

Collection and analysis of particulate matter deposition around the Port of Koper

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ABSTRACT:

Particulates are tiny particles of solid or liquid suspended in a gas. Sources of particulate matter can be anthropogenic or natural. Increased levels of fine particles in the air are linked to health hazards such as heart disease, altered lung function and lung cancer. Human activities, such as burning of fossil fuels in vehicles, power plants and various industrial processes also generate significant amounts of particulates.

Results of epidemiological studies show an influence of polluted air on health status of inhabitants of Municipality of Koper. There are many different sources of air pollution in the region, mainly warehouse of petroleum products, chemical industry, waste incinerator, ironworks, Port of Koper, traffic pollution, elevated ground-level ozone in summer months, etc. The degree of each pollutant on total air pollution and health risk is difficult to establish. For this reason, a study was conducted in an effort to determine the extent of dust emissions from iron and coal ore manipulation at the Port of Koper on total particulate matter deposition in the surrounding area.

The results indicate that the Port of Koper has a partial impact on total dust emissions. During the sampling period from October 2005 to October 2006, the total amount of particulate matter deposition at all sampling sites in the area of the municipality of Ankaran and surroundings exceeded nine times the emission limit value of 350 mg/m² a day. The highest monthly values were measured in the period from 15. 9. 2006 to 15. 10. 2006. During this period, the monthly emission limit value was exceeded at nearly all of the sampling sites. A similar increase of particulate matter deposition values was seen also in the period from 15. 2. 2006 to 15. 3. 2006 and in the period from 15. 6. 2006 to 15. 7. 2006. New strategies with which the emissions of dust at Port of Koper will be reduced should be considered.

KEY WORDS:

Air Pollution, Particulate Matter Deposition, Iron Ore and Coal Dust Emissions

Received: 20. 2. 2009.

Accepted: 2. 6. 2009.

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INTRODUCTION

The quality of ambient air in residential districts is influenced mostly by air emissions of substances from nearby sources. The pollution results from the transport of these substances over large distances. Aerosols are small solid and liquid particles suspended in a gas phase. They originate as a result of processes on the surface of the earth and in the atmosphere. They vary in shape, size, density and chemical composition [1]. Primary particles in the air are the result of direct emissions from traffic, industry and combustion installations, agriculture, while secondary particles are the result of various physical-chemical processes in polluted atmosphere [1]. In addition to the anthropogenic sources, a portion of dust particles is also a result of natural processes in the environment (volcano eruptions, forest fires, wind erosion, plants pollen, etc.). Particles that arise from different processes have consequently a different chemical structure, shape and physical characteristics. Various factors influence the elevated concentration of pollutants, such as climatic characteristics, meteorological phenomena, physical-chemical processes of transformation of substances in the air and the topographical structure of the area [2].

Particulate matters can impact the health merely because of their presence, or because of a specific chemical composition or morphological structure that has additionally harmful influences on health. In most cases, the most susceptible organs are the lungs and the respiratory tract. Some air pollutants, such as fine particles, can also damage other organs. Coal particles belong by categorization of the International Agency for Research on Cancer (IARC) in group 3 – the agent is not classifiable as to its carcinogenicity to humans, which means that there were not enough studies made to confirm carcinogenicity, however it is not excluded [3]. Coal particles are, due to their chemical structure and their morphology, capable of binding with diverse pollutants, causing synergistic effects between various substances. Few if any studies regarding these phenomena have been reported. When a particle such as coal gets into the lungs, it reacts with the pulmonary cells, irritates them and releases the pollutants bound to it. Coal particles can cause allergic reactions in upper respiratory tract and lungs. That is why its toxicity is connected mainly to the size of the particles and the presence of other pollutants in the air. Kodavanti et al. [4] established a connection between metals in small particulate matters (particularly zinc) and the phenomenon of pulmonary inflammation in laboratory animals. Pulmonary inflammation in most cases resulted in the damage of the cardiac muscle. Authors concluded that the fine particles in outdoor air contain important concentrations of various metals, in most cases also an important concentration of zinc. Beside zinc in particulate matter deposition a considerable portion of other metals, such as lead and iron is present [5]. Iron oxide is a potentially dangerous substance because it is a catalyst for the reaction according to Fenton's chemistry [6]. In this reaction hydrogen peroxide, which is a normal metabolite in pulmonary cells in the presence of iron forms a hydroxyl radical, which is the most active and most aggressive free radical. Hydroxyl radicals directly

