

**BABEȘ-BOLYAI UNIVERSITY CLUJ – NAPOCA  
FACULTY OF ENVIRONMENTAL SCIENCES AND  
ENGINEERING**



**STUDIES ON PHYSICAL AND CHEMICAL PROPERTIES  
AND THE DISTRIBUTION OF MAIN ATMOSPHERIC  
POLLUTANTS IN CLUJ COUNTY**

**DOCTORAL DISSERTATION ABSTRACT**

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**Keywords:** atmospheric pollutants, ambient air, pollution sources, EMEP/CORIMAR, Emission Inventory Guidebook, concentrations, mineral crystallography, X-ray diffraction, mineral optical microscopy, pollution dispersion modeling, atmospheric pollutants distribution.

## Chapter I. Introduction

Currently, environmental protection is a major global concern. The factors leading to environmental pollution are of technological, economic, and social origin.

The main characteristic of the environment is its self-regulation ability and capacity; however, if certain limits are exceeded, gradual environmental degradation occurs, leading to a destructive state. [Muntean et al. 2000]

Air pollution is defined as the existence of chemical compounds, in gaseous, solid, or liquid form, in certain amounts and lengths of time which render ambient air as hazardous for human health and the environment.

The purpose of the present study is an investigation concerning the physical and chemical properties of the main atmospheric pollutants in Cluj County and their mapping. These studies have involved multiple stages during the research project, which started in 2007.

During the first stage, the studies focused on the identification of the existing pollution sources in Cluj County. For this purpose, extensive due diligence and research were performed with respect to the classification and nature of atmospheric pollutants.

Therefore, we elaborated a comprehensive summary of main atmospheric pollutants in Cluj County, based on a comprehensive source inventory, along with a classification of sources of such pollutants.

The above classification was based upon the field where the pollution source exists, various raw materials and fuel types involved in production process, as well as the technological characteristics of various retention systems employed for the retention of gaseous pollutants resulting from industrial processes. A quantitative database of pollutants from various sources was consequently developed.

The second stage involved an in-depth theoretical approach, to establish the array of analytical methods for the targeted pollutants. The legislative background, national standards, and the currently enforced norms based upon which the atmospheric pollution is assessed were researched and identified. This was complemented by the study of the basic scientific principles, and the evaluation of the necessary scientific instrumentation for sample collection and analytical work.

The third stage strictly targeted the research concerning the physical and chemical properties of the main atmospheric pollutants in Cluj County and their distribution. We carried out measurements of the main indicators present in the ambient air: NO<sub>x</sub>, NO, NO<sub>2</sub>, SO<sub>2</sub>, CO, O<sub>3</sub>, PM<sub>10</sub>, Pb, Cd, and Ni from PM<sub>10</sub> and monitored the evolution of their respective

concentrations. Using a comparison of recorded data with the corresponding data in the emission inventory, as well as meteorological information for all studies indicators, we applied the OML-multi model of Gaussian dispersion. The results revealed the distribution of atmospheric pollutants in Cluj county, based on the elaborated dispersion models.

In order to have a comprehensive view of air quality, the studies have continued – by additional crystallographic and mineral investigations of solid suspended materials sampled from air from various spots within the Cluj-Napoca city area.

Due to the fact that during winter the concentrations of  $PM_{10}$  were recorded at elevated levels, compared to the warmer months, we were concerned about the reasons behind the increase. Consequently, the purpose of our study was widened and we sought to establish the influence of ice-melting materials applied on the roads upon the air quality.

## **Chapter II. Pollution Sources in Cluj County**

Pollutants do not act locally, around their emission area, but they are disseminated in space, in the ambient air, in surface and ground waters, in ground, affecting the global environment.

The following categories of pollution sources in Cluj County were investigated:

- ❖ Traffic
- ❖ Industrial activities
- ❖ Storage and distribution of fossil fuels
- ❖ Solvents use
- ❖ Incineration of residual materials
- ❖ Water reclamation
- ❖ Agriculture
- ❖ Other sources

### **Chapter III. Methods of Analysis for Air Pollutants**

In order to establish an array of analytical methods covering the range of targeted pollutants, legislation, national standards, and the currently enforced norms based upon which the atmospheric pollution is assessed were researched and identified. Additionally, the evaluation of the necessary scientific instrumentation for sample collection and analytical work was completed.

### **Chapter IV. Mineral and Crystallographic Investigations of Air Samples**

For material samples, representing airborne powder collected in Cluj-Napoca, were studied.

Considering the occurrence of material particles of mineral origin in ambient air of urban centers [Joshi et al, 2009] Tasic et al, 2006, Elihn, Berg, 2009; Navaro, Sebastian, 1996; Moldoveanu, 2005] a mineral and crystallographic investigation of the samples was deemed necessary.

A comprehensive characterization of the samples was carried out by correlating the X-ray diffraction results with the results obtained by optical microscopy in transmitted and cross-polarized light [Arghir, Ghergari, 1989; Cullity, Stock, 2001; Arghir, 1993].

All four samples were investigated under identical conditions, at room temperature and atmospheric pressure.

Due to the adherence of subject particles to the wall of the crucible, we used doubly distilled water for their removal, followed by their placement on glass plates for microscopy, and the amorphous cuvettes for X-ray diffraction.

X-ray diffraction analysis was performed by a DROM-3 type instrument, equipped with Matmec VI.0. data acquisition and recording software.

The diffraction range was 20-100 degrees  $2\theta$  with post-acquisition fine structure detailing across the 20-45 degrees  $2\theta$ . We used a Cu anticathode with  $\text{Cu}_{k\alpha}$   $\lambda = 1.54184 \cdot 10^{-10}$  m, at a recording speed of one degree/minute. The source settings were: horizontal aperture 4 mm, vertical aperture 8 mm, Soller collimator 1.5 deg div, Ni filter thickness 20  $\mu\text{m}$ , horizontal aperture 0.5 mm, vertical aperture 12 mm, time constant 5 seconds, pulse range  $1 \cdot 10^3$  imp/sec. The corresponding interplanar distances were calculated based in Bragg's Law, aided by Matmec VI.0. software.

The mineral content of the sample were identified by comparing the calculated interplanar distances with those included in the specific database Match – Phase Identification from Powder Diffraction, Crystal Impact Co. 2008.

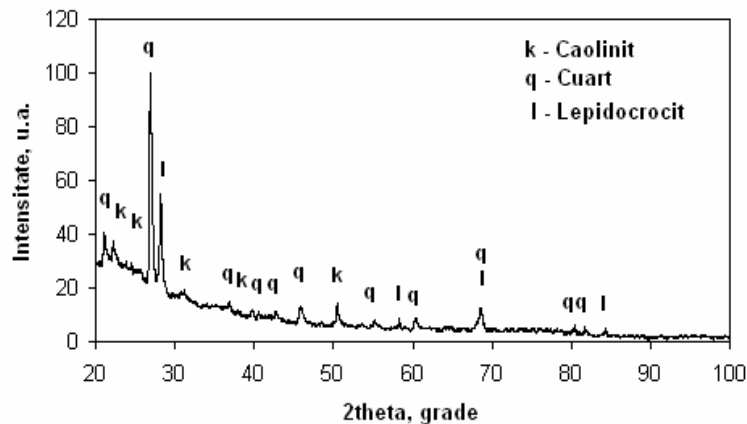
The morphologic aspects of the PP sample were determined by dark field optical microscopy. We used an IOR-8 optical microscope equipped with a Samsung 8 MPx digital camera and the Image J 1.40g specialized microphotographic analysis software (National Institute of Health, USA) [Rasband, 2009].

The mineral microscopic analyses were performed with an optical microscope Laboval 2 – Carl Zeiss Jena, using transmitted and cross-polarized light. Digital captioning was achieved using a Samsung 8 MPx camera.

#### 4.1. Investigation of air samples accumulated during weekdays and weekends

Sample 1 – collected from Dambovitei Street, accumulated during the January 17-21, 2011 period, in an amount of 0.020 g, as deposited on crucible’s wall.

The diffractogram for the P1 sample is shown in Figure 1. We noticed well defined diffraction maxima, indicative of crystalline components that are well developed at microstructure level. The appearance of a net maximum followed by a relatively complex succession of less intense maxima indicates a multi-component, multi-phase crystalline sample.



thorns, which erode in time due to reciprocal abrasion. Such particles can be found in a wide dimensional range, in various sand types.

The second determined component, in terms of presence in the sample, is lepidocrocite, its interplanar distances corresponding to the PDF #88-2212 reference data [26]. This represents  $\gamma$  iron hydroxide, with the chemical formula  $\text{FeOOH}$ .

Caolinite –  $\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4$  – hydrated aluminum silicate. This material crystallizes in a bipyramidal, orthorhombic shape, which leads to the granular, scales-like, appearance of the particles [Arghir, Gherghari, 1989].

The third mineral component of the P1 sample is caolinite; the interplanar distances correspond to the database (PDF #01-0527) [Hanawalt et al. 1938].

The relative intensities of its specific maxima do not exceed 25%, which denoted its less significant presence in the sample, as compared with quartz and lepidocrocite. It is part of the clay minerals category, crystallizing in triclin shape [Arghir, Gherghari, 1989] and it is an unual component of the soils that are adjacent to roads, being frequently found in road dust. [27]

The distribution of minerals in the bulk of sample P1 can be revealed by mineral optical microscopy. One can observe (Fig. 2a and 2c) the overall morphologic appearance of the sample, particularly the larger entities, with an average diameter of 25  $\mu\text{m}$  – illustrated as dark-colored and accompanied by very fine particles located behind.

The microscopic examination under cross-polarized light (Fig. 2b and 2d) distinctly reveals the particles' match with the crystalline species identified by X-ray diffraction. Quartz appears as grey-greenish, lepidocrocite as reddish-rusty, and caolinite bright white. [Arghir, Gherghari, 1989].

We also noticed an over 50% dominance of quartz particles within the investigated microstructures, lepidocrocite accounting for around 30%, while the balance is attributed to caolinite (the very fine particles visible under polarized light as white dots, sometimes yellowish-white).

