preparation of phosphates at the Djebel-Onk phosphate plant, unlike natural particles where the surface is smooth and the grains are spherical in shape.
- The results of the analysis by atomic absorption spectrometry presents an excess in the values of Cadmium (Cd) and mercury (Hg), the levels of the latter are higher than the German limit values (TA Luft 2002).

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З. Мекті[1],
E-mail: mektizohir@yahoo.fr;
А. Бутемеджет[1],
E-mail: boutemedjetassia@yahoo.fr;
О. Секіу[2],
E-mail: o.sekiou@cre.dz;
С. Бердуді[1],
E-mail: berdoudisaid@yahoo.fr;
А. Шайб[1],
E-mail: achaib_as@yahoo.fr;
Ф. Куйдер[3],
E-mail: kouiderfareskul016@gmail.com;
А. Рахмані[3],
E-mail: Abdun81@gmail.com;
А. Бенселгуб[2],
E-mail: benselhoub@yahoo.fr;
[1]Лабораторія валоризації гірничих ресурсів та навколишнього середовища, Гірничодобувний відділ,
Університет Баджі Мохтара, Аннаба, Алжир;
[2]Дослідницький центр навколишнього середовища (C.R.E), Аннаба, Алжир;
[3]Інженерний факультет, Навколишнє середовище та інформаційні технології, Університет SEGi, Малайзія

## ЗАБРУДНЕННЯ АЕРОЗОЛЬНИМИ ЧАСТИНКАМИ, ЩО ПОВ'ЯЗАНЕ З ЕКСПОРТОМ ФОСФАТУ У ПОРТУ АННАБА

*Протягом кількох останніх десятиліть забруднення твердими частинками повітря (ТЧ) стає однією з основних проблем політиків у всьому світі через його несприятливий вплив на клімат, здоров'я людей, навколишнє середовище та різні соціально-економічні сектори. Процес експорту фосфатів у великих обсягах є одним з основних джерел, що генерують забруднювачі повітря, такі як викиди твердих частинок різної форми, концентрації та розміру, які можуть спричинити захворювання органів дихання. Метою даної роботи є визначення рівня запиленості, що утворюється в процесі розвантаження, зберігання та завантаження фосфату навалом на судна портовою установкою в порту Аннаба (схід Алжиру).*

*Вимірювання ТЧ відбувалось відповідно до NF X 43-014, воно проводилось на 08 точках відбору проб, розташованих у порту Аннаба та на периферії з радіусом до 2,3 км. Отримані результати показали дуже високий рівень пилу під час вивчення серії зразків 05, що значно перевищує допустимий поріг (1000 мг/м²/добу). Рівень важких металів у пилу було оцінено за допомогою атомно-абсорбційного спектрометричного аналізу. Аналіз EDAX показав, що фосфатні ТЧ мають кутову морфологію з видовженими частинками та гачкоподібною формою.*

*Ключові слова: ризик для здоров'я, важкі метали, довкілля, вплив, північний схід Алжиру.*

З. Мекти[1],
E-mail: mektizohir@yahoo.fr;
А. Бутемеджет[1],
E-mail: boutemedjetassia@yahoo.fr;
О. Секиу[2],
E-mail: o.sekiou@cre.dz;
С. Бердуди[1],
E-mail: berdoudisaid@yahoo.fr;
А. Шайб[1],
E-mail: achaib_as@yahoo.fr;
Ф. Куйдер[3],
E-mail: kouiderfareskul016@gmail.com;
А. Рахмани[3],
E-mail: Abdun81@gmail.com;
А. Бенселгуб[2],
E-mail: benselhoub@yahoo.fr;
[1]Лаборатория валоризации горных ресурсов и окружающей среды, Горнодобывающий отдел,
Университет Баджи Мохтара, Аннаба, Алжир;
[2]Исследовательский центр окружающей среды (C.R.E), Аннаба, Алжир;
[3]Инженерный факультет, Окружающая среда и информационные технологии, Университет SEGi, Малайзия

## ЗАГРЯЗНЕНИЕ АЭРОЗОЛЬНЫМИ ЧАСТИЦАМИ, СВЯЗАННОЕ С ЭКСПОРТОМ ФОСФАТА В ПОРТУ АННАБА

*На протяжении последних десятилетий загрязнение твердыми частицами воздуха (ТЧ) становится одной из основных проблем политиков во всем мире из-за его неблагоприятного влияния на климат, здоровье людей, окружающую среду и различные социально-экономические секторы. Процесс экспорта фосфатов в больших объемах является одним из основных источников, генерирующих загрязнители воздуха, такие как выбросы твердых частиц разной формы, концентрации и размера, которые могут вызвать заболевания органов дыхания. Целью данной работы является определение уровня запыленности, образующегося в процессе разгрузки, хранения и погрузки фосфата навалом на суда портовой установкой в порту Аннаба (восток Алжира).*

*Измерение ТЧ производилось в соответствии с NF X 43-014, оно проводилось на 08 точках отбора проб, расположенных в порту Аннаба и на периферии с радиусом до 2,3 км. Полученные результаты показали очень высокий уровень пыли при изучении серии образцов 05, что значительно превышает допустимый порог (1000 мг/м²/сутки). Уровень тяжелых металлов в пыли был оценен с помощью атомно-абсорбционного спектрометрического анализа. Анализ EDAX показал, что фосфатные ТЧ имеют угловую морфологию с удлиненными частицами и крючкообразной формой.*

*Ключевые слова: риск для здоровья, тяжелые металлы, окружающая среда, влияние, северо-восток Алжира.*