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The most exposed group are children, because the particles penetrate deeper in the lungs as compared to adults, they spend more time outdoor and are more active, subsequently they also breathe deeper and faster, the polluted air slows down the development of pulmonary functions in children.

One of the major sources of dust in the area of the Port of Koper is the activities at the EET. It is important to emphasize that the emission of particles results from the manipulation of ore (off- or on- loading).

destroy cells, degrade cellular membranes, proteins and cellular deoxyribonucleic acid (DNA) and cause mutations. In the legislation iron oxide is not yet declared as toxic, however the latest studies support the above mentioned mechanism of action [6,7]. Also in the case of iron oxide the size of the particles has the main role in determining its danger for health.

The presence of particles in the air is associated with asthma, chronic bronchitis and reduction of pulmonary function [5,8]. Various epidemiological studies [9,10,11] showed a connection between the elevated concentration of particles in the air and the increase of diseases of the respiratory tract and cardiovascular diseases, regardless of the chemical structure of the particulate matter. Exposure to particles in air can affect health, while elevated concentration of fine particle has a direct influence on morbidity and mortality [9,10,12,13]. The most exposed group are children, because the particles penetrate deeper in the lungs as compared to adults, they spend more time outdoor and are more active, subsequently they also breathe deeper and faster [14], the polluted air slows down the development of pulmonary functions in children [15,16]. Senior citizens, especially those with a weakened cardiovascular and respiratory system, are a high risk group too [5]. Another risk group is patients with chronic pulmonary emphysema, asthma or cardiovascular diseases [5].

Also in Slovenia there is a problem of elevated air pollution. Results of epidemiological studies show an influence of polluted air on health. Analysis of the study made by the Institute of Public Health Celje [17] on the health status of the residents in the Municipality of Koper show an increase of chronic respiratory diseases and allergic illnesses among children. Also an increase in the number of women with lung cancer, which is 1.6 times higher than the Slovenian average, a result of the pollution load on the discussed area, was detected.

The basic activities performed in the Port of Koper are cargo handling and warehousing. At the European Energy Terminal (EET) in the Port of Koper, large amounts of coal and iron ore are handled and warehoused. The storage capacity of the landfill is 450,000 t for coal ore and 350,000 t for iron ore. The area covered by the coal and iron ore is 108,500 m². Unloading daily capacity of coal is 17,000 t (cape size vessel) or 15,000 t (panamax vessel) and for iron ore (cape size vessel) is 25,000 t. One of the major sources of dust in the area of the Port of Koper is the activities at the EET. It is important to emphasize that the emission of particles results from the manipulation of ore (off- or on-loading). A big source of emission represents also the landfill, particularly during strong winds. Dust control measures already introduced include construction of an 11 m high anti-dust emission wall, spraying the body of the landfill with water and wet cleaning the transportation roads around the landfill at least twice per day. However, when a strong wind blows unexpectedly, the dusting is not suppressed completely. Because the landfill is not completely closed, it is still exposed to the weather conditions.

For many years the inhabitants of Ankaran (distance from the iron ore and coal storage sites is approximately 1800 m) and its surroundings (the closest residential area is 1000 m from the storage sites) have been pointing out the problem of the pollution of their residential area with emissions from the terminal EET in the Port of Koper. Particulate matter depositions are visibly perceived on their yards, places of residence, linen and vegetables (Figure 6). The results of the study of environment pollution in Ankaran [18] made by the Environment Agency of the Republic of Slovenia (ARSO) show the 24 hour limited value for particles PM_{10} was exceeded. With respect to wind direction, the distribution showed an increased concentration of particles during the mistral wind. Consequently, we can assume, regarding the location of the sampling site, a direct influence of the Port of Koper, specifically, the landfill EET. The same study ascertained a very strong correlation between the particles PM_{10} and the presence of iron. The correlation factor is 0.91.