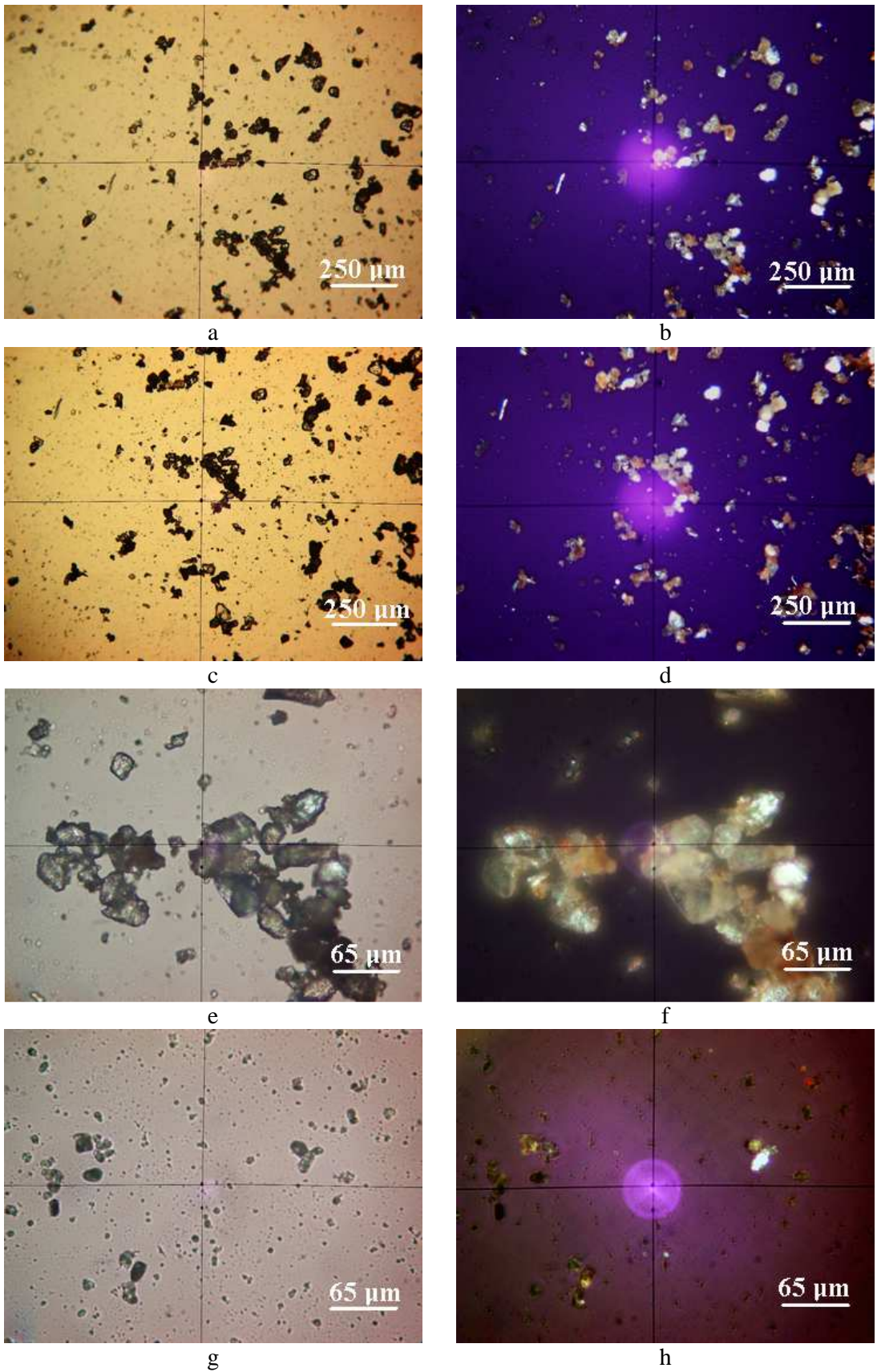


Figure 2. Microscopic aspects of P1 sample: a, c, e, g) micropictures with transmitted light; b, d, f, h) in cross-polarized light

Zoomed details (Fig. 2e and 2f) further underline the general observations with respect to the overall appearance of the P1 sample. The equiaxial shape of quartz particles can be distinctly observed in both transmitted light (Fig. 2e) and cross-polarized light (Fig. 2f).

In this micropicture we observed an interesting detail – one of the quartz particles includes lepidocrocite, fact which is in agreement with other literature reports [28,29].

Zoomed microstructural details (Fig. 2g and 2h) reveal the presence of PM<sub>10</sub>-type material particles (the larger), and PM<sub>2.5</sub> - type (the smaller). The larger appear colored in shades that are typical for quartz, whereas some of the very small particles appear as white or yellowish-white dots, the latter being caolinite particles.

The very fine caolinite particles are also observed in Figures 2b and 2d. In specialized references it was reported the tendency of caolinite to form flaky particles with dimensions consistent with the PM<sub>1</sub> and PM<sub>2.5</sub> range, rather than PM<sub>10</sub> [Zbik et al, 2008; Houwing, Rijn, 1998; Hosu Prack et al, 2010].

The determination of the very fine particles of PM<sub>2.5</sub> and Pm10 type, the sample P1 was subjected to an optical inspection by dark field reflected light, complemented by quantitative analysis, in order to establish the mean diameter of such particles.

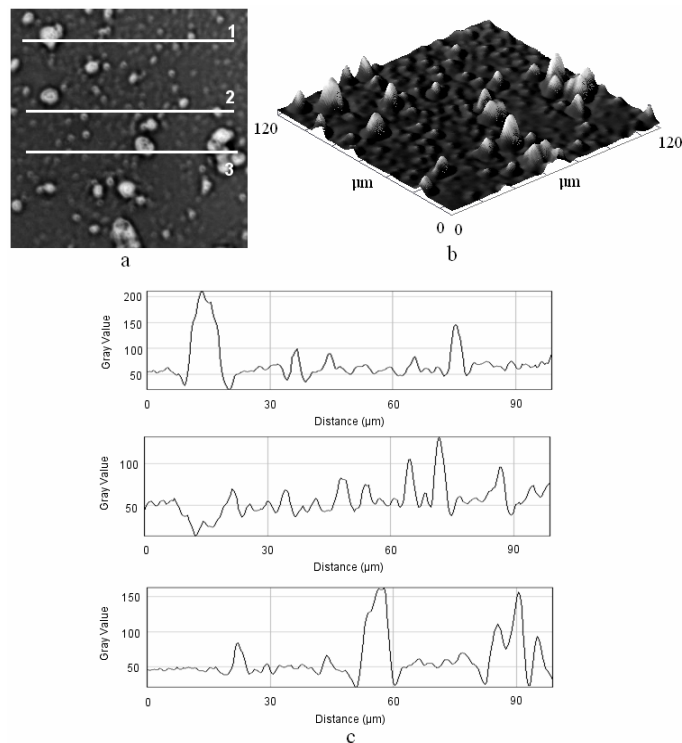


Figure 3. Microscopic quantitative analysis (dark field) for sample P1: a) the resulted micropicture, b) the 3-D presentation of figure 3a; c) profile details corresponding to the white lines in figure 3a

In Figure 3 we observed the very fine particles corresponding to PM<sub>2.5</sub>, as very small white dots, while the larger ones are apticles of the PM<sub>10</sub> type. The aspect of the latter can be better observed in a 3-D presentation (Fig. 3b).

The profile details revealed by dark field light analysis (Fig. 3c), one can determine the diameter of particles at half-height of each particle in the profile. Therefore, the mean diameter of the PM<sub>2.5</sub> particles are around 2 μm, while the mean diameter of the PM<sub>10</sub> particles is around 9 μm.

As a result, sample P1 consists of microscopic particles, moderately sized, with the mean diameter of approximately 25 μm, formed by quartz and lepidocrocite accompanied by PM<sub>2.5</sub> and PM<sub>10</sub> consisting of all three mineral species identified.

The presence of material mineral particles larger than 10 μm is consistent with very strong mixing of street dust due to heavy traffic during weekdays.

Sample 2 – collected from Dambovitei Street, accumulated during the weekend of January 21-23, 2011, in an amount of 0.004 g, as deposited on crucible's wall.

The diffractogram for sample P2 is presented in Figure 4.

Figure 4 reveals well defined maxima, less intense than those in sample P1, and we notice significant band widening, indicating the contribution of crystalline materials, correlated with the massive presence of fine particles of PM<sub>2.5</sub> and PM<sub>10</sub> type.

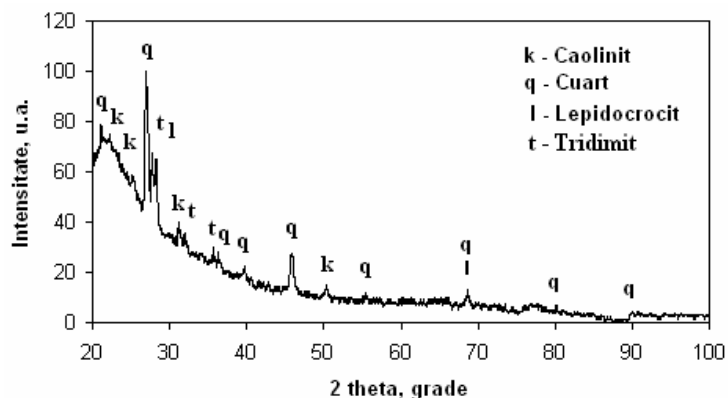


Figure 4. Sample P2 diffractogram – collected January 21-23, 2011, Dambovitei Street

The most intense maximum is assigned to quartz, while we identified eight more distinct maxima. The context confirms quartz as being the predominant component. The second contributor, in terms of importance, is lepidocrocite.

In a similar fashion to the analysis of P1 sample, we identified kaolinite, with the same ratio of relative intensities.

The major difference is that in sample P2 we identified tridimite, along which we identified other two distinct maxima, fact which confirms literature findings [30].

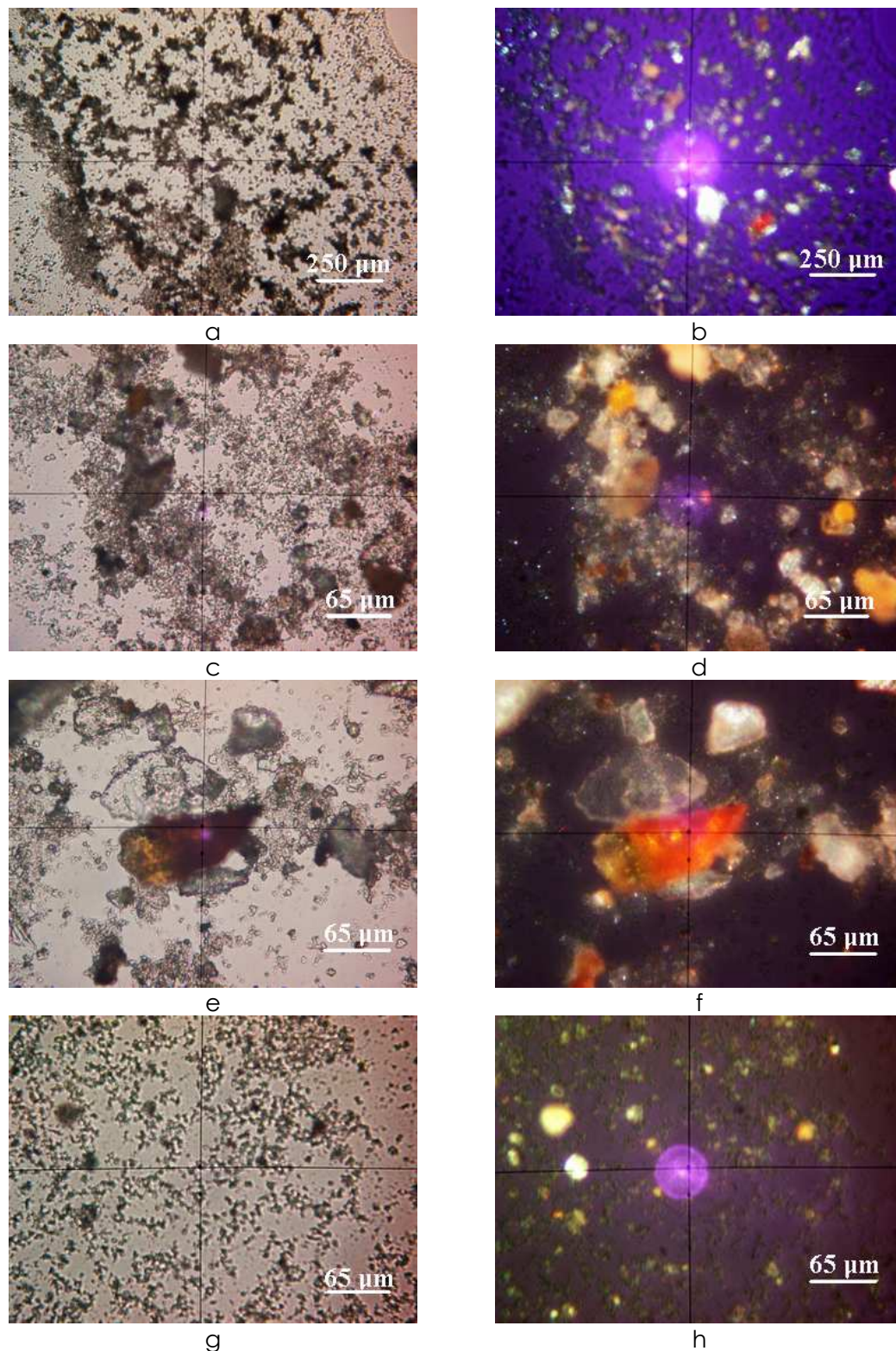


Figure 5. Microscopic aspects of P2 sample: a, c, e, g) micropictures with transmitted light; b, d, f, h) in cross-polarized light

