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Emission Sources and Their Contributions to Ambient Air Concentrations of Pollutants

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1. Introduction

Understanding the chemical and physical processes in the atmosphere and emission sources of various technologies is the key to the development of cost-effective and health-protective air pollution control strategies. A number of species have been designated "hazardous air pollutants" or toxic air contaminants. Most are directly emitted into the air, but some also have significant secondary sources, i.e., are formed by chemical reactions in air. Furthermore, the ultimate health impacts are determined not only by the emission and formation of such compounds in air but also by their atmospheric fates. In short, some pollutants react in air to form less toxic species, whereas other form more toxic compounds (Barbara et al., 2000).

Today, no region of the global atmosphere is unaffected by anthropogenic pollution. Urban regions are affected by inorganic, light organic and heavy organic gases (Jacobson, 2002) and aerosols. Import heavy organics in urban air, such as toluene and xylenes, break down chemically over hours to a few days; thus, most of the free troposphere is not affected directly by these gases, although it is affected by their breakdown products.

Heating and cooling of the surface affect the stability of the atmosphere. The Earth's surface is a much more efficient absorber and emitter of radiation than the atmosphere above. During daytime, heating of the surface increases air temperatures close to the surface, resulting in an unstable atmosphere where air moves freely up and down. At sunset the land surface begins to cool, setting up stable conditions near the surface. Diurnal variation in atmospheric stability over land surfaces has important implication for urban air pollution; ventilation of cities tends to be suppressed at night and facilitated in the daytime (Jacob, 1999).

The mathematical models based on fundamentals of atmospheric chemistry and physics are essential tools in tracking emissions from many sources, their atmospheric transport and transformation, and finally their contribution to concentrations at a given location (receptor). The receptor models are enabling to attack the source contribution identification problem in reverse order, proceeding from particulate concentrations at the receptor site backward to responsible emission sources. The corresponding tools, named receptor models, attempt to relate measured concentration at a given site to their sources without reconstructing the dispersion patterns of the material (Seinfeld and Pandis, 1998).

2. Emission Sources in the Industrial Town Pančevo

Pančevo (20^0 40' N, 44^0 53' E) an industrial town of about 80,000 inhabitants is located about 20 km northeast of Belgrade. A major industrial complex includes petrochemical plant a fertilizer plant and a major oil refinery. Industries for refining oil and for the production of petroleum derivatives and fertilizer manufacture are situated in the South Industrial Zone of Pančevo (Fig. 1).

The dominant wind flows from the south-eastern segment, i.e. from the direction of the Southern Industrial Zone, towards the town of Pančevo. Industrial furnaces and oil refinery torches, which are emission sources of SO_2 and NO_x and other products of burning, are also situated within the South Industrial Zone.

Fig. 1. Map of Pančevo

The Vatrogasni dom receptor is located in the urban part of Pančevo - in the traffic area, at about 5 km from its industrial zone (Fig. 1 red point a). The "Vojlovica" receptor (Fig. 1, red point b) is at about 300 m from the nearest traffic artery, which minimizes the influence of traffic.

The factories for refining oil are emission sources of oil-type NMVOCs, including benzene, toluene, xylenes, as well as sulphur compounds, methyl-mercaptan and TRS (total reduced sulphur). NMVOCs are emitted from sources of technological processes for the production of petroleum derivatives. However, non-hermetic equipment and installations at the oil refinery and the petrochemical industry, from which NMVOCs can freely evaporate, contribute more significantly to the emissions. These are mainly non-hermetic oil- and oil derivative-tanks, dispatch facilities for petroleum products at the oil refinery, dispatch facilities for pyrolytic gasoline at the petrochemical industry and open-air waste water treatment plants at the oil refinery and petrochemical industry, where NMVOCs freely evaporate from the surface of the waste water. There is also a significant contribution of NMVOCs from traffic.