The focus of the study was to collect and analyze the particulate matter deposition around the port of Koper. Standard and modified methods of particulate matter deposition collection were employed. Six sampling sites at different locations of Ankaran and its surrounding were established.

The basis of the Slovene legislation in the field of outdoor air quality is the Environmental Protection Act [25] and its subsequent Decrees. The particulate matter deposition and its limit values were regulated by the Decree on limit values, alert thresholds and critical emission values for substances into the atmosphere [26]. However, it was annulled on July 27th 2007 [27]. The change of the Slovene regulations is a consequence of the harmonization of the Slovene legislation with the EU one. The decree on sulfur dioxide, nitric oxides, particles and lead in the atmosphere [28], which regulates the area of emission of particles into the atmosphere, applies to only the supervision of particles PM_{10} and $PM_{2.5}$. It does not supervise particulate matter deposition.

METHODS

Location of sampling sites

The choice for sampling sites location on the micro level was based on the anticipated influence of emissions in the nearby area. Location of the measurement spot and influences on it by location (distance between the nearby buildings, trees etc.) was meant by the term micro location. The sampling gauge was placed 1.5 – 2.0 meters above the ground (the height of breathing of an adult person). In close proximity of the sampling sites were no obstacles that could have influence on the air flow passing the sampling gauge. The micro-location of individual sampling sites was well ventilated. The study included six sampling sites in the area of Ankaran and its surrounding (Figure 1). Some sites were chosen for their proximity to potential dust sources, and other sites were located downwind from a dust source. The first five sampling sites were placed in the prevailing direction of the wind or in the direc-

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Figure 1:

Geographical location of the sampling sites.



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The same method was applied also in the proposition of guidelines of the Association of clean air societies of Yugoslavia n. 201.

tion of the residential area, all in closed proximity to the Port of Koper. The sixth sampling place was used as a control sample of the background (located in the residential area of Hrvatini, where due to distance from Port of Koper and the height above sea level a negligible influence of the terminal of strewn cargo of the Port of Koper was expected).

The sampling design for this study was not statistically based; rather, sites were chosen to provide data on dust flux at the selected sites and to answer specific questions about the relation of port activities with the total dust deposition-emission regarding the distance from source, climate influence and other. Samples were collected and analyzed (gravimetrically) on the 15th day of each month for a one year period from October 2005 to October 2006.

Collection of particulate matter deposition

The collection of particulate matter deposition was conducted with the standard Bergerhoff sedimentators according to Commission Reinhaltung der Luft im VDI und DIN Guideline VDI 2119 part 2 (1996) – Measurement of Particulate Precipitations Determination of dust precipitation with collecting pots made of glass [19]. The same method was applied also in the proposition of guidelines of the Association of clean air societies of Yugoslavia n. 201 [20]. The stated method is used as the standard method for measuring the particulate matter deposition in Slovenia.

Modified method for collection of particulate matter deposition

The method for collection of particulate matter deposition is not perfect, because it does not entirely include the wind contribution since when wind blown through the sampling gauge, majority of particles are

not deposit on the ground or in the sampling gauge but blown away with the wind and precipitate when wind stop blowing or when hits the obstacle. That why metal screen as obstacle directed toward ore depot was added. That is the reason the method was modified by adding a metal screen (20 cm x 30 cm – sampling sites marked with letter B), which intercepted particulate matter spread horizontally. The flow chart of sampling sites is shown in Figure 2. Distillate water was also added to prevent the wind blowing away the particulate matter deposition from the containers. The main purpose of the modification was to establish and quantify the particulate matter brought also by the wind from the observed directions. Distilled water was added to improve the method.