Tridimite is an allotropic form of silica dioxide, which crystallizes in an orthorhombic shape, as opposed to quartz, which crystallizes hexagonally. Tridimite is formed by heating quartz to elevated temperatures (i.e., 1470 deg. C), followed by cooling. The modification is catalyzed by the presence of certain impurities, such as iron or calcium. [Baldo, Santos, 2002]. The specific shape of tridimite particles is tabular-lamellar.

An overall view of sample P2 can be observed in Figures 6.36 a and b. There are two types of particle association – associations of medium sized microscopic particles, and associations of small size microscopic particles of PM2.5 and PM10 type. We observed the predominance of small microscopic particles. In cross-polarized light, we observed grey-greenish quartz particles, and brown-reddish lepidocrocite particles.

We also observed (Fig. 5b) certain grey-greenish particles with tabular-lamellar structure corresponding to tridimite, whereas caolinite particles appear as small white-yellowish dots. Zoomed images (fig. 5c and 5d) allow a clearer distinction of the above. In Figure 5c, we very clearly observed the association of larger particles, with a mean diameter of approximately 25  $\mu\text{m}$  (smaller than in sample P1).

The same view, in polarized light, clearly reveals three light brown-reddish lepidocrocite particles, a few equiaxial quartz particles, and a larger quartz particle (oblong, 65  $\mu\text{m}$  in length and 50  $\mu\text{m}$  in diameter), with fine inclusions of lepidocrocite.

We observed very fine particles of caolinite, white-yellowish in color, as a function of their position with respect with the microscope's optical axis. The details in Fig 5e and 5f include two particles that are particularly interesting: the reddish particle, lepidocrocite, with a length of 75  $\mu\text{m}$  and a width of 50  $\mu\text{m}$ , and a tabular, grey-greenish [article of tridimite, with a length of approximately 70  $\mu\text{m}$  and a width of 50  $\mu\text{m}$ .

The arrangement of the very fine particles (Fig 5g and 5h) is visible under polarized light, with some quartz and white-yellowish caolinite particles being visible. The quantitative microscopic investigation in dark field (Fig. 6) underlines similar details with those presented in Fig. 5g.

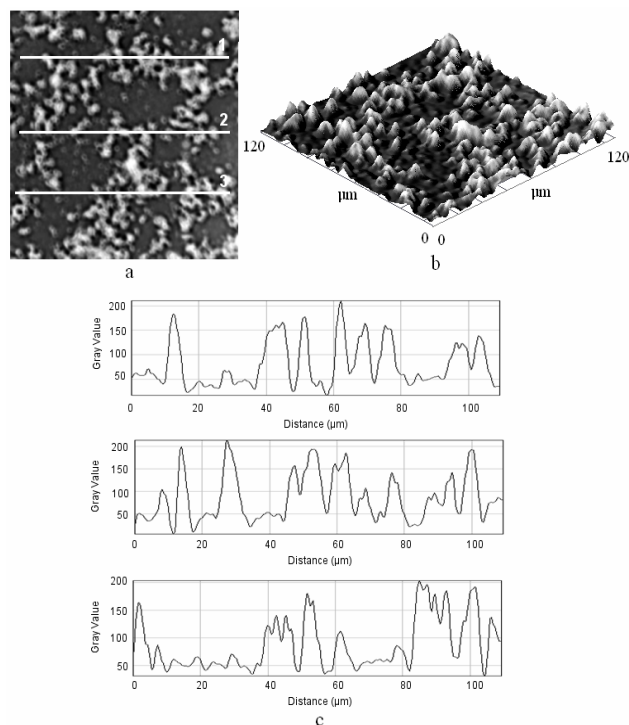


Fig. 6. Microscopic quantitative analysis (dark field) for sample P2: a) the resulted micropicture, b) the 3-D presentation of figure 6a; c) profile details corresponding to the white lines in figure 6a.

Based on the profile details in Figure 6 we could establish the mean diameter of PM<sub>2.5</sub> as being 2  $\mu\text{m}$ , while PM<sub>10</sub> may be associated with an approximate value of 7  $\mu\text{m}$ . We noticed that these values are significantly smaller than those resulted for sample P1.

Sample 3 – collected from Aurel Vlaicu Street, accumulated during the week of January 17-21, 2011, in an amount of 0.014 g, as deposited on crucible's wall.

The diffractogram for sample P3 is presented in Figure 7. Its general appearance consists of well-defined maxima, very pronounced, which is consistent with a predominance of crystalline components in the bulk of sample P3. The interpretation of Figure 7 allows the determination of diffraction data and the identification of crystalline compounds (phases) in the sample.

It was observed that the main component of sample P3 is quartz, silica dioxide, hexagonally crystallized. This is represented by the most intense maximum, and it is accompanied by eight additional representative maxima, the interplanar distance values being consistent with the data reported in the literature [Hanawalt et al, 1938].

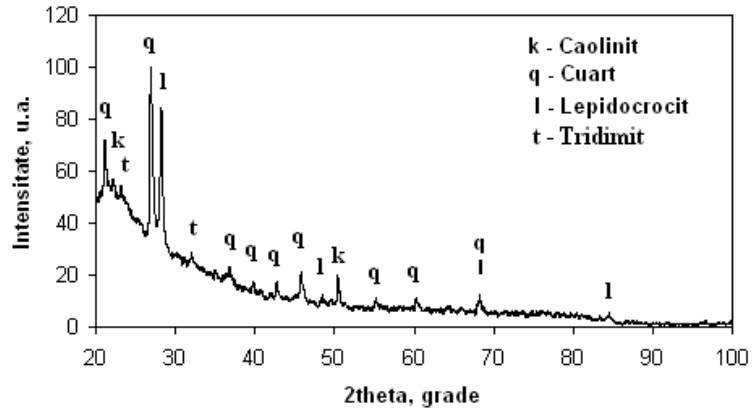


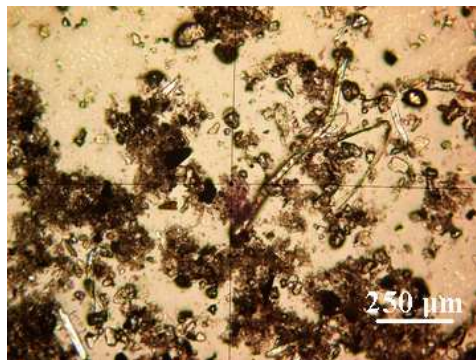
Figure 7. Sample P3 diffractogram – collected January 17-21, 2011, Aurel Vlaicu Street

The next compound in the sample, importance-wise, is lepidocrocite. The third identified compound is caolinite. These facts allowed us to estimate its proportional contribution to the mass of the sample to about 15%. The rest of 5% is allocated to tridimite, whose presence is indicated by the two relatively less intense maxima (15% and 18% respectively). This component being orthorhombic silica dioxide, specific to elevated temperature heating, there is a possibility that it originated in street dust.

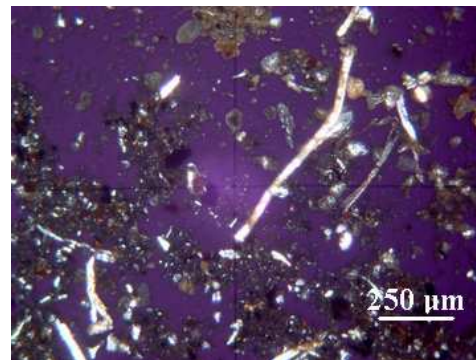
An overall view of sample P3 is shown in Fig. 8a and 8b. Equiaxial particles of medium size, with a mean diameter of about 60  $\mu\text{m}$  are observed, surrounded by accompanying smaller particles.

The massive presence of some fibrous materials (probably traces of textile material) which do not belong to minerals class was also noted.

Under polarized light (Fig. 8b) we observed the predominance of quartz particles, grey-greenish in color, and some dark brown lepidocrocite particles.