The fertilizer plant produces NH_3 using natural gas, which is first desuphurized with ZnO and subsequently, after mixing with water vapour, reformed and methanized to give a mixture with following composition: CO and CO_2, CH_4, H_2, N_2 and Ar. This gas mixture is compressed for the synthesis of NH_3 from the H_2 and N_2. One part of the recycled gas is driven to the reforming section, as a fuel, to keep the partial pressures of N_2 and H_2 constant. The production of NH_3 and its use in the manufacture of synthetic fertilizers is an only one emission source of NH_3 to the ambient air of Pančevo, while the combustion of the recycled gas is one of emission source of NO_x, besides traffic and other industrial furnaces.

2.1 The Contents of Specific Pollutants in the Ambient Air of Pančevo

In this study the results of continuous monitoring obtained from the municipal monitoring system of the industrial town Pančevo were used. The air pollutants in the urban area of the industrial city Pančevo was continuously monitored for one month (August, 2004). The hourly concentration of ammonia, Total Reduced Sulphur (TRS), NO_2, SO_2, PM_{10}, and hydrocarbons recorded minute-by-minute fluctuations were used.

For example, in August 2004, the average fuel consumption of the energy plant of the petrochemical industry, which produces steam for the industrial processes, was: 135 538 Nm^3 day^{-1} of natural gas, 70 t day^{-1} of fuel oil and 42 t day^{-1} of pyrolytic oil. The content of H_2S in the natural gas was between 0.86 and 5.40 mg l^{-1}, the content of sulphur in the fuel oil was 2.3 % and in the pyrolytic oil up to 4.5%. Assuming that combustion at the energy plant was complete, and that oxidation of the reduced forms of sulphur was complete, the maximum possible emission of SO_2 from high altitude emitters at this industrial firebox alone was approximately up to 350 kg h^{-1}, in August 2004. The emission sources of high altitudes originating from the combustion of liquid and gaseous fuels in industrial furnaces are expected to be the dominant contributors to the SO_2 and NO_2 in the ambient air but, near ground concentrations of these pollutants were low.

Routine measurements for several years showed that the NO_2 emission from the fertilizer plant was approximately 290 kg h^{-1}. However, NH_3 was also emitted, but the amount was not precisely quantified, either from technological balance or by measurements.

			Hourly			
			Min	Max	Mean	1σ
Vatrogasni dom		SO$_2$	10.8	143.1	19.3	10.2
		Benzene	0.1	160.5	18.0	25.3
		Toluene	0.1	180.0	18.8	22.9
		Me-me	0.2	20.0	2.3	3.1
		NH$_3$	0.3	401.5	10.8	26.2
		NO$_2$	1.5	159.3	17.0	14.3
		NMVOCs	0.6	497.8	75.4	110.7
Vojlovica		SO$_2$	0.7	63.2	5.5	4.5
		Benzene	0.4	150.2	12.5	22.5
		Toluene	0.7	184.7	15.9	27.8
		Xylenes	0.1	114.7	5.2	10.9
		TRS	0.7	12.3	2.7	1.8
		PM10	0.1	232.9	50.7	35.0

Table 1. The concentrations of the measured pollutants at the receptors Vatrogasni dom and Vojlovica in August 2004

Sulphur dioxide and nitrogen oxides as a combustion products, are emitting from industrial furnaces but it also originate from traffic. The high altitude industrial stacks in the Industrial Zone emit combustion products continuously, but traffic too.

2.2 Daily Variation of the Ambient Air Concentration

Based on the analysis of the mean values and standard deviations it is noticeable that no statistically significant differences existed in the concentrations of PM$_{10}$ at Vojlovica receptor; 48.6 ± 35.8 μgm^{-3} and 49.6 ± 34.2 μgm^{-3} during the night and daytime periods as well as SO$_2$ during the day; 19.4 ± 10.5 μgm^{-3} and night; 19.4 ± 9.1 μgm^{-3} (Fig. 1 and Tab. 1).

Statistically significant differences between the concentrations during day and night times periods are noticeable in the cases of benzene; 11.5 ± 14.5 μgm^{-3} and 22.8 ± 29.2 μgm^{-3} at Vatrogasni dom receptor; and 7.4 ± 14.8 μgm^{-3} and 17.0 ± 25.4 μgm^{-3} at Vojlovica receptor; respectively.

