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# MUTAGENICITY AND PAH CONTENT OF AIRBORN PARTICULATES AND OF FALLEN DUSTS FROM TWO HUNGARIAN TOWNS AND EMISSION SAMPLES FROM ALUMINUM REDUCTION AND POWER PLANTS

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Summary. - *Urban air particulates (suspended particles and settling dust), furthermore dust emitted by a Soderberg aluminum reduction plant and a coal burning power plant from an industrial town, Ajka (30,000 inhabitants) were analysed for PAH content (liquid chromatography) and mutagenicity (Salmonella microsome test). Air particulates from Papa - a town of similar largeness without considerable heavy industry - and corresponding plant emission from Inota, a third town in the study, served as controls. The dust content and the PAH concentration, as well as the mutagenic potency of the air in Ajka were higher than in Papa. Mutagenicity of the airborne particulates showed a clear seasonality with a winter maximum and a summer minimum in both towns. The mutagenic potency of air correlated well with the air BaP and total PAH content in Ajka, but not in Papa. The amounts of extractable organic material and mutagenic potency as calculated for unit quantity of airborn particulate matter was higher in the Papa samples. Similar differences between the two towns were observed in the case of fallen dust, too. On the basis of examination of emitted dust, it can be stated, that in the mutagenicity of urban air, the aluminum plant emission plays considerable higher role than the power plant emission, which is the main component of air dust pollution in Ajka.*

Riassunto (Mutagenicità, contenuto di IPA di particolati aerei e polveri di ricaduta da due città ungheresi e di emissioni da impianti di riduzione di alluminio e di produzione di energia). - *Si è analizzato il contenuto in IPA e la mutagenicità in Salmonella del particolato urbano aereo, della polvere emessa da un impianto di riduzione Soderberg dell' alluminio e di un impianto per la produzione di energia a carbone della città di Ajka. Come controlli sono stati usati il particolato di Papa (una città di grandezza simile senza grandi impianti industriali) e le emissioni di un impianto di Inota. Il contenuto in polvere, la concentrazione di IPA e la mutagenicità dell' aria di Ajka sono risultate maggiori di quelle di Papa. La mutagenicità aveva un andamento stagionale, con massimi in inverno e minimi in estate in ambedue le città. La potenza mutagena dell' aria era correlata con il BaP e gli IPA totali in Ajka, ma non in Papa. La quantità di materiale organico estraibile e la potenza mutagena per unità di particolato era maggiore in Papa. Simili differenze tra le due città sono state osservate anche per la polvere caduta. Sulla base delle analisi sulla polvere emessa, si può affermare che l' emissione dell' impianto di alluminio ha un ruolo maggiore per la mutagenicità dell' aria urbana rispetto all' impianto per la produzione di energia, il quale è il principale responsabile dell' inquinamento in polvere nella aria di Ajka.*

## Introduction

Several epidemiological studies show positive correlation between tumour incidences of respiratory tract and air pollution [1-3]; lung cancer incidence for example is higher in urban than in rural areas [4-6]. Among the many factors (smoking, industrial exposure, indoor formaline exposition, etc.) polluted ambient air is definitively responsible for a certain percentage of the tumours.

There are experimental evidences for carcinogenicity of organic extracts of aerosols, collected in cities [7-11]. Such experiments or chemical analysis and identification of known mutagens is however, not applicable to large scale screening. The Salmonella/microsome (Ames) test is generally applied to assess mutagenicity not only of pure substances, but complex mixtures and extracts of environmental samples, including ambient air [12-15].

Air pollution have become a major concern all over the world and in some part of Hungary, too. A major program has been initiated in 1982-1983 to assess the health implication of air pollution in towns around an aluminum reduction plants [16].

Our main goal was to assess, analyse and possibly quantify the airborn particulates, the extractable organic content, the total PAH and BaP content and the mutagenic activity in two middle-sized towns in Hungary. We further studied these parameters concerning the fallen dust and some of the major emission sources.