a



b

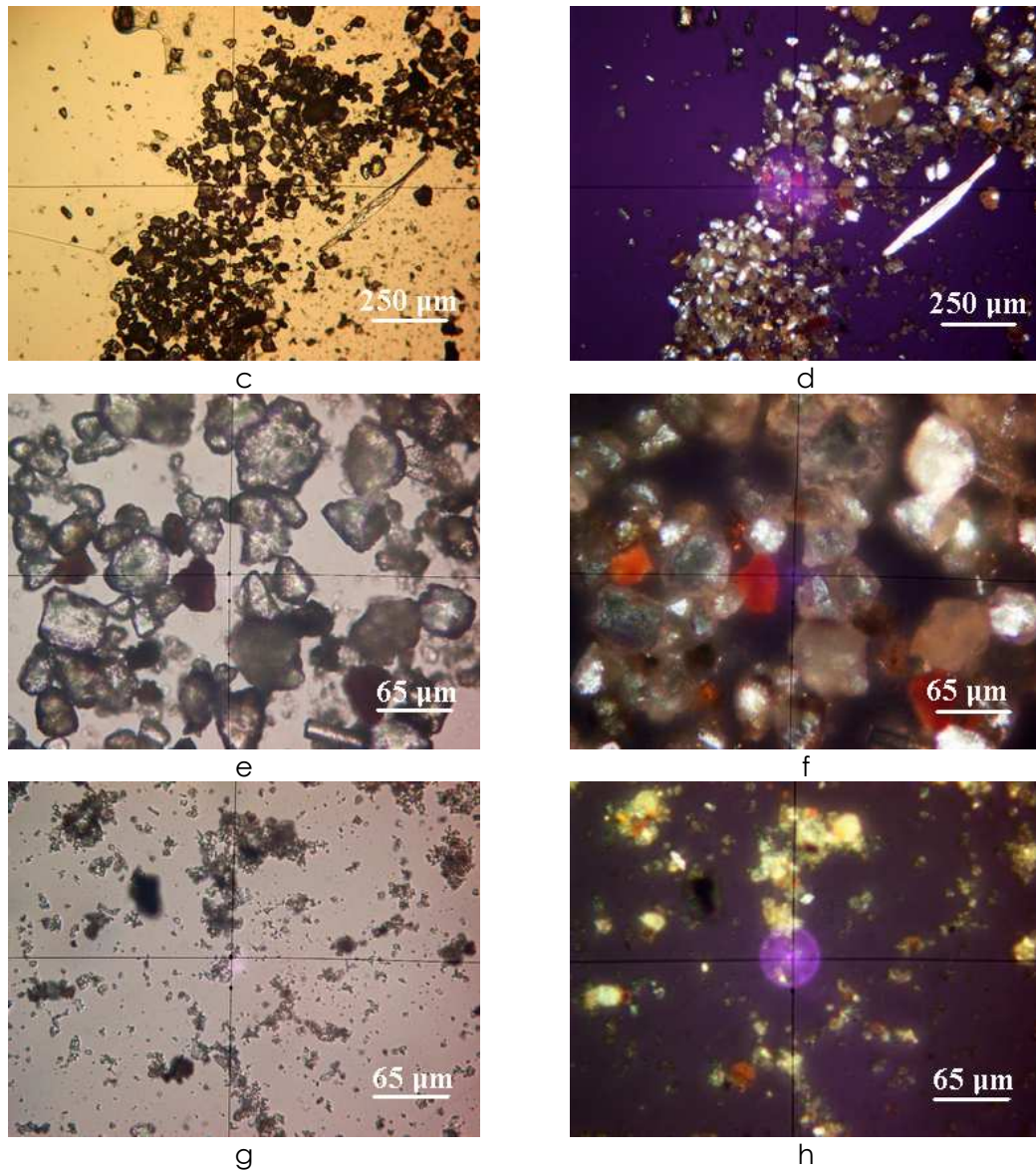


Figure 8. Microscopic aspects of P3 sample: a, c, e, g) micropictures with transmitted light; b, d, f, h) in cross-polarized light

In Figures 8c and 8d we observed a different overall microscopic zone, where various mineral particles, identified by X-ray diffraction, could be clearly distinguished.

The most interesting part of figure 8b is zoomed in figures 8e and 8f.

In these, we observed the microscopic dimensions of the particles, which confirm the previous findings, namely a mean diameter of about 60  $\mu\text{m}$ .

The polarized light results (Fig. 8f) are more interesting, as three brown-reddish lepidocrocite particles are surrounded by grey-greenish quartz particles. These facts are in full agreement with the results and observations yielded by X-ray diffraction analysis.

The structuring of the fine particles via polarized light is evident in figures 8g and 8h, where the fine quartz fragments are surrounded by smaller, white-yellowish caolinite particles. Also, brown fragments of lepidocrocite particles were observed.

This structuring of fine particles were subject to microscopic quantitative analysis in dark field (Fig. 9)

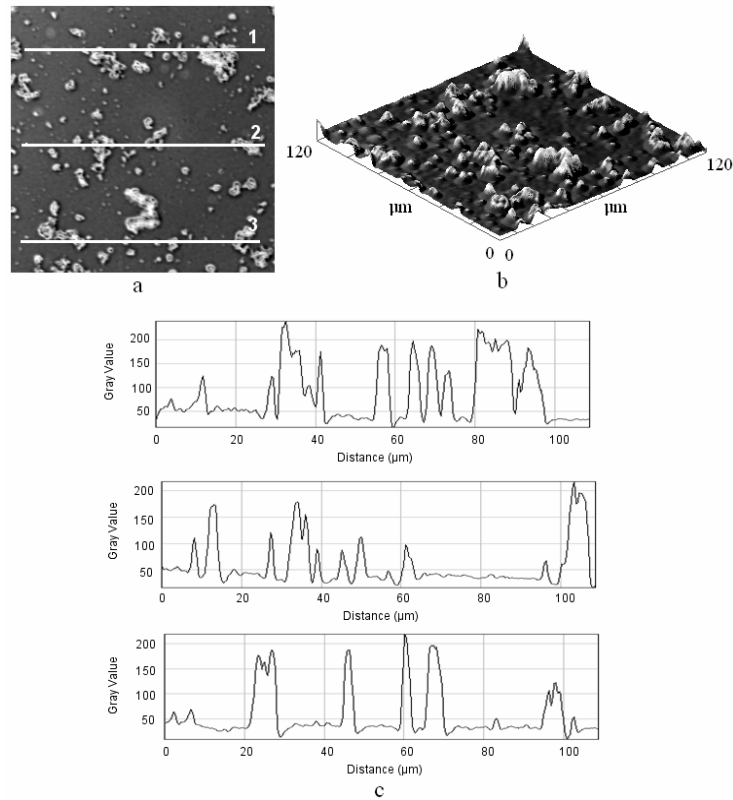


Figure 9. Microscopic quantitative analysis (dark field) for sample P3: a) the resulted micropicture, b) the 3-D presentation of figure 9a; c) profile details corresponding to the white lines in figure 9a

In the 3-D image (Fig. 9b) the accurate shape of the very fine particles is evident. The profile details in figure 6.40.c allowed the determination of mean diameters – hence, a mean diameter of approximately 3  $\mu\text{m}$  was determined for  $\text{PM}_{2.5}$ , whereas for  $\text{PM}_{10}$ , a mean value was found to be around 8  $\mu\text{m}$ .

Sample 4 – collected from Aurel Vlaicu Street, accumulated during the weekend of January 21-23, 2011, in an amount of 0.005 g, as deposited on crucible's wall.

The diffractogram for sample P4 is presented in Figure 10. It presents somewhat distinct diffraction maxima, characteristic of crystalline materials, but with more reduced

intensities and wider bands than in sample P3. This fact is indicative of the presence of a very fine powdered material.

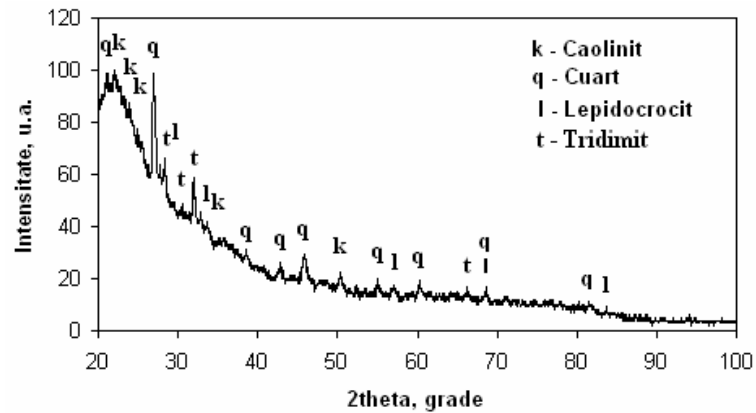


Figure 10. Sample P4 diffractogram – collected January 21-23, 2011,  
Aurel Vlaicu Street

We identified the same minerals that were present in sample P3, as follows: the predominant component is quartz, followed by lepidocrocite, caolinite, and tridimite. What differentiates P4 from P3 is the extent to which quartz exceeds the other components. The estimated quartz content in sample P4 is 70%, the rest being allocated the aforementioned minerals.

This assessment is confirmed by the overall microscopic observations, obtained under polarized light analysis, as shown in figure 6.42.d, where the bulk of the picture is almost entirely occupied by finely dispersed quartz, grey-greenish in color. Very few brown-reddish particles (lepidocrocite), or white-yellowish (caolinite) were observed, interspersed with the predominant quartz particles. Another overall microscopic view (Fig. 11a) reveals some larger equiaxial particles, of approximately 25  $\mu\text{m}$  mean diameter, but very few with respect to the finer particles association.

In polarized light (Fig. 11b), we observed that these correspond primarily to quartz, and a single particle matches tridimite (tabular, grey-greenish in color).

Long, fibrous materials appear in various colors under polarized light. These were attributed to colored textile fibers, and the observed colors were due to the fiber coloring pigments which exhibit characteristic colors when exposed to polarized light.

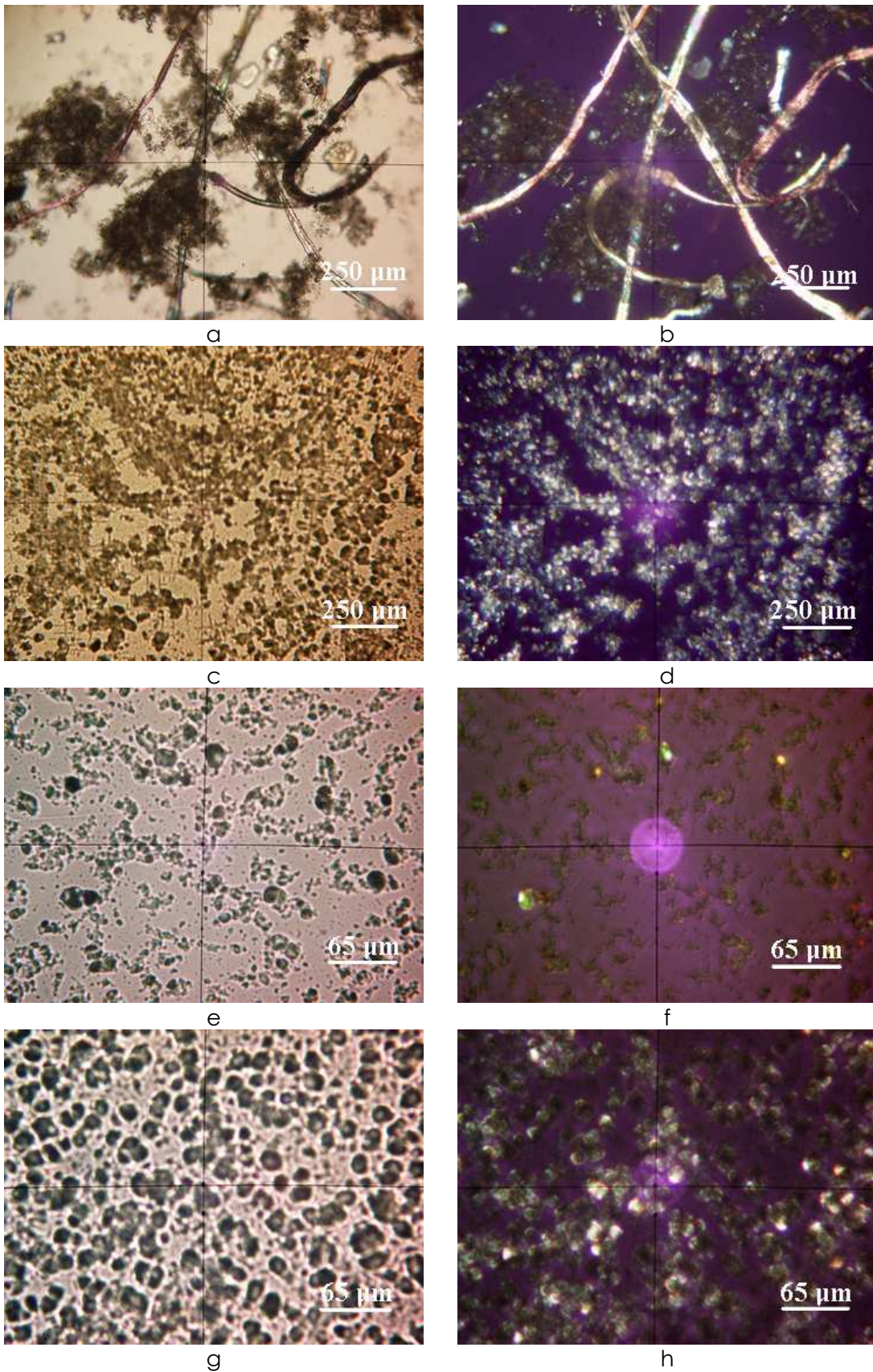


Figure 11. Microscopic aspects of P4 sample: a, c,e, g) micropictures with transmitted light;  
b, d, f, h) in cross-polarized light

The details of mineral optical microscopy are presented, in zoomed fashion, in figures 11 e-h. We observed fine equiaxial particles of PM<sub>10</sub> type, grey-greenish – which reveal that they belong into the quartz category. Fine PM<sub>2.5</sub>-type particles, accompanying the PM<sub>10</sub>-type, are even more evident in figure 11e.