Statistically significant differences between the concentrations during day and night time periods are noticeable in the cases of toluene too; 11.6 ± 14.5 μgm^{-3} and 22.8 ± 29.2 μgm^{-3} at Vatrogasni dom receptor, and 8.5 ± 15.8 μgm^{-3} and 22.8 ± 32.6 μgm^{-3} at Vojlovica, xylenes; 2.5 ± 4.6 μgm^{-3} and 7.8 ± 13.4 μgm^{-3} as well as total reduced sulphur (TRS); 2.1 ± 1.3 μgm^{-3} and 3.3 ± 1.9 μgm^{-3} at Vojlovica receptor respectively.

The mean concentrations of NH$_3$ were higher during the daytime (12.3 ± 31.3 μgm^{-3}) than those during the night period (7.5 ± 11.6 μgm^{-3}) at Vatrogasni dom receptor, while the mean of daily concentrations of NO$_2$ during the daytime (12.9 ± 14.8 μgm^{-3}) were statistically significantly lower than those during the night (20.1 ± 11.5 μgm^{-3}) at Vatrogasni dom receptor, unlike the daily variations of the concentrations measured in the ambient air of the Kaohsiung Petroleum Refinery in Taiwan (Chiu et al., 2005a; Chiu et al., 2005b). The results showed that the concentrations of pollutants originating from low altitude emission sources like organic pollutants were higher twice and more at night. However, SO$_2$ and NO$_2$ are also emitted from a near-ground source – traffic.

Statistically significant differences (mean $\pm$ 1σ) between the concentrations during day and night time periods are noticeable in the cases of benzene (7.4 $\pm$ 14.8 μg m^{-3} and 17.0 $\pm$ 25.4 μg m^{-3}), toluene (8.5 $\pm$ 15.8 μg m^{-3} and 22.8 $\pm$ 32.6 μg m^{-3}), xylenes (2.5 $\pm$ 4.6 μg m^{-3} and 7.8 $\pm$ 13.4 μg m^{-3}) and TRS (2.1 $\pm$ 1.3 μg m^{-3} and 3.3 $\pm$ 1.9 μg m^{-3}). Thus, the concentrations of these pollutants were higher during the night than during the daytime period, which is in accordance with the results of measurements in the ambient air of the Kaohsiung Petroleum Refinery in Taiwan (Chiu et al., 2005a; Chiu et al., 2005b). These pollutants were emitted from low altitude sources located at the oil refinery and petrochemical industry. Low altitude sources combined with surface temperature inversion of the atmosphere represent extremely favourable conditions for pollution of the near ground atmosphere. An investigation of the frequency of surface inversion in the Belgrade region showed that average incidence of night time inversions was the highest (as many as 26 cases) in August (Vukmirović et al., 2003).

When the boundary layer of surface inversion moves towards the ground, the combustion products inside a plume of smoke emitted from high altitude industrial stacks spreads above the boundary layer and therefore near ground concentrations of these pollutants is lower. This phenomenon is more pronounced at night and as a result, the concentrations in the near-surface atmosphere are lower at night. The upper edge of a boundary layer is a natural barrier to the vertical dispersion of pollutants by diffusion.

The primary particles (Seinfeld and Pandis, 1998) are emitted from industrial furnaces and traffic, but they also can be formed as the products of atmospheric reactions between NH_3, emitted from the fertilizer factory, and acid oxides (SO_2 and NO_2), emitted from industrial furnaces and traffic, whereby in observed ambient conditions are enabled $(NH_4)_2SO_4$ and $(NH_4)NO_3$ aerosol production in the neutralisation processes (Olszyna et al., 2005). In atmospheric reactions, through SOA mechanisms, atmospheric aerosols are formed from organic air pollutants (Hamilton et al., 2005; Jenkin and Clemitshaw 2000; Knippinga et al., 2004 etc.).