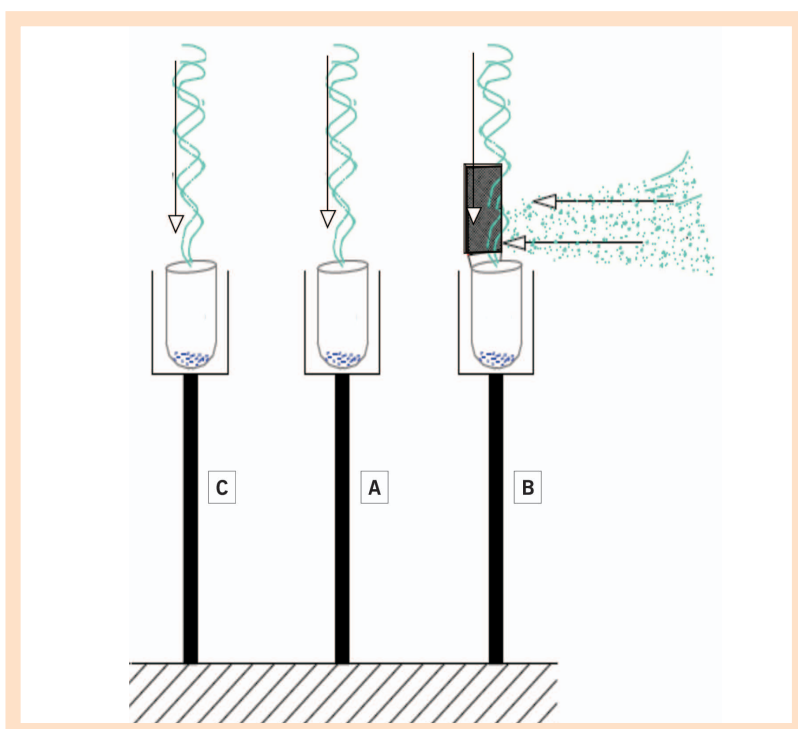


Figure 2:

Appearance of sampling site A, B and C

Three sample collectors were placed at each sampling site (Figure 3). The mass of material deposited in the first container, container A in Figure 2., was determined monthly. The mass of material deposited in the



Figure 3:

View from sampling site No. 5

The samples were analyzed by the gravimetric method and the amount of dust deposition was determined in mg/m^2 a day.

second container, container C, was meant to be determined annually. The mass of material deposited in the third container, container B, from both the horizontal and vertical directions was determined monthly. The yearly sample from container C was unusable because of the presence of decayed insects and plant leaves and contamination with algae. For this reason the yearly value of particulate matter deposition was calculated by the monthly values.

GRAVIMETRICAL ANALYSES

Each month, the glass collecting bottles from each sampling site were taken to the laboratory. When insects or parts of insects, parts of leaves, etc. were observed to be present, they were removed prior to the analysis. The samples were analyzed by the gravimetric method and the amount of dust deposition was determined in mg/m^2 a day. The samples were filtered through a cellulose nitrate membrane filter of porosity size of $3 \mu\text{m}$ (ME 29 Scheilcher and Schuell) using a vacuum filtering device. A modified procedure ISO 11923 [21] was used. Three blank filters were used prior to analyzing real samples. A part of filtrate was evaporated in an evaporating dish. Before weighting the samples and blank filters were thermostated for 60 minutes at $105 \text{ }^\circ\text{C} \pm 5 \text{ }^\circ\text{C}$ in both procedures (Binder FED 53, Tuttlingen, Germany). At the end filter paper and evaporating dishes were weighted and the total dust amount was calculated. An analytical balance, capable of weighing with a precision of at least $\pm 0,1 \text{ mg}$, was used (Mettler Toledo AT 261, USA).