In polarized light (Fig. 11 f), these PM<sub>2.5</sub>-type particles tend to exhibit white-yellowish color, consistent with caolinite. These results are in full accordance with the X-ray diffraction analytical results.

These particles were examined by dark field optical microscopy, to establish the dimensional characteristics. The microscopic quantitative analysis results are illustrated in figure 12.

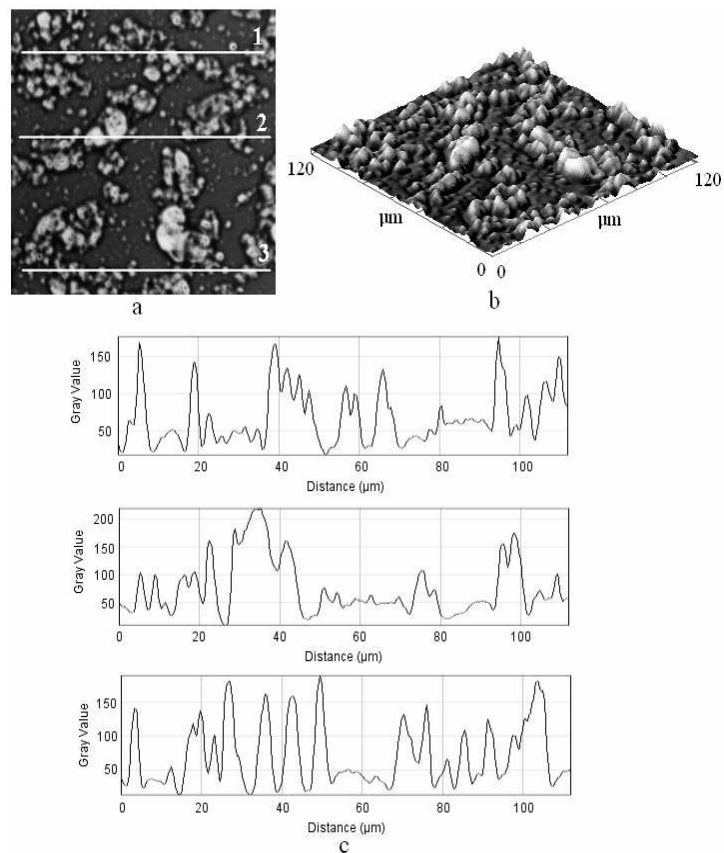


Fig. 12. Microscopic quantitative analysis (dark field) for sample P3: a) the resulted micropicture, b) the 3-D presentation of figure 12a; c) profile details corresponding to the white lines in figure 12a

The structural details yielded by the dark field optical micropicture shown in figure 12 a is better noticed in the 3-D presentation (Fig. 12b) and they are in full agreement with the observations obtained via transmitted light microscopy (Fig. 11e).

Based on the profile details in Figure 12 we established the mean diameter of the PM-type particles. Accordingly, the PM<sub>2.5</sub> have a mean diameter of 2.5 μm, and the PM<sub>10</sub> have an approximate mean diameter of 9 μm.

We also noticed that the fine particles represent a larger fraction of sample P4 than they did with respect to sample P3.

#### 4.2 The effects of atmospheric pollution with airborne pollutants on human health

We have demonstrated the fact that quartz - SiO<sub>2</sub> and lepidocrocite - FeO(OH) are predominant in PM<sub>10</sub>.

The iron hydroxides are very dangerous for the respiratory tract and lungs, causing acute inflammations [Harris, 1999; Muntean et al, 2012].

To evaluate the invasive capacity of PM<sub>10</sub> and PM<sub>2.5</sub>, and what the effects of airborne polluting particles upon human health, we performed a study, based on optical microscopy.

A volunteer was exposed, for the duration of one hour, to the effect of airborne particles, in the area of Dambovitei Street, during normal traffic conditions and volume. After the exposure, a sample of the mucous membrane was subjected to optical microscopy analysis.

The PM<sub>10</sub> and PM<sub>2.5</sub> particles are observed as black dots, surrounded by white blood cells (WBC) and several germs (G) (Fig. 13) [Muntean et al. 2012]

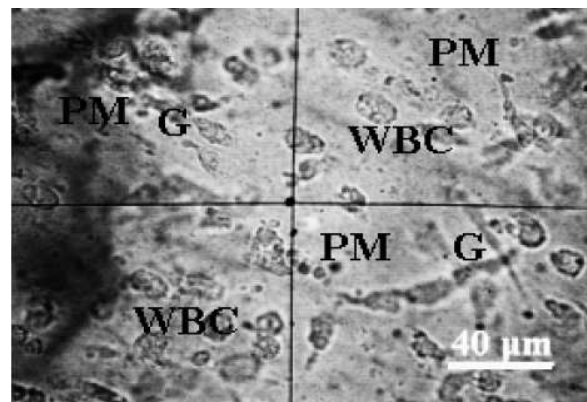


Fig. 13. Optical microscopy picture of a mucous membrane sample, exposes in vivo, for the duration of one hour, to the effect of airborne particles, in the area of Dambovitei Street, during normal traffic conditions and volume. PM = material particles, WBC = white blood cells, G = germs.

The above demonstrates that the presence of PM in mucous membrane induces a bodily reaction, which affects the human health.

#### 4.3 The effect of application of deicing and anti-skidding materials on roads, during winter season, upon the air quality in Cluj-Napoca

Due to our finding that the concentration of airborne particles including the PM<sub>10</sub> fraction during winter season, we performed a study regarding the effect of the application of deicing and anti-skidding materials in roads upon the air quality in Cluj-Napoca, from the standpoint of the presence of airborne particles.

In this context, we collected a representative sample of such deicing and anti-skidding material, from a typical lot used in Cluj-Napoca, on February 11, 2011. The sample consisted of a granular heterogeneous material, with variable diameters from 2 mm to 15 mm. The sample was prepared for the mineral investigation as follows:

- A representative amount of the overall material was mechanically powdered to evaluate its global mineral composition via optical microscopy.
- Several particles were separated, from each different component of the sample. Each component was mechanically powdered to evaluate its individual mineral composition via optical microscopy.

To identify the minerals contained in the sample of deicing and anti-skidding material, we applied X-Ray diffraction analysis.

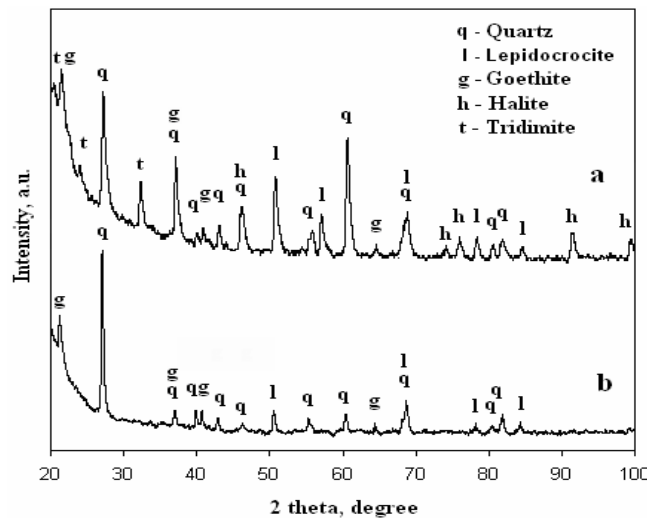


Fig. 14. The X-Ray spectrum of: a) the anti-skidding material and b) a sample with a high iron hydroxide content [Muntean et al, 2011]

The resulting X-ray spectrum, fig.14, reveals very well defined peaks, corresponding to a crystalline state of the probe.

We found large quantities of quartz –  $\alpha$   $\text{SiO}_2$ , lepidocrocite –  $\gamma$   $\text{FeO}(\text{OH})$ , goethite –  $\alpha$   $\text{FeO}(\text{OH})$ , halite –  $\text{NaCl}$ , and traces of tridimite –  $\gamma$   $\text{SiO}_2$ . [Arghir, Gherghari, 1989]

The morphology of the initial sample of anti-skidding material is presented in Fig. 15. The quartz particles appear as rounded, white-grey in color, halite appears as square, translucent particles, while the particles containing iron hydroxide are brown-red, [Catariu, Arghir, 1988], and the tridimite traces appear as rounded dark particles. Initially, the diameter of the global anti-skidding material particles was between 5-10 mm.

The quartz particles, and also tridimite are instrumental to increasing the road adherence, while halite reduces the melting point during winter season, under traffic conditions.

Optical microscopy analysis (Fig. 15 c and d) reveals the high content of lepidocrocite and goethite (brown-orange color), mixed with fine quartz particles (grey-greenish).

Most probably, the particles containing iron hydroxide originate from the rust from the vehicles' chassis, which mixes on the road surface with the deicing and anti-skidding material.

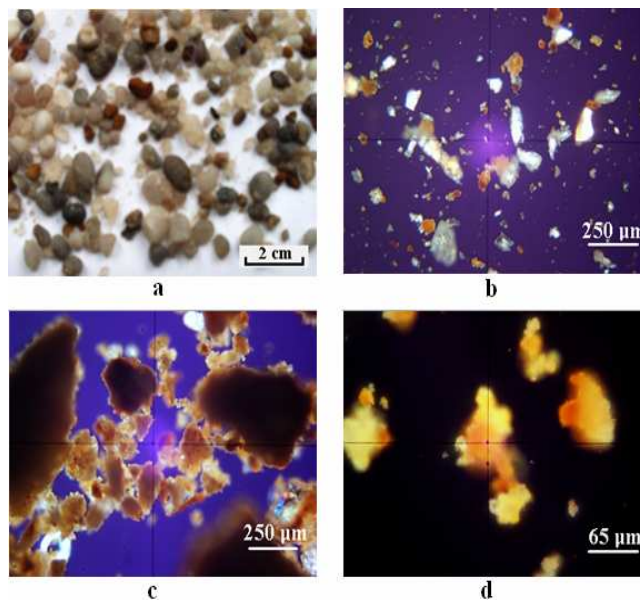


Fig. 15. Morphology and mineral distribution in primary samples: a) initial anti-skidding material; b) cross-polarized light micropicture of anti-skidding sample, c) cross-polarized light micropicture of anti-skidding sample with iron hydroxide content, and d) zoomed cross-polarized light micropicture of anti-skidding sample with iron hydroxide content [Muntean et al, 2011]

Due to the known hazards associated with airborne iron hydroxides, a procedure to reduce their emission is necessary. We proposed magnetic separation as a solution.