2.3 Statistical Cluster Analysis with a View to Source Identification

In order to identify the main contribution sources of some pollutants in the receptor region (sampling site), the most commonly implemented subset of clustering method, which is generally referred to as an agglomerative hierarchical method, was employed (Facchinelli et al., 2001; Đorđević et al., 2004; Lee et al., 2004). This method is appropriate to evidence correlations between variables. The parameter dendrogram based on Pearson's correlation coefficients is summarized in the dendrogram shown in Fig. 2. The smaller the value on the axis is, the more significant is the association.

Hierarchical cluster analysis was used to obtain dendrograms (Fig. 2) established using average linkage between groups according to the Pearson correlation of combinations of pollutants.

By cluster analysis of output data of measurements, the clusters of benzene (cluster 2) and NMVOCs (cluster 7) variables from the »Vatrogasni dom« are related to each other with the highest coefficient of correlation ($r_{Benzene - THMNC}$ = 0.899) in the first stage of combinations in the day time, representing the strongest association (Fig 2a, Tab. 2). The second strong association is between toluene (10) and me-me (11) meaesured in the night time with the correlation coefficien $r_{Toluene-me-me}$ = 0.887. The next strong association is between benzene (9) and NMVOCs (14) measured in the night time, with $r_{Benzene-NMVOC}$ = 0.870. In the fourth

stage of agglomeration appears the association of toluene (3) and me-me (4) measured in day time with $r_{Toluene -me-me} = 0.864$ followed by association between benzene (9) and toluene (10) measured in the night time related to $r_{Benzene-Toluene} = 0.851$. The association between benzene (2) and toluene (3) ,measured in the day time, is related to $r_{Benzene-Toluene} = 0.831$. NH_3 (5) and NO_2 (6) ,that are agglomerated in the seventh stage, are bound with $r_{Benzene-Toluene} = 0.803$.

Vatrogasni dom (a)				Vojlovica (b)			
Stage	Cluster combined		Correlation coefficient	Stage	Cluster combined		Correlation coefficient
	Cluster 1	Cluster 2			Cluster 1	Cluster 2	
1	2	7	0.899	1	8	9	0.975
2	10	11	0.887	2	2	3	0.965
3	9	14	0.870	3	2	4	0.929
4	3	4	0.864	4	8	10	0.854
5	9	10	0.851	5	2	5	0.592
6	2	3	0.831	6	8	11	0.513
7	5	6	0.803	7	2	6	0.414
8	2	5	0.523	8	8	12	0.300
9	8	13	0.523	9	2	8	0.179
10	9	12	0.410	10	1	7	0.161
11	1	2	0.396	11	1	2	0.103
12	8	9	0.359				
13	1	8	0.265				

Table 2. Agglomeration Schedule

It is important to highlight that the clusters of benzene (2) and the NH_3 (5) measured in the day time, and the clusters of benzene (9) and NH_3 (12) measured in the night time are bound in both cases. Even the coefficient of correlation between the clusters of benzene and the NH_3 measured in the night time is lower ($r_{Benzene-NH3} = 0.410$). These parameters are linked in considerable correlation, this fact indicating the sources of benzene in the industrial zone beacuse the NH_3 originatig from just one sorce located in the »Azotara« Fertilizer plant . On the other hand the smallest values on the axis (Fig. 2) are agglomearated for benzene, toluene, NMVOCs and me-me measured in the day time, and for the same variables in the night time on the »Vatrogasni dom« receptor, indicating most considerable associations. Ammonia and nitrogen dioxide form a minor association , that becomes considerable in the day time and shows the influence of the »Azotara» Fertilizer plant .

As far as the results obtained on Vojlovica receptor are concerned, the associations are similar (Fig. 2b, Tab. 2). The most considerable and strongest associations are between benzene, toluene, xylenes, regardless of whether in the day time or in the night time.