Ajka, with about 30,000 inhabitants, has the reputation to be one of the most air-polluted town in Hungary. An aluminum reduction plant, further a power plant represent the heavy industry. Papa is a town of similar largeness without heavy industry. Two series of studies were carried out with airborn particulate matter from Ajka (1982-83 and 1983-84). From Papa, only winter, spring and summer samples were analysed in 1983-84. Inota served as a control having aluminum reduction and power plant. Both aluminum production plants are based on Soderberg technology, using horizontal stud in Ajka and vertical stud in Inota. Both power plants use coal for burning.

## Materials and methods

Emission samples from the aluminum reduction plants or power plants were taken from the exhaustion systems. Fallen dust was collected two times for 6 months: between October-March and April-September, 1985-86. Urban air particulates were collected on filters by a conventional samplers, operating typically for 8 h (about 800 m³). After weighing the filter, extraction was carried out either in Soxhlet apparatus with benzene: acetone mixture (19:1) for 8 hours or by sonication in acetone for 5 min, followed by dichloromethane for 5 min. After evaporating the solvents, the residue was weighed and dissolved in DMSO and this stock solution was kept at - 35 °C in $N_2$ atmosphere until use.

The same sampling and extraction procedure was carried out for chemical determination of total BaP and PAH content of the samples (HPLC, Varian 4100).

The mutagenicity studies were carried out as described by Maron and Ames [17] in two strains (TA100 and TA98) by plate incorporation technique, with and without metabolic activation (10% rat S9 fraction).

Out of the several parameters registered in the study, we focus here only to the: a) mutagenic ratio (MR), which represents a ratio of the number of induced and control revertant colonies (solvent or extraction controls); b) mutagenic potency (MP): number of net (induced minus control) revertant colonies, calculated for m³ of air or mg of dust.

## Results

### *Airborn particulate matter in Ajka and Papa*

The amount of particulates, organic extracts, PAHs and BaP is shown in Fig. 1.

Comparing the parameters in the two towns, the amount of collected dust, the extractable organics, total PAH and BaP are consistently higher in Ajka than in Papa. Total PAH and BaP content of the extracts shows a definite winter maximum in Ajka. Similar seasonality with a winter maximum was observed in the mutagenic potency, as expressed by revertants/m³ (Fig. 2). In winter samples,