By applying dry magnetic separation, we achieved a high extraction efficiency, but the mixture of goethite and lepidocrocite is impurified by the presence of trace quartz and tridimite.

The extraction efficiency is beneficial for the environment, but the resulting product has no industrial applicability, due to the quartz contamination.

In order to explore a viable industrial solution, we applied wet magnetic separation, where the extraction efficiency was reduced, but the purity of the resulting product was significantly improved.

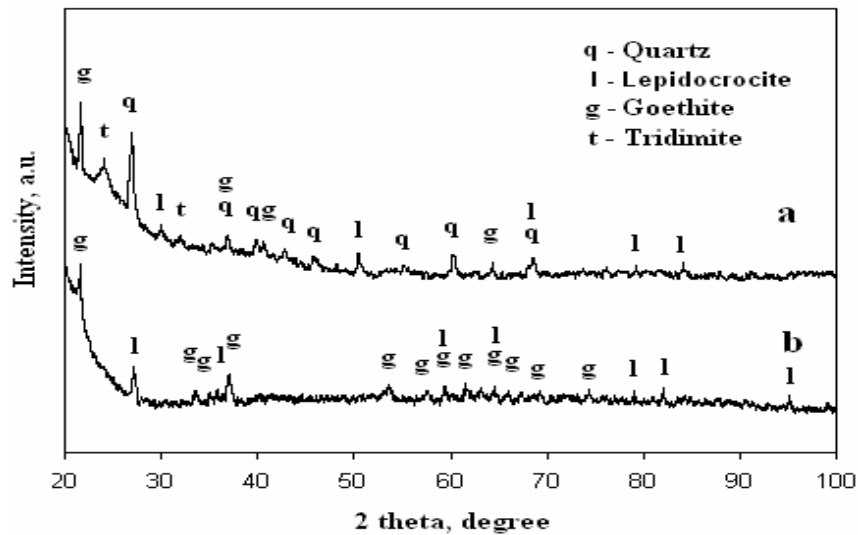


Fig. 16. The X-ray spectrum of particles of anti-skidding material with high iron hydroxide content after: a) dry magnetic separation and b) wet magnetic separation [Muntean et al, 2011]

It is observed that, following the wet magnetic separation procedure, goethite and lepidocrocite are the only detected minerals (Fig. 67).

The powdered iron hydroxide resulted by wet magnetic separation seems to have uniform dimensions, 15  $\mu\text{m}$  in diameter, which could be adequate for use in metallurgy, or, it could be subjected to hydrogen reduction to obtain a purity appropriate for iron powder.

The technical procedure of wet separation can be further improved, in order to use it as a source of raw material in metallurgy or other processes, such as the manufacturing of permanent magnets from synthesized barium ferrite [Catarig, Arghir, 1998; Arghir, 1978].

Our research aimed at iron hydroxide in the environment proved that the anti-skidding material samples collected during December 2010 – February 2011 contain significant amounts of iron hydroxides (goethite and lepidocrocite), amounting to approximately 20%. Their presence could be very hazardous to human health.

The applied magnetic separation method has proved beneficial for the environment, by enhancing its quality.

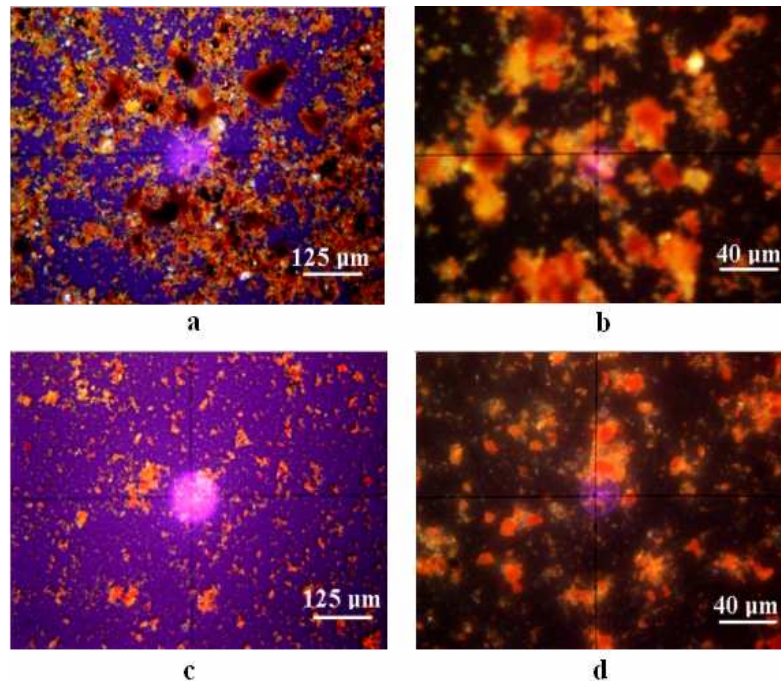


Fig. 17. Cross-polarized light micropictures of samples for magnetic separation: a) dry, medium zoom, b) dry, high zoom, c) wet – medium zoom, and d) wet – high zoom [Muntean et al, 2011]

The proposed separation method can be useful in the reduction of the iron hydroxide content in the urban ambient air.

#### 4.4 Concluding Remarks

The following aspects result from the analysis of the four samples of powders, subjected to mineral crystallographic analysis:

➤ All four samples have the same basic composition, having quartz as predominant, but accompanied by lepidocrocite, in variable proportion. In all samples we identified caolinite, as minority component. With the exception of sample P1, we detected tridimite in trace amounts. Since the points of sample collection (Dambovitei Street and Aurel Vlaicu

Street) are at least 1 km apart proves that the originating source of these particles has a widespread geographic area.

➤ For the samples collected during weekdays we noticed a large fraction of airborne particles with mean diameters between 25 and 60  $\mu\text{m}$ , for both collection points (Dambovitei Street and Aurel Vlaicu Street), corresponding to  $\text{PM}_{2.5}$  and  $\text{PM}_{10}$ -type particles. During weekends, we noticed a relative decrease of levels associated to these large particles. The fact is correlated with higher traffic intensity during weekdays, traffic which triggers the air dispersion of larger particles to a higher extent.

➤ Fine microscopic particles of  $\text{PM}_{2.5}$  and  $\text{PM}_{10}$  type are within specific dimensional range in all samples, regardless of the time and place of collection, with the observation that their size distribution is lower by 1-2  $\mu\text{m}$  during weekends.

➤ Quartz and lepidocrocite are prevalent in  $\text{PM}_{10}$  and the large microscopic particles, while caolinite corresponds rather to the fine fractions of  $\text{PM}_{2.5}$  particles.

It is interesting to note the quartz association with lepidocrocite in all four investigated samples. This association reveals the existence of a common source of such material particles, covering vast geographic areas, given the two relatively distant collection points (Dambovitei Street and Aurel Vlaicu Street).

We observed that the application of anti-skidding material, applied during winter season, is a problem, due to the harsh contact between the tires with the road surface, associated with the reduced hardness of such particles. Particles of  $\text{PM}_{10}$  type become airborne, along with a resuspension of road dust, both of which lead to an increase in the overall airborne concentration.

## **Chapter V. The distribution of main atmospheric pollutants in Cluj County**

Atmospheric emission of pollutants originating in various human and natural factors affects not only the air quality, but other environmental components – water, vegetation, soil – with consequences on ecosystems and human life quality. [Hancu, Marin, 2008; Coldea, 2002]

Diffusion (molecular or turbulent) takes place as a result of molecular exchange between the fluid layers (molecular diffusion), or a mass exchange in fractions (macroparticles) that fluctuate between layers (turbulent diffusion). Macrodiffusion (fluid

particles) takes also place during laminar movement of fluids, due to the different relative speed between the fluid layers. [Sandu et al, 2004]

A dispersion model is a mathematical simulation of physical and chemical phenomena in atmosphere, which uses data entries into the characteristic dispersion equations, resulting in concentration values for various receptors. [Olendrzynski, 2000]

Several steps were taken to map the atmospheric pollutants:

- ❖ We identified the air pollution sources in Cluj County
- ❖ We elaborated the annual inventory of atmospheric pollutants in Cluj County; based on this we obtained data about the quantities of emitted pollutants
- ❖ We measured the main indicators present in atmosphere:  $\text{NO}_x$ ,  $\text{NO}$ ,  $\text{NO}_2$ ,  $\text{SO}_2$ ,  $\text{CO}$ ,  $\text{O}_3$ ,  $\text{PM}_{10}$ ,  $\text{Pb}$ ,  $\text{Cd}$  and  $\text{Ni}$  from  $\text{PM}_{10}$ , and we monitored their concentrations
- ❖ We monitored the weather conditions, such as temperature, pressure, wind speed and direction, humidity, solar radiation, and precipitation amount in Cluj-Napoca.

By corroborating the data obtained from the emission inventory with weather information and the measurements for all the above indicators, we applied the Gaussian OML-multi dispersion model, and the results yielded the main pollutants distribution over the area of Cluj County.

The county-wide emission database includes three categories for the emission sources:

- ❖ Point sources (chimneys)
- ❖ Surface sources (fuels used for heating in all localities in Cluj County)
- ❖ Linear sources (traffic)

## 5.1. NO<sub>x</sub> Concentration

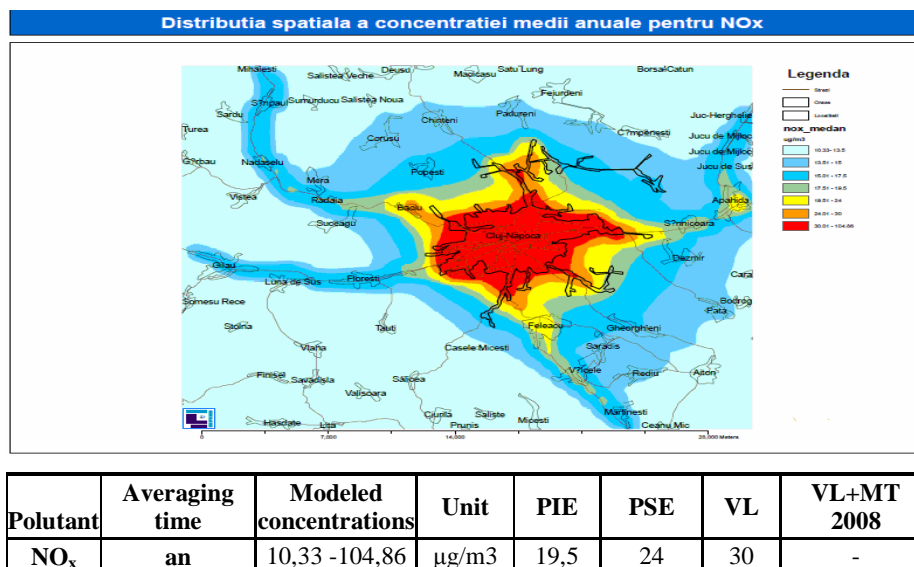


Fig. 18. Spatial distribution of annual average concentration of NO<sub>x</sub>

The limit of the medium annual concentration is exceeded across the entire area of Cluj-Napoca, particularly the entry and exit roadways (Fig. 18).