The results of the cluster analysis show the existence of common strong emission sources having the effect of evaporation, because benzene, toluene and xylenes are within the scope of petrogenic hydrocarbons from the Oil Refinery and Petrochemical plants, but also in the scope of NMVOCs from the traffic. The prevailing winds in the region come from the south east direction. These winds from the South Industrial Zone carry pollutants to the town Pančevo (Fig. 1).
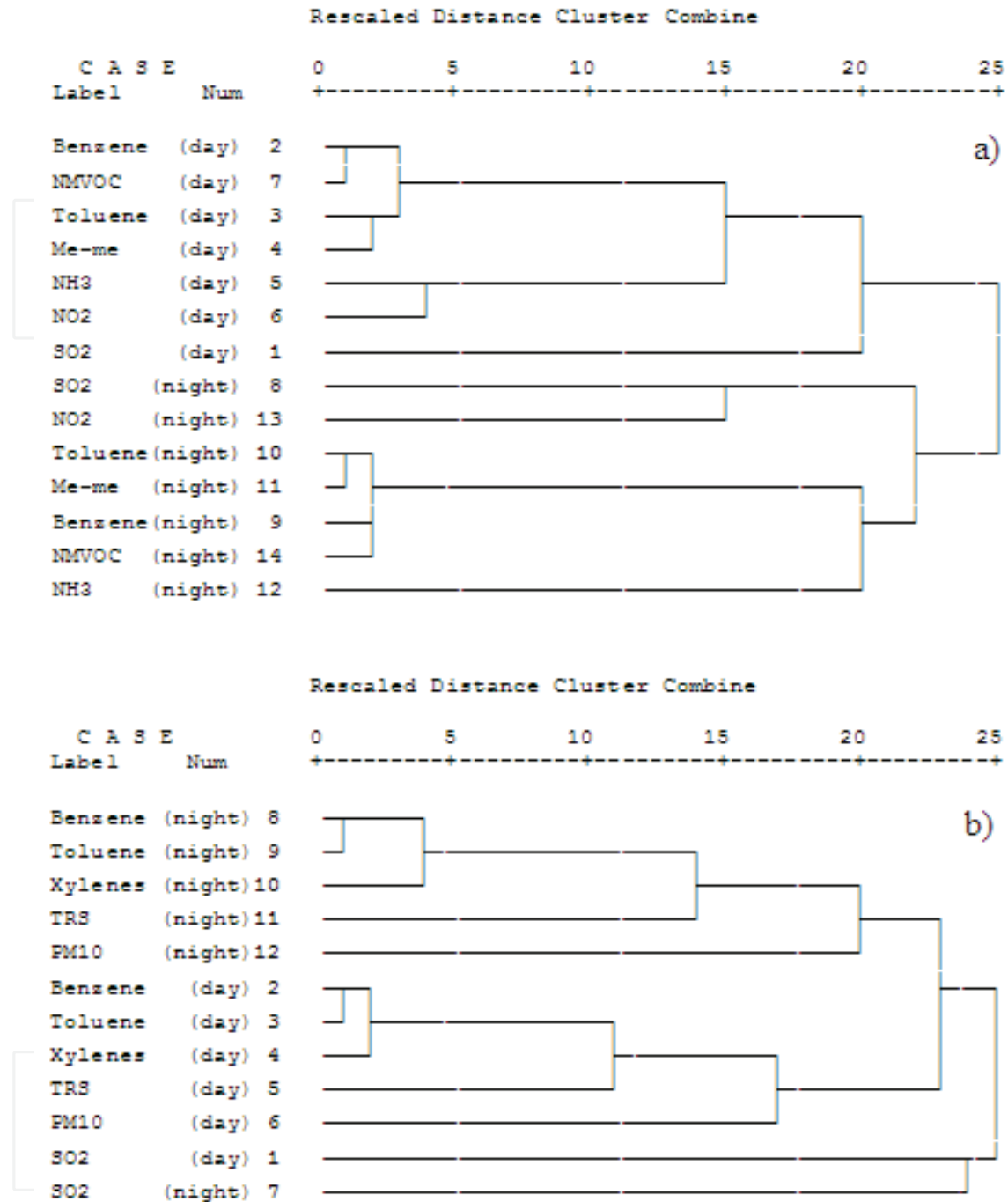
Rescaled Distance Cluster Combine

```
   C A S E       0        5       10       15       20       25
Label      Num   +--------+--------+--------+--------+--------+

Benzene  (day)  2                                              a)
NMVOC    (day)  7
Toluene  (day)  3
Me-me    (day)  4
NH3      (day)  5
NO2      (day)  6
SO2      (day)  1
SO2    (night)  8
NO2    (night) 13
Toluene(night) 10
Me-me  (night) 11
Benzene(night)  9
NMVOC  (night) 14
NH3    (night) 12
```