Analysis of the amount of different metals

The depositions of aluminum, cadmium, chromium, copper, iron, lead, nickel and zinc were quantified as mg/m^2 a day by atomic absorption spectrometry of the material collected from the combined one year samples at each site (Shimatzu AA 6701 instrumentation). These determinations were performed according to methods EPA 7380 [22], EN ISO 15586 [23], ISO 8288 [24]. Samples were prepared for atomic absorption spectrometry by acid microwave-assisted digestion (Milestone 1200 Mega unit, Milestone Ethos plus, Rome, Italy). A multi-element Standard Reference Material SPS-WW2, Wastewater Level 2 (SPS 2002) was used for calibration and Quality Assurance/ Quality Control (QA/QC) procedures. The analysis was performed in the laboratory of the Public Health Institute of Koper.

Results and discussion

The monthly particulate matter deposition was collected from the 15th of each month for a one year period from October 2005 to October 2006. Figure 4 and Figure 5 represent the measured monthly emission of particulate matter at each sampling sites (in mg/m^2 a day). Vertical deposition is differentiated from vertical plus horizontal deposition as A and B, respectively.

During the sampling period from October 2005 to October 2006, the total amount of particulate matter deposition at all sampling sites in the

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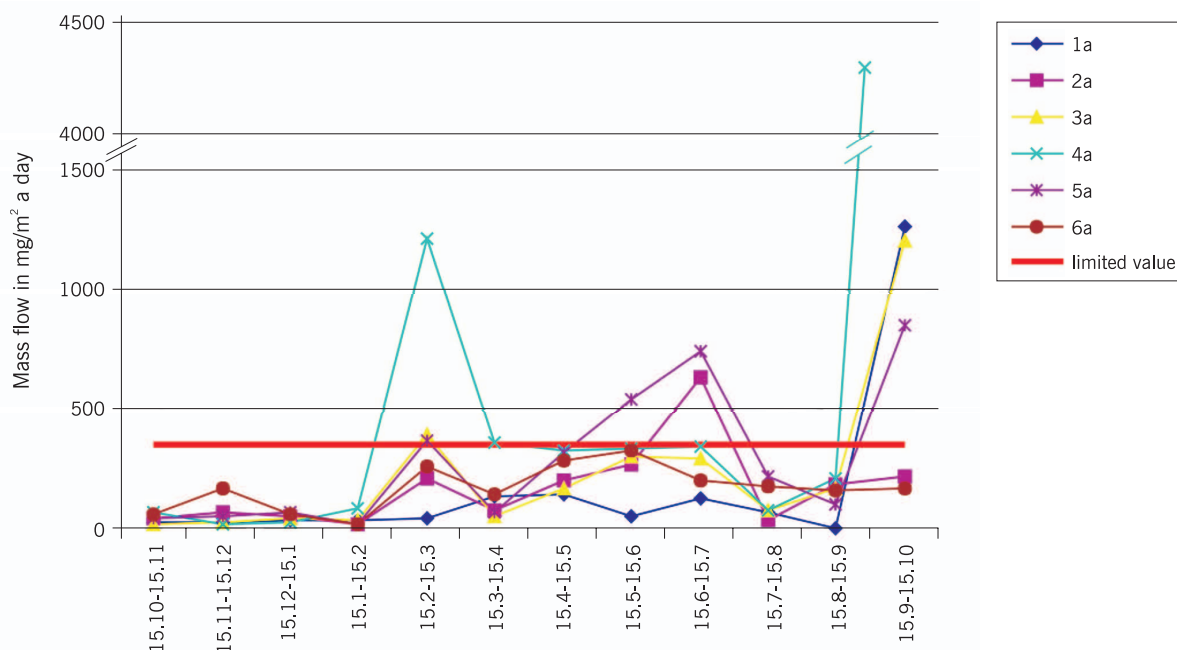


Figure 4:
Monthly mass of particulate matter
– vertical deposition (collected by
standardized method).