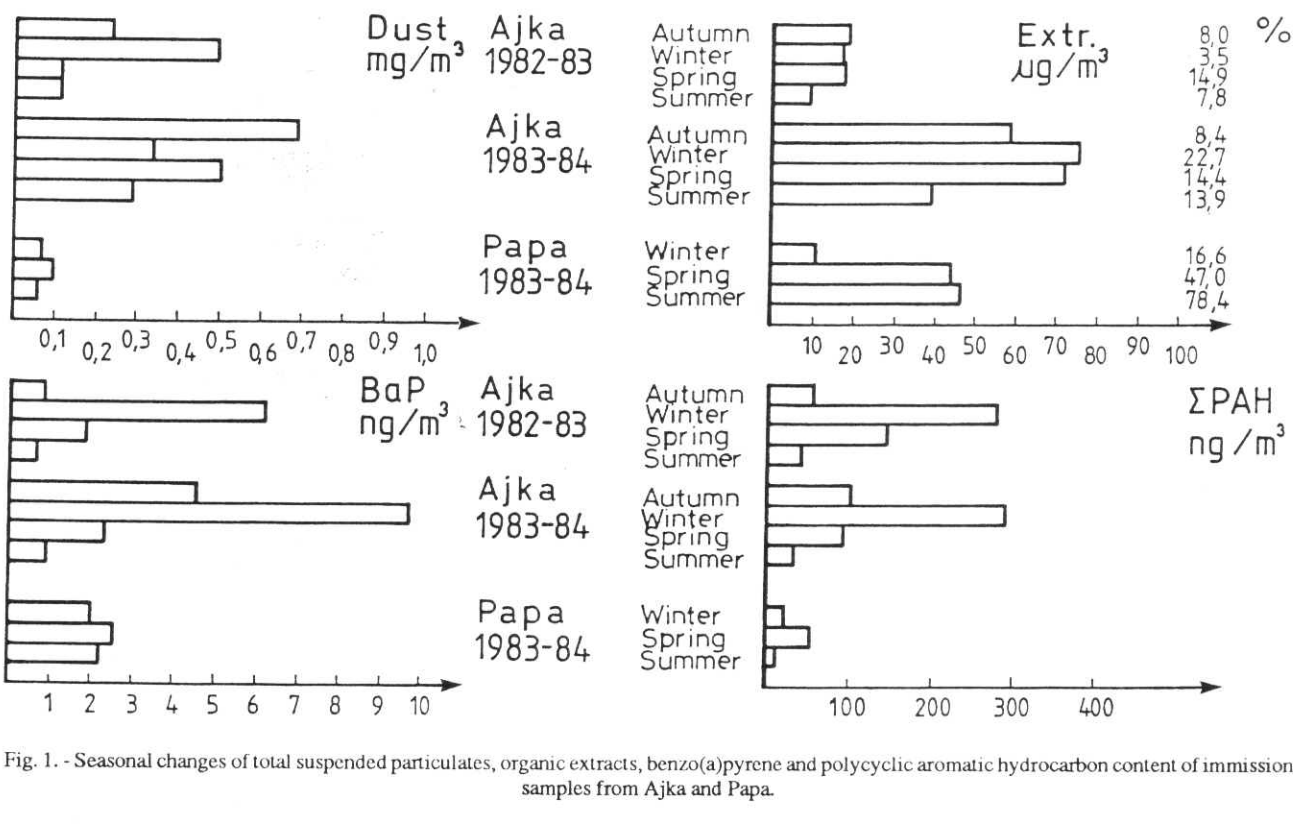


Fig. 1. - Seasonal changes of total suspended particulates, organic extracts, benzo(a)pyrene and polycyclic aromatic hydrocarbon content of immission samples from Ajka and Papa.