Rescaled Distance Cluster Combine

```
   C A S E       0        5       10       15       20       25
Label      Num   +--------+--------+--------+--------+--------+

Benzene (night)  8                                             b)
Toluene (night)  9
Xylenes (night) 10
TRS     (night) 11
PM10    (night) 12
Benzene  (day)   2
Toluene  (day)   3
Xylenes  (day)   4
TRS      (day)   5
PM10     (day)   6
SO2      (day)   1
SO2    (night)   7
```

Fig. 2. Dendrograms established of variable combinations of pollutants at the receptors: a)Vatrogasni dom, and b) Vojlovica

The production processes of the oil refinery and petrochemical industry involve the use of volatile hydrocarbons as basic raw material as well as end products. Inevitably, processes with volatile hydrocarbons result in their evaporation into the atmosphere, especially when the production and storage equipment is non-hermetic. There are devices and parts of old technologies in both the oil refinery and petrochemical industry which enable the undisturbed evaporation of significant amounts of hydrocarbons into the atmosphere. An example of such devices are decanter pipes which are used for the shipping of petroleum

products at the loading facilities for *van-* and *truck*-decanters in the oil refinery and for the shipping of pyrolytic gasoline at the loading facilities for *truck*-decanters at the petrochemical factories. When filling a tank with gasoline and other products, there is undisturbed evaporation of saturated vapour from the interior of the tank into the atmosphere.

Oily waste water treatment plants at the oil refinery and petrochemical industry are a second significant emission source of volatile organic compounds into the atmosphere.

For example, Fig. 3 shows the influence of the once-for-all pouring of pyrolytic oil on the occasion of shipping 500 m³ from the petrochemical plant that was taking place on August 23 in the afternoon's and the night's shifts, as well as on the occasion of shipping 100 m³ of pyrrolytic oil that was taking place on August 24 in the morning's shift. On the "Vatrogasni dom" measurement site, situated in the direction of the dominant southeast wind in relation to the Petrochemistry, considerable concentration growth of benzene, together with the concentration growth of toluene and methylmercaptans, was registered.
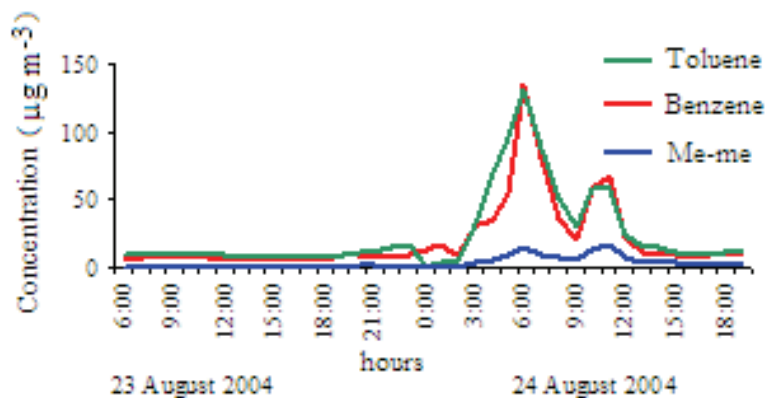
Fig. 3. The influence of pyrolytic oil shipping in petrochemical plant on concentrations of organic compounds at the "Vatrogasni dom" receptor, 5 km from the source

All these facts indicate that several types of emission sources contribute to different degrees to the concentration levels of pollutants in the ambient air, among which low altitude sources as dominant contributors have been identified in the industrial zone.