## 5.2. NO<sub>2</sub> Concentration

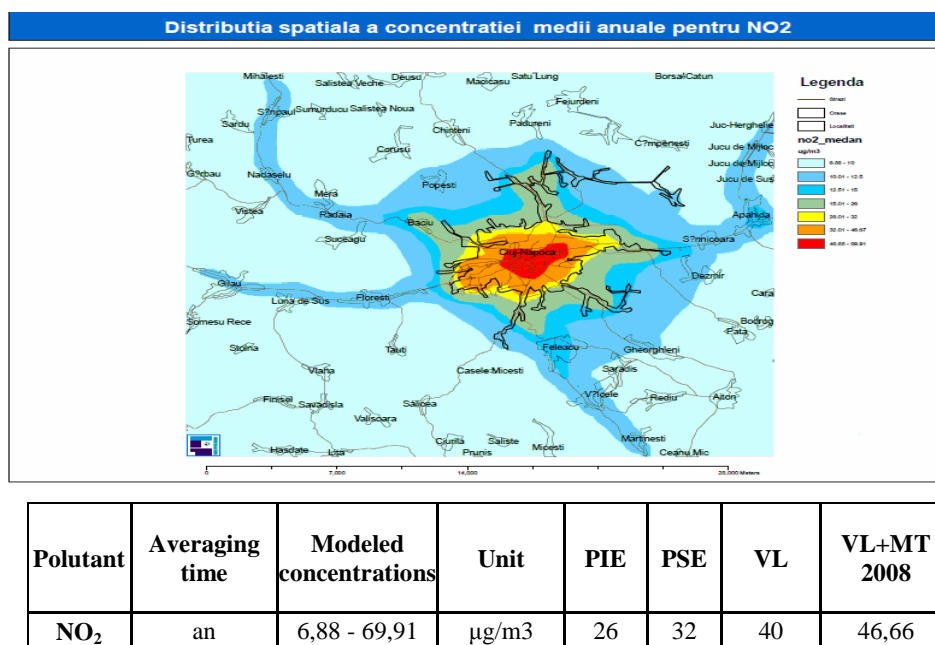


Fig. 19. Spatial distribution of annual average concentration of NO<sub>2</sub>

The limit (including the VL+MT tolerance) of the medium annual concentration is exceeded across the reduced area, in the central zone of Cluj-Napoca (Fig. 19).

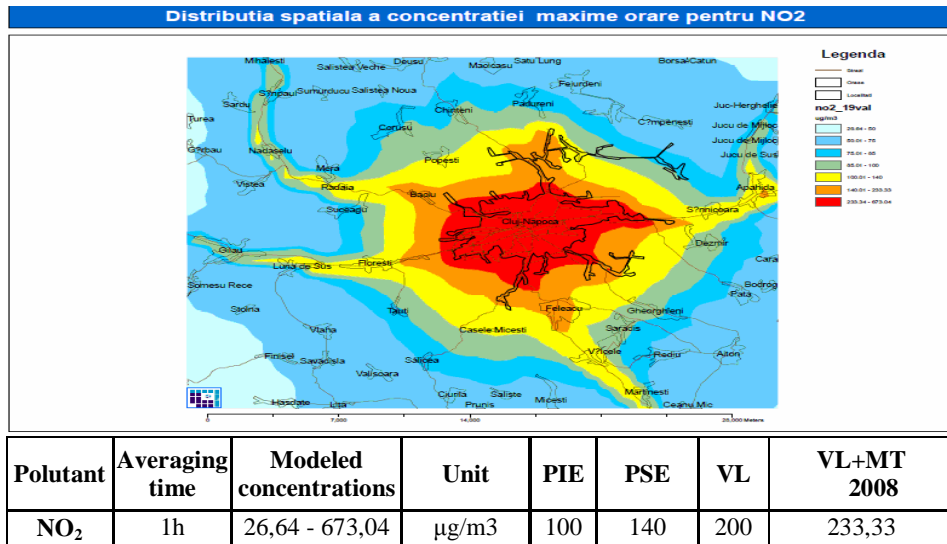


Fig. 20. Spatial distribution of maximum hourly concentration of NO<sub>2</sub>

The limit (including the VL+MT tolerance) concentration is exceeded almost across the entire Cluj-Napoca area (Fig. 20).

### 5.3. SO<sub>2</sub> Concentration

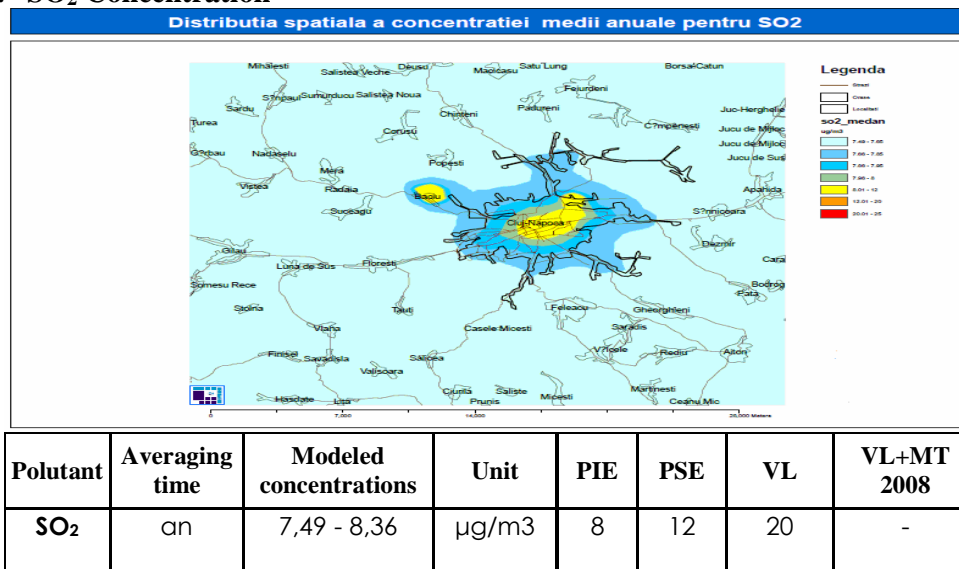


Fig. 21. Spatial distribution of annual average concentration of SO<sub>2</sub>

The limit of the medium annual concentration is not exceeded in Cluj County (Fig. 21).

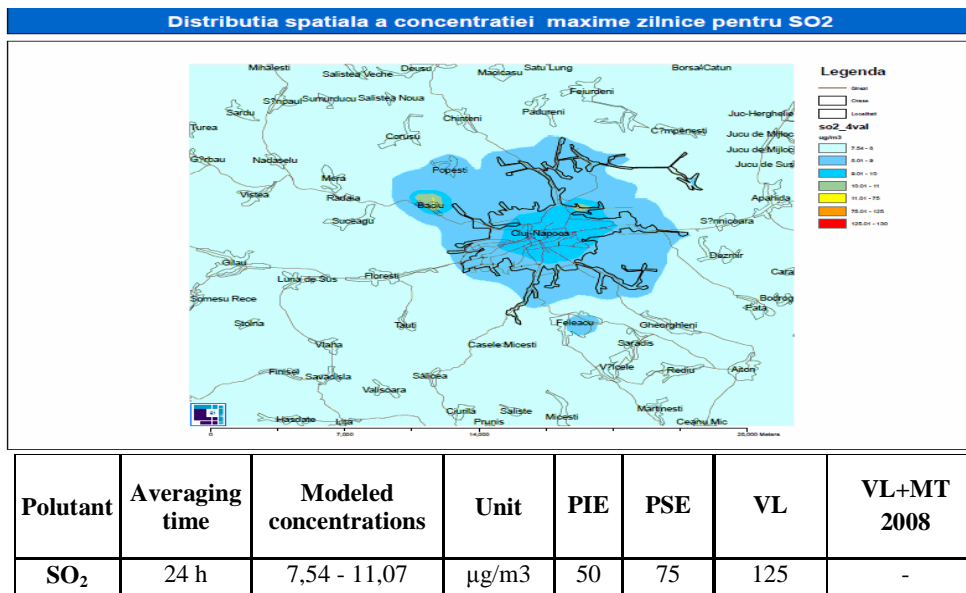


Fig. 22. Spatial distribution of maximum daily concentration of SO<sub>2</sub>

The limit of daily concentration, PSE and PIE, is not exceeded in Cluj County (Fig. 22).

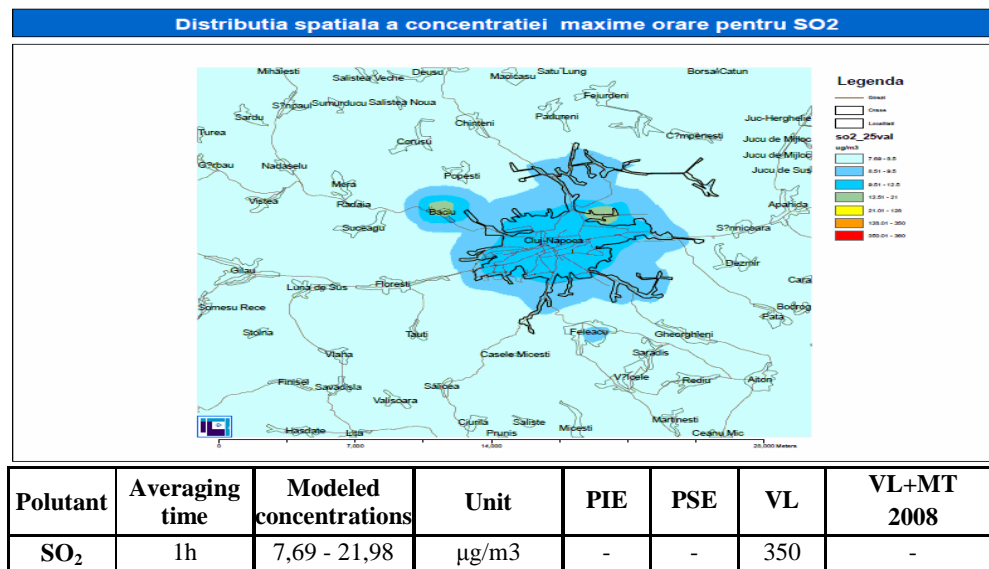


Fig. 23. Spatial distribution of maximum hourly concentration of SO<sub>2</sub>

The limit of daily concentration, PSE and PIE, is not exceeded in Cluj County (Fig. 23).