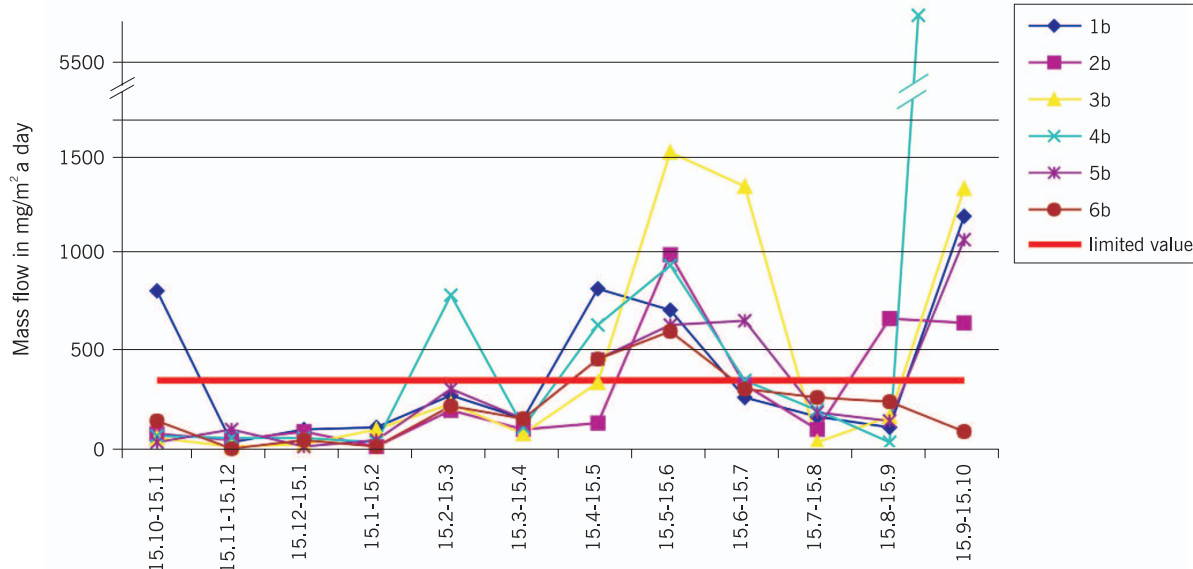


Figure 5:
Monthly mass of particulate matter
– vertical plus horizontal deposition
(collected by modified method).

area of the municipality of Ankaran and surroundings exceeded nine times the emission limit value of $350 \text{ mg/m}^2 \text{ a day}$. The highest monthly values were measured in the period from 15. 9. 2006 to 15. 10. 2006. During this period, the monthly emission limit value was exceeded at nearly all of the sampling sites. A similar increase of particulate matter deposition values was seen in the period from 15. 2. 2006 to 15. 3. 2006 and in the period from 15. 6. 2006 to 15. 7. 2006.

The highest values of dust deposition were measured particularly in the period 15. 9. 2006 to 15. 10. 2006. These could be attributed to strong wind, which blew from the direction of the sea across the ore storage pile toward the residential area for a short time (in a few hours). The consequences were reported also by the local residents as dust deposition seen on yards, gardens and facades. Figure 6 (a and b) were taken in the proximity of the sampling site 4 in October 2006, in the period when the values of particulate matter deposition was extremely high.

Figure 6:

Rožnik residential area (approx. 1000 m from landfill) – after strong wind, October 2006.

a) clean gauze and

b) gauze after wiping the floor.



The amount of the collected dust and its frequency was higher than that obtained with the standard method.

With the modification of the sampling device by adding a metal screen (20 cm x 30 cm) oriented towards the iron ore and coal storage piles, more dust was obtained in samples. The amount of the collected dust and its frequency was higher than that obtained with the standard method (Figure 4 and Figure 5). With this screen also the horizontal transport of the dust with wind could be obtained. However, with the addition of distilled water, the loss of dry dust sample from the glass by the action of strong wind was prevented.