2.4 Valid Limited Value of Benzene Concentration in the Ambient Air

According to Directive 2000/69/EC, ANNEX I, relating to limit values for benzene and carbon monoxide in ambient air the limited value for benzene standardized at a temperature of 293 K and a pressure of 101,3 kPa, for the protection of human health is 5 µg/m³ on the averaging period of calendar year with margin of tolerance of 5 µg/m³ (100%) on 13 December 2000, reducing on 1 January 2006 and every 12 months thereafter by 1 µg/m³ to reach 0 % by 1 January 2010. Date by which limit value of 5 µg/m³ on the averaging period of calendar year, is to be met is 1 January 2010, except within zones and agglomerations within which a time-limited extension has been agreed in accordance with Article 3(2).

According to Article 3(2) of Directive 2000/69/EC, when the limit value laid down in Annex I is difficult to achieve because of site-specific dispersion characteristics or relevant climatic conditions, such as low wind speed and/or conditions conducive to evaporation, and if the application of the measures were to result in severe socio-economic problems, a Member State may ask the Commission for a timelimited extension. The Commission, acting in accordance with the procedure laid down in Article 12(2) of Directive 96/62/EC, may, at the

request of a Member State and without prejudice to Article 8(3) of this Directive, grant one extension for a period of up to five years if the Member State concerned:

— designates the zones and/or agglomerations concerned;
— provides the necessary justification for such an extension;
— demonstrates that all reasonable measures have been taken

to lower the concentrations of the pollutants concerned and to minimise the area over which the limit value is exceeded, and — outlines the future developments with regard to the measures which it will take according to Article 8(3) of Directive 96/62/EC.

The limit value for benzene to be granted during that timelimited extension shall, however, not exceed 10 µg/m3.

2.5 Emission Control and Decreasing Benzene Concentration in the Ambient Air Pančevo

High concentrations of evaporable carbon hydrogen and benzene having been found out in the ambient air of Pančevo, and taking into consideration the limitations set by the Directive 2000/69/EC, there have been made priorities of overhaul of key sources of emissions within the industrial zone. In support of the model Chemical Mass Balance (Seinfeld and Pandis, 1998) were identified installations and engines in both the Oil Refinery and the Petrochemistry, whose contribution to the concentration of carbon hydrogen in the ambient air is considerable. These are first of all tanks for storing evaporable products originating from oil, then pouring stations for the shipping oil derivatives, and also installations for the refinment of waste waters in the above mentioned factories. These installations make part of ground floor sources of emissions, altitudes up to 10 m. Firstly was made the plan of activity in factories of the industrial zone (Đorđević et al., 2004), which excludes pouring of evaporable derivatives during unfavourable meteorological conditions (feeble southeast wind which blows the clouds of pollution over the city), and sets new location for pouring of pyrolytic oil, at about 9 km from the previous pouring location, toward Danube (Fig. 1). The system of loading pyrolytic oil in shipping wagons was moved out from the Petrochemistry factory in 2004. This was followed by numerous actions, such as hermetic closure of tanks in both the Oil Refinery and the Petrochemistry, then substitution of one part of obsolete systems for pouring of oil derivatives on the occasion of their loading for the shiping from the Oir Refinery, then hermetic closure of installations for the refinement of waste waters in the Petrochemistry, and filally harmonization of activities of pouring evaporable carbon hydrogen with meteorological conditions. When meteorological conditions are unfavorable, i.e. when in concrete case a feeble southeast wind blows in the direction Idustrial zone - the town, and the near ground floor inversions of atmosphere are considerable (that usualy take place in the night time). In these cases the activities of pouring have been stopped, i.e they have been planned in harmony with weather forecast. These measures resulted in considerable fall of concentration of benzene on the Vatrogasni dom receptor (Fig. 1a) as of 2004.