## 5.4. PM<sub>10</sub> Concentration

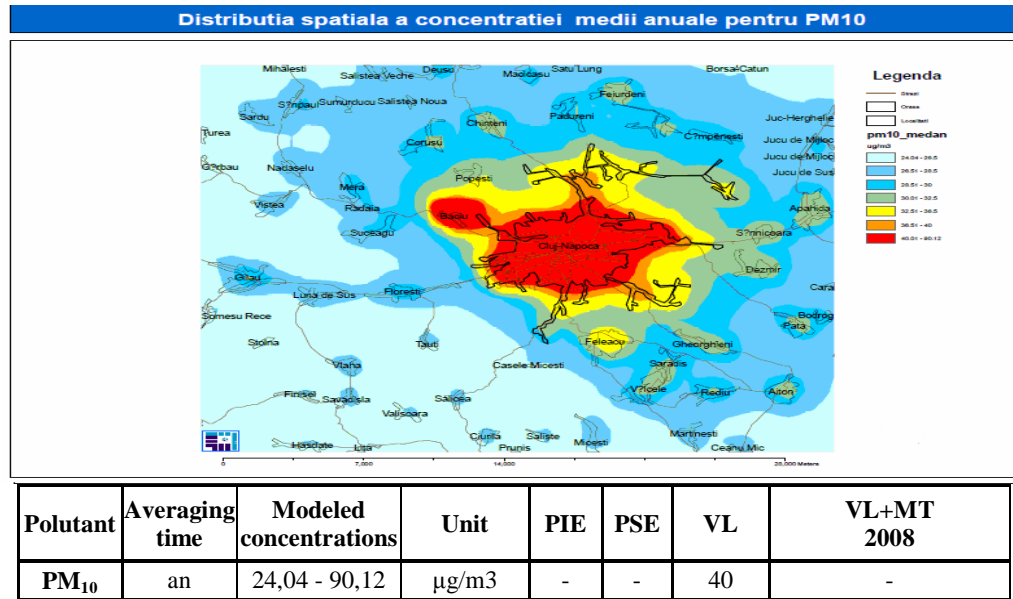


Fig. 24. Spatial distribution of annual average concentration of SO<sub>2</sub>

The limit of the medium annual concentration is exceeded in Cluj Napoca and Baciu village (Fig. 21).

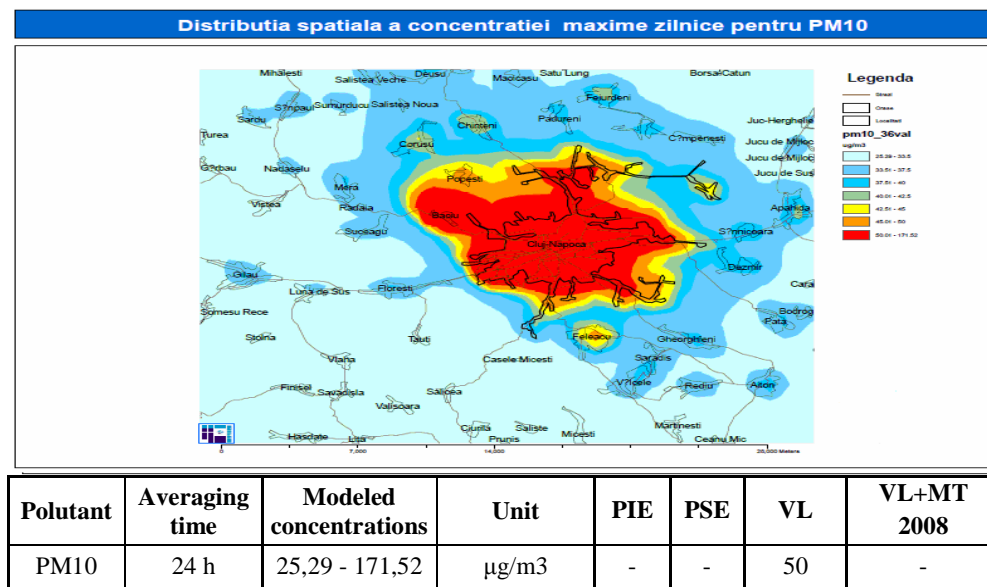


Fig. 25. Spatial distribution of daily maximum concentration of PM<sub>10</sub>

The limit of the medium daily concentration is exceeded across the entire area of Cluj Napoca, extended to suburban Baciu village (Fig. 25). These exceeded limits are mainly due to the heating sources of low height, associated to residential heating, heavy traffic, and construction zones in the area.

## 5.5. Pb Concentration

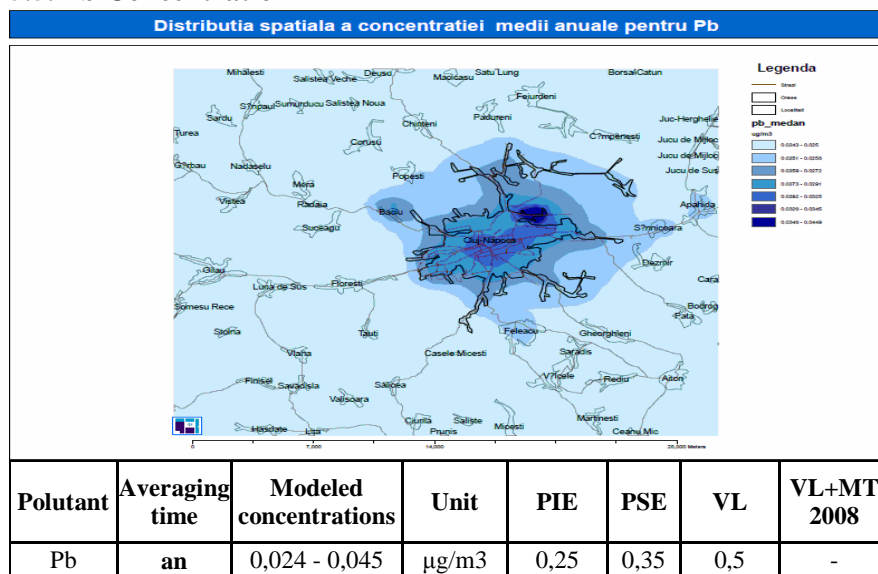


Fig. 26. Spatial distribution of annual average concentration of Pb

The limit of the medium annual concentration, PSE and PIE is not exceeded across Cluj County area (Fig. 26).

## 5.6. CO Concentration

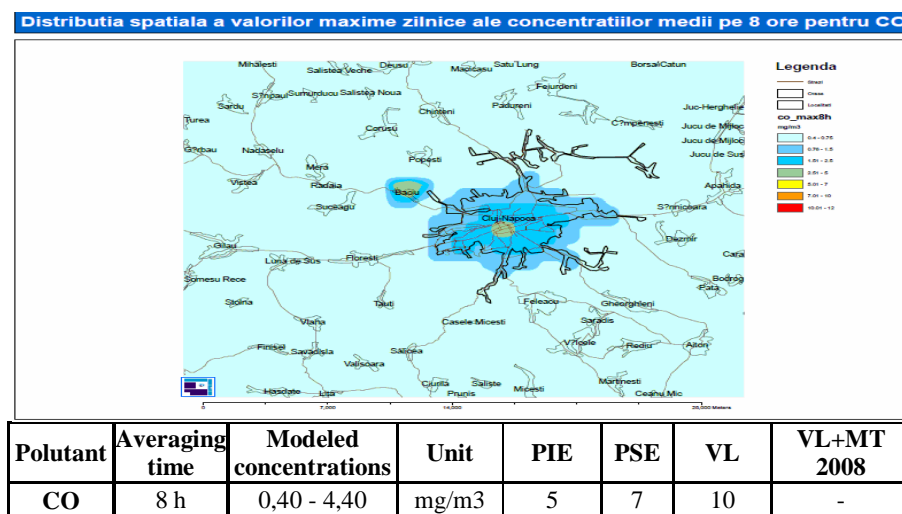


Fig. 27 Spatial distribution of maximum daily concentration (8-hour average) of CO

The limit of the maximum daily concentration (8 hours), PSE and PIE is not exceeded across Cluj County area (Fig. 27).

## 5.7. C<sub>6</sub>H<sub>6</sub> Concentration

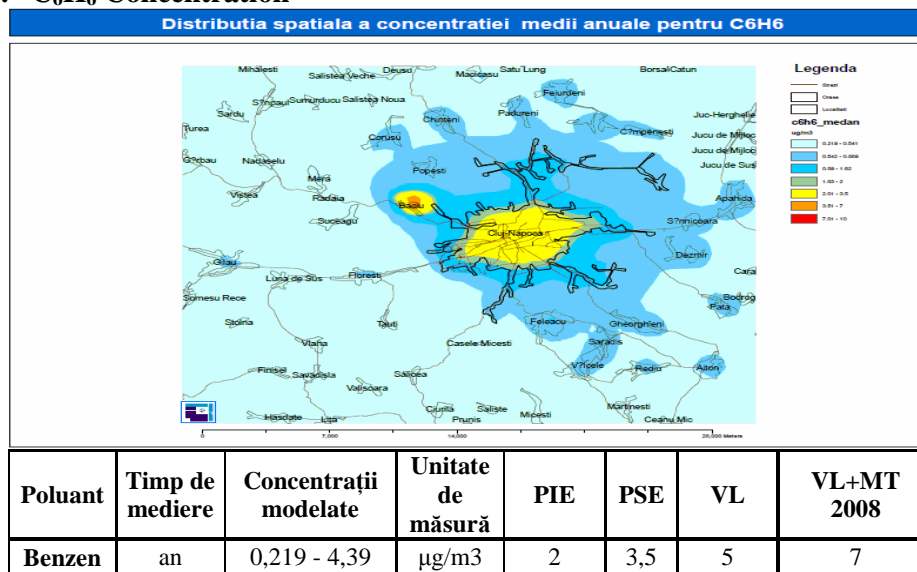


Fig. 28 Spatial distribution of annual average concentration of C<sub>6</sub>H<sub>6</sub>

The limit of the medium annual concentration is not exceeded across Cluj County area (Fig. 28).

## Chapter VI. Conclusions and personal contributions

Air pollution represents a great challenge of the recent decades, due in part to the aggressiveness of pollutants upon human health, but also to their impact on all environmental components – air, water, soil, vegetation.

Our studies, carried out in Cluj County during 2007-2011, revealed higher than limit values for the PM<sub>10</sub> indicator; this fact is due to:

- Road traffic (linked to the Diesel fuel consumption by trucks and cars)
- Construction zones, which increased in number during 2007-2009
- Application of deicing and non-skidding materials on roads during winter
- The fossil fuel burning facility in Cluj County (IMA), with installed capacity larger than 50 MW)
- Local sources, in increasing numbers, due to county-wide industrial development, during 2008-2009

These studies led to assessments with respect to air quality in Cluj County, and to conclusions aimed at measures aimed at reducing pollution in Cluj County, particularly PM<sub>10</sub>.

Such measures should aim at:

- Infrastructure redesign and modernization, county-wide
- Widening mass transportation options
- Larger parks and park areas and proper maintenance of such
- Assurance of the necessary parking lots and spaces
- Facilitation of bicycle use for daily activity
- Irrigation systems in the central city zones
- Enhancements to the road maintenance by:
  - Using non-polluting anti-skidding materials and mechanical road cleaning means
  - Regular cleaning of urban streets and sidewalks
  - Proper maintenance of parks and park areas, increasing the grassed areas around

roundabouts and roadsides

- Stricter criteria for the issuance of construction permits by:
  - Mandatory use of wet protection curtains, particularly in demolition activities, to prevent dust dispersion
  - Obligation of washing the vehicle tires upon exiting construction zones
  - Isolation of construction zones to restrict access/exit to specially designed ways
  - Tight coverage of vehicles transporting construction materials
  - Direct storage on containers of all materials from construction activities
  - Immediate removal of refuse and residual products by properly equipped vehicles

Numerous studies carried out within the European Union as well as Romania, emphasize the seriousness of air pollution upon human health, particularly the airborne particles. A series of conservative estimates, based on specialty studies, demonstrate that several adults and children under one year of age who die every year could be saved by imposing cleaner air norms.

It is very important that each and all of us to realize the importance of these facts and individually and collectively contribute to improving the air quality and implicitly, to a better health state for the population.

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