The average particulate matter deposition per year (Figure 7) was calculated from the results of the monthly sampling intervals. Only sampling sites A (vertical deposition) were comparable with limited value since limiting value was set for standardize method. In any individual month as well as on an annual basis, the highest amounts were collected at the sampling site 4 – Rožnik, which lies approx. 1000 m north – north east from the European Energy Terminal storage piles. These results can be explained by the morphology of the terrain and the position of sampling site regarding the proximity of the coal and iron ore depot.

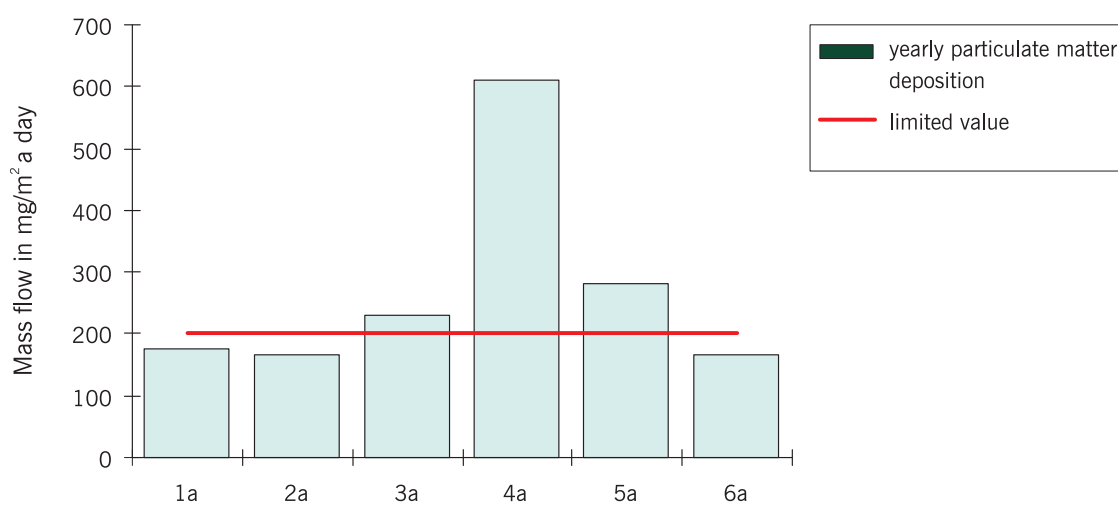


Figure 7:

Yearly particulate matter depositions on sampling sites A – vertical deposition.

The results from the determinations of aluminum, cadmium, chromium, copper, iron, nickel and zinc in the one – year samples of particulate matter show that the limit value of the metals Zn, Cd and Pb [26] were not exceeded. The only exception was the concentration of zinc at the sampling site 5 A. The current regulations do not specify limits for Al, Cr, Cu, Fe and Ni. Results also show increased values of Fe, Al, Cu, Zn and Pb at the sampling site 4. High values of metals in the deposition are present also on the control location (sampling site 6a). Sampling site 6 is located in the residential area Hrvatini, where influences from Trieste and its surroundings as well as those from the Gulf of Koper are possible. In comparison with the control (6a) it is possible to note the presence of particulate matter depositions also from other sources and not only as a consequence of the coal and iron ore depot from the Port of Koper.

It was not possible to distinguish the coal and iron ore originating at the Port of Koper from other organic sources of carbon (burning of fossil fuel, traffic, pollen, etc.) and iron compounds in the soil on the basis of the results from the metal determinations.

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Table 1:Values of metals in the yearly sample of particulate matter deposition ($\mu\text{g}/\text{m}^2$ a day).

	1a	2a	3a	4a	5a	6a	Limit value ($\mu\text{g}/\text{m}^2$ a day) [26]
Fe	3,054	7,740	14,433	29,450	6,892	13,081	/
Al	2,584	7,263	16,805	31,942	6,110	8,748	/
Cu	66	93	93	371	80	172	/
Zn	146	239	278	861	93	324	400
Cd	<0.3	<0.3	<0.3	1.2	<0.3	<0.3	2
Pb	25	35	40	87	24	60	100
Cr	27	39	90	98	28	46	/
Ni	14	26	35	50	14	42	/

/ – the limited value was not determined.