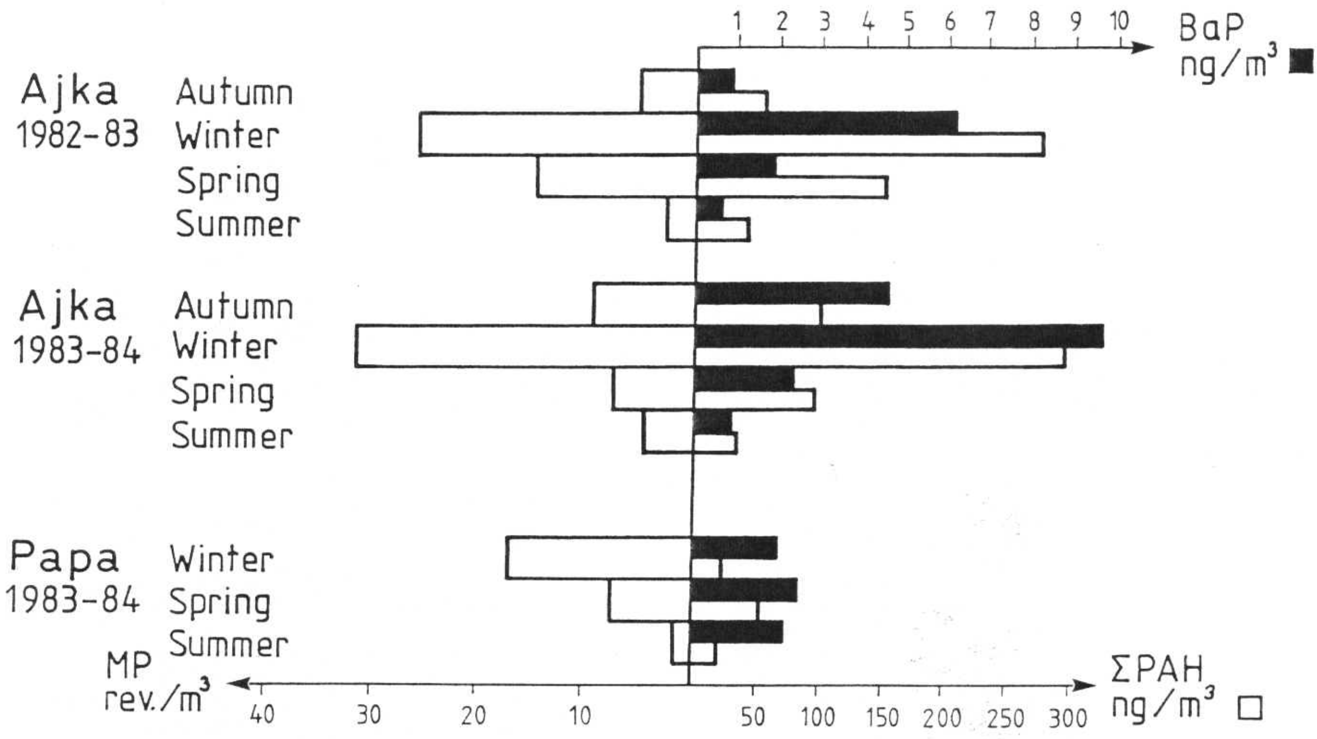


Fig. 2. - Seasonal comparison of PAH content and mutagenic potency of air samples from Ajka and Papa.

mostly the frameshift mutagens (predominantly indirect) were responsible for the activity, but mutagens causing base changes, were also present. If one compared the total PAH and BaP content with the mutagenicity, there was good correlation in the Ajka samples in two years. Winter samples from Papa had relatively low PAH and BaP content, but the mutagenic potency of these samples was relatively high (Fig. 2), that means that unlike to Ajka, no such good correlation between mutagenicity and BaP and PAH content is observed in Papa. This indicates, that the compounds, responsible for the mutagenicity in Papa are different from that in Ajka which means that PAHs play a less important role.

*Fallen dust*

We compared several parameters of fallen dust, collected in towns (Ajka and Papa) during heating and non-heating season (Fig. 3).

Similar to the airborn particulates, the amount of the dust was higher, but the percentage of extractable organics was lower in Ajka than in Papa.

As far as the non-heating season is concerned, the BaP and PAH content, further the mutagenic potency was higher in Ajka than in Papa and these parameters showed rather good correlation. The mutagenic potency in the heating season in Papa was found to be much higher than in Ajka. This increased mutagen potency did not correlate well with the PAH and BaP content in Papa.

In evaluating the importance of mutagenicity of fallen dust, one should bear in mind that during the long collection period many secondary reactions, decompositions and interactions may occur which influence the final

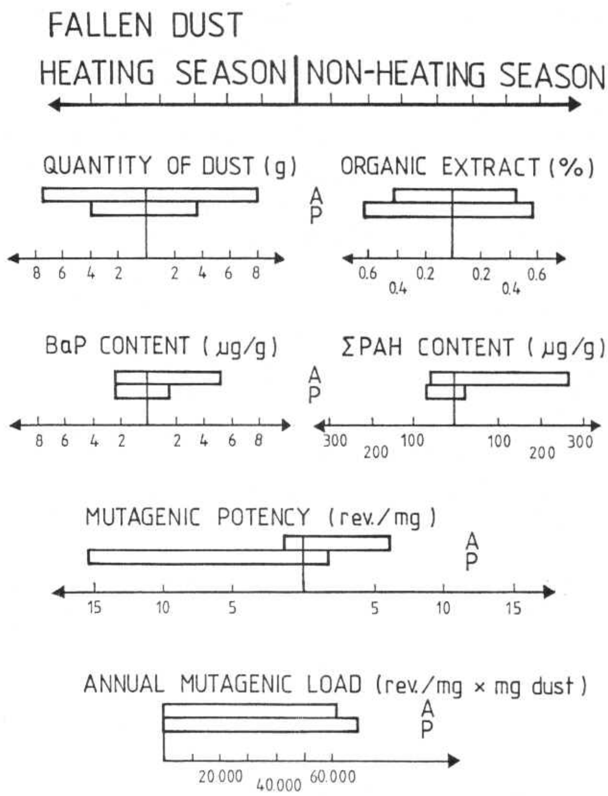


Fig. 3. - Comparion of several parameters of fallen dust collected in heating and non-heating seasons in Ajka and Papa.

results. On the other hand, it always contains soil, which has consistently been shown to be mutagenic.

If one calculates the possible annual mutagenic load (rev/mg x mg dust) this value is slightly higher in Papa than in Ajka.