With the exception of the period of time 1994 – 1996 when, due to heavy economic crisis, the production was reduced to the minimum or completely suspended, the average annual concentration of benzene in the period of time 1997 - 2003 surpassed border line values set in the Directive 2000/69/EC (Fig. 4). New temporary measures of controlling key sources after their identification, undertaken with the aim to reduce emission of evaporable organic compounds, began to be applied as of 2004.
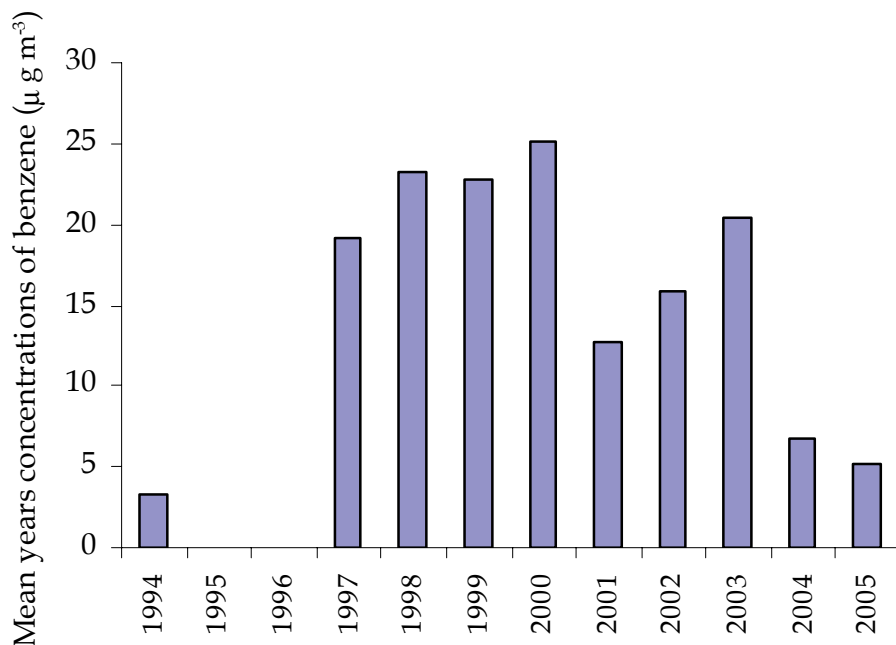
Fig. 4. Trend of annual benzene concentration in the ambient air at Vatrogasni dom receptor

Some investigations in the European cities indicate the presence of benzene originating from traffic in ambient urban air. Measurements have shown that mean annual value of the concentration of benzene in urban areas is the lowest in Copenhagen (around 3 μg m^{-3}); while in Athens the mean annual value is 20 μg m^{-3}. In these cities, the benzene derives mainly from traffic and the differences in the concentrations are the consequence of different meteorological conditions (Cocheo et al., 2000).

3. Conclusion

Harmful substances in the ambient air can be reduced only on condition that their emiters are kept under control. It is indispensable, for that purpose, to know technological processes, the nature of emiters, the link between the emission flux on the emiter and the concentration of pollutants in one part of receptors, as well as meteorological base of the area. Main emiters of pollutants have been identified, on application of statistical methods, upon analysis of technological processes in factories of the South industrial zone in Pančevo (Refinery, Petrochemistry and Fertilizer factory), by measurement of pollutants' concentrations in the ambient air and meteorological base of the area. The measures of control of emission - such as extinguishing key sources (pouring station of pyrolytic oil in the Petrochemistry), planning of emiters' activities in harmony with meteorological conditions, and reconstruction of one part of sources in both the Refinery and Petrtochemistry – had as the result the reduction of benzene concentrations in the Vatrogasni dom receptor's part for more than twice. The benzene concentrations on the measurement site which is under high influence of industrial sources reached in the years 2004 and 2005 the values acceptable with regard to the limit value of the Directive 2000/69/EC. It is important to highlight that the identification of emission sourcs on the base of measurement of pollutants' concentrations in the ambient air will be more exact and more precise if a grater number of various kinds of pollutants are measured, and if among

them there are some specific emiters' tracers. It is also importnant to highligh that the control of emissions need not alwasy be followed by high investments in order to make the effect on the ambient air more visible.

4. Acknowledgements

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