The results show significant differences between sampling sites and monthly and seasonal levels.

The results indicate periodical extremely elevated values of particulate matter deposition around the Port of Koper. According to gravimetric results the highest measured amounts of particles were observed during time period from 15. 9. 2006 to 15. 10. 2006 at sampling site 4. With standardize method of collection (sampling site A) 4,283 mg/m^2 a day was measured; this value exceeded the limited value (350 mg/m^2 a day) for the factor 12. Furthermore, on sampling site B, 5,743 mg/m^2 a day was measured, the limited value was exceeded for the factor 16.5, most probably due to wind contribution. During this time period also in all other sampling sites elevated values were observed. During entire study time period also two other elevations of particulate matter deposition were observed (15. 2.-15. 3. and 15. 6.-15. 7.); however, measured values were lower and limited value was not exceeded in all sampling sites. The results show significant differences between sampling sites and monthly and seasonal levels. Increased values are noticed in the hot dry periods, mainly in summer and autumn months due to the dryness of the terrain and strengthening of the wind, which blew from the sea across the storage piles of ore toward the sampling sites. During winter, a north wind, which blows across the storage piles towards the sea, prevails in the study area. Due to only one year sampling period more accurate conclusions can not be made due to unpredictability of weather and variations of climate. It is suggested that monitoring should continue for longer time period before accurate conclusion can be done.

Several conclusions can be drawn from this study. (1) Monthly monitoring intervals may be too large to correlate the influence of cargo manipulation on the quantity of the particulate matter deposition in the environment. (2) The off-loading, on-loading and storage of the iron ore has a small influence on the dustiness of the environment. (3) The main influence is the sudden strong wind, which blows from the sea crosses the storage piles and continues towards the residential area [29]. (4) The quantity of particulate matter depositions depends mostly on weather conditions, especially the strength and direction of the wind and also the quantity of rainfall, which determined the dryness of the

terrain and also dryness of coal and iron ore. Building the 11 m high anti-dust emission wall and the spraying of the landfill with water lowers the emissions of dust particles in the environment. Unfortunately no data on emission from the period before the anti-dust emission wall was build are available. Thus, it is not possible to evaluate how much this measure decreased the emission in the environment from coal and iron ore depot. However the Port of Koper is considering the adoption of additional new strategies with which the emissions of dust will be reduced even more in the future.

In the area of the municipality of Koper are various point and dispersed sources of emissions of various air pollutants. In this area there are located a large storage depot of petroleum products (emissions of benzene, toluene), chemical industry (emissions of formaldehyde, acetaldehyde), waste incinerator and the ironworks in Trieste (dioxins, furans, dust particles). Additionally heavy traffic pollution, ground-level ozone in summer months, transport of polluted air with the western winds from the Po river basin, etc is present. For these reasons, it is necessary to take measures to reduce the environmental burden with various pollutants from diverse sources. By no means should additional emissions into the environment, which would burden the air, be allowed.

The results indicate that Port of Koper has some impact on particulate matter emissions. For this reason further studies should be carried out to estimate the percentage ratio of particulates origin from the Port of Koper. For future work analyze of samples with electron microscopy will be performed to determined the shape, the size and also the chemical composition of the individual particle. Additionally the $^{13}\text{C}/^{12}\text{C}$ ratio in samples will be analysed. Results from both method will be used to determinate the origin of particulate matter. Additionally it would be reasonable to make risk assessment, specially focused on respiratory disease incidence. According to the results of epidemiological study [17] additional epidemiologic research on respiratory problems in the impact area would also be reasonable to be made.

Clean air is one of the necessities for healthy life now and in the future. Achieving this goal requires the active participation of legislators, experts from various fields and all those who burden the air with their activities.

Acknowledgements

Financial support to the study from Port of Koper is gratefully acknowledged.

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