Table 1. - *Polycyclic aromatic hydrocarbon content and mutagenic potency of emission samples in Ajka and Papa*

| | Extracts (%) | BaP μg/g | PAH μg/g | MP rev./mg |
|---|---|---|---|---|
| **Aluminum Reduction Plants** | | | | |
| Ajka 1982 | | 955.50 | 4670.70 | 318.8 |
| Ajka 1986 | 12.674 | 356.10 | 10131.99 | 204.66 |
| Inota 1982 | | 17.50 | 2040.40 | 1569.8 |
| Inota 1986 | 1.990 | 76.77 | 1502.00 | 1150.2 |
| **Power plants 1986** | | | | |
| Ajka cyclon | 0.121 | 0.035 | 2.52 | 0.14 |
| Ajka electrofilter | 0.104 | 0.042 | 13.074 | 0.21 |
| Inota | 0.109 | 0 | 4.128 | 0 |

*Emission samples*

Samples taken from two power plants had no or very low activity. This result is rather comforting since the majority of air dust pollution, which causes major public concerns in the town, comes from the power plant (Table 1). Comparing the emissions of the two alu-plants, the amount of organic extracts , the BaP and PAH content of the Ajka sample was higher than that of Inota, the mutagenic potency, however, was much higher in Inota. The total PAH and BaP content did not parallel with the mutagenic potency, the measured values of these compounds were higher in Ajka (Table 1). As far as the nature of mutagenicity is concerned, indirect frameshift mutagens were predominant in Ajka samples, but indirect base-change mutagens were present in high amount in the Inota sample. The high direct frameshift activity might be attributed to polar aromatic hydrocarbons (nitro-or oxy-derivatives).

Table 2 summarizes the mutagenic potencies of different emission and immision samples.

Table 2. - *Summary of mutagenic potencies of different immission and emission samples*

| Airborn particulates | | | rev./mg |
|---|---|---|---|
| Ajka | 1982-83 | Winter | 53.1 |
| | | Summer | 18.0 |
| | 1983-84 | Winter | 95.0 |
| | | Summer | 15.0 |
| Papa | 1983-84 | Winter | 250.0 |
| | | Summer | 26.5 |
| **Fallen dust 1985-86** | | | |
| Ajka | | Heating season | 1.45 |
| | | Non-heating season | 6.24 |
| Papa | | Heating season | 15.54 |
| | | Non-heating season | 1.82 |
| **Aluminum reduction plant emissions** | | | |
| Ajka | | 1982 | 318.75 |
| | | 1986 | 204.66 |
| Inota | | 1982 | 1569.75 |
| | | 1986 | 1150.20 |
| **Power plant emissions 1986** | | | |
| Ajka | | cyclon | 0.14 |
| | | electrofilter | 0.21 |
| Inota | | | 0 |

## Conclusions

Summarizing the data obtained in Ajka and Papa, the following conclusions can be drawn.

*Ajka*

There is a good correlation between total PAH, BaP content and the mutagenic activity of airborn particulates. Mutagenic potency of ambient air shows definite seasonality; in winter, a considerable part can be ascribed - besides relative constant industrial emission - to heating. In summer, the importance of increased traffic, agricultural work, etc. is probable. The role of atmospheric changes (wind, fog, etc.) could not be estimated - in the absence of relevant data. Not considering other emission sources, about 85% of the mutagenic load can be attributed to aluminium reduction plant and 15% to the power plant. The mutagenic potency of ambient air in Ajka is rather similar in magnitude to other industrial towns - as judged from published data [18, 19].

*Papa*

Contrary to Ajka, no correlation could be established between mutagenic potency, PAH and BaP content of the airborn particulates. In absolute terms, the mutagenicity of the air samples is less than that in Ajka but the relative mutagenicity (as calculated to mg dust) is higher, than in Ajka - that raises the possibility of the existence of other important emission sources.

The results of the study provided further evidences that the mutagenic activity of air dusts is - besides chemical analysis - an important factor in assessing human genotoxic hazard.

The terms of mutation and cancer is well known and convincing to initiate measures for reducing emission from the aluminum reduction plants. The real solution - a complete change in the technology - is unfortunately a remote possibility.

According to the IARC evaluation, working in aluminum production is carcinogenic to humans. We can add that living in the vicinity of such aluminum plant poses also increased mutagenic load and carcinogenic hazard.

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