Urban Air Pollution by Carcinogenic and Genotoxic Polyaromatic Hydrocarbons in the Former USSR

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The content of major carcinogenic and genotoxic polyaromatic hydrocarbons (PAH) in urban air, vehicle, and industrial emissions is assessed. A sensitive, specific, and selective method for PAH and nitro-PAH quantitation was developed on the basis of low-temperature luminescence-spectra of frozen polycrystalline solutions. Polyarene contents in urban air and urban industrial emissions, as well as vehicle exhausts, are compared to the Russian Ministry of Health standard of maximal permissible concentration for benzo[a]pyrene in the ambient air. Special attention was paid to the study of PAH profiles and characteristics of different sources. The feasibility of using benzo[a]pyrene to assess PAH content in industrial and vehicle emissions was explored. PAH profiles in the ambient air and local emissions are compared. In addition, genotoxicity of the emissions of coke plant and diesel engine exhausts are compared to their polyarene content. — Environ Health Perspect 102(Suppl 4):49–53 (1994).

Key words: carcinogenic and genotoxic polyaromatic hydrocarbons (PAH), industrial emissions, luminescence spectra, maximal permissible concentration (MPC), nitro-PAH, urban air pollution, vehicle exhausts

Introduction

The respiratory-cancer risk is closely related to the content of polycyclic aromatic hydrocarbons (PAH) and their nitro-derivatives in urban air and industrial areas (1). At the International Symposium on Cancer and Air Pollutions (Stockholm, March 1977), special scrutiny was paid to the identification of benzo[a]pyrene (B[a]P). This substance was seen as an indicator of the carcinogenic hazard in environmental pollution (2). Later, in 1985, at the International Agency for Research on Cancer (IARC) Workshop, it was reported (according to Redmond’s data (3)) that B[a]P could be a suitable index for the carcinogenic potential of PAH in air from the topside of a coke oven, as well as PAH in the ambient air. (The cumulative cancer risk has a linear correlation with the concentration of B[a]P.)

From 1972 to 1973, in the former USSR, the maximal permissible B[a]P concentration (MPC) in the ambient air (1 ng/m³) was established (4). Health Councils of several countries, such as the Netherlands, have also recommended to decrease the B[a]P concentration in the ambient air below 1 ng/m³. These proposals are aimed at the concomitant reduction of PAH concentrations.

Another group of genotoxic and carcinogenic compounds, nitropolyarenes, may be formed from PAH in emissions from various sources, in particular, diesel engine exhausts combined with the presence of nitrogen oxides. So, the recommendation should be considered with regard to both groups of these carcinogenic and genotoxic substances present in the urban air.

Results and Discussion

Epidemiological Studies

Epidemiological studies were carried out in the Moscow Cancer Research Center (5) with the aim of assessing the impact of ambient air pollution on the lung cancer incidence rate in 26 industrial cities in the former USSR. Studies of dominant industries have demonstrated (Table 1) that the lowest standardized incidence rate was characteristic of the cities with developed food and textile industries (it appeared to be 63.0 for males, 9.0 for females). The highest rate was characteristic of the cities with developed ferrous and nonferrous metallurgy (90.7 for males, 11.1 for females). It was shown that the major constituent, particularly of the metallurgical emissions, were PAH.

Table 1. Respiratory cancer incidence rate in the cities of the former USSR with different types of industry.

| Cities          | Males | Females |
|-----------------|-------|---------|
| Ferrous and nonferrous metallurgy, chemical industry, machine building |       |         |
| Magnitogors | 95.5  | 12.7    |
| Ust-Kamenogorsk | 95.4  | 13.0    |
| Lipetsk       | 81.2  | 12.0    |
| Krasnoyarsk  | 81.8  | 11.9    |
| Sverdlovsk    | 72.1  | 9.8     |
| Donezk       | 66.8  | 11.5    |
| Chelyabinsk  | 95.2  | 10.5    |
| Eravan       | 51.9  | 12.4    |
| Kurgan       | 77.6  | 9.5     |
| Ryazan       | 82.1  | 8.9     |
| Ferrous and nonferrous metallurgy, machine building |       |         |
| Zaporozhe   | 64.6  | 8.6     |
| Krivoi-Rog  | 96.4  | 10.4    |
| Norilsk     | 110.6 | 9.2     |
| Leninoorsk  | 91.1  | 9.9     |
| Chemical industry (coke and petrochemical industries included) |       |         |
| Omsk         | 85.1  | 9.3     |
| Kemerovo    | 83.2  | 11.0    |
| Baku         | 81.0  | 10.9    |
| Ufa          | 3.5   | 9.4     |
| Rostov-na-Donu | 70.5  | 12.2    |
| Kursk       | 56.2  | 8.6     |
| Voronez     | 54.1  | 7.9     |
| Vilnus      | 63.6  | 7.7     |

Our investigations have shown that the B[a]P total emission was 2 kg/day at the Rustay Metallurgical Plant (B[a]P) maximal concentration at the topside of coke-oven battery—11 mg/m³ (6). Since the emissions cross the borders of the sanitary protection zone.

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area, the B[a]P MPC is exceeded 50-to 100-fold (according to our data) in the area surrounding the coke plant.

In cities with heavy traffic, MPC is sometimes exceeded 22-fold. Background B[a]P concentrations for mountainous areas (Central Asia) is 0.05 ng/m³, and for the industrial regions (the Baltic coast, Lithuania) and park areas in the industrial cities (Moscow, Baku), it is 1 ng/m³ (7). The MPC for B[a]P is exceeded in urban areas because of local emissions.

**Carcinogenic Risk Assessment by the Analysis of Carcinogens in Air Pollutions**

The studies carried out at our laboratory are aimed at cancer risk-assessment through analyzing the pollutant content of the environment, and of the ambient air in particular. The essential points of our program are as follows (Figure 1):  

- **a)** development and improvement of the methods for identification and quantitation of carcinogenic substances of different chemical classes, including PAH, nitropolycyclic and carcinogenic N-nitrosamines;  

- **b)** Practical realization of these methods for air pollution monitoring and hygienic control in the former USSR (8) as well as the application in industry of regulations for maximal permissible B[a]P emissions (MPE). MPE would ensure air-quality standards for B[a]P (MPC) and act as a guideline for step-by-step reduction of emissions;  

- **c)** Study of the industrial and vehicular emissions of carcinogenic substances (PAH in particular). Special attention is paid to examining the PAH profiles characteristic of different emission sources. There is also a focus on possible employing B[a]P as a gauge for assessing PAH content in plant emissions and vehicle exhausts;  

- **d)** Study of air pollution and role of different sources in PAH pollution. PAH profiles in the ambient air and local emissions are compared; and  

- **e)** Because the study of carcinogenicity and genotoxicity of atmospheric emissions cannot be limited just to identification and quantitation of PAH and nitro-PAH, short-term genotoxicity assays were used in conjunction with chemical characterization of the emissions.

**Examples of Research**

In the following examples special attention was given to the development and improvement of our methods of analysis (9,10).

**Method for Identification and Quantitation of PAH and Nitro-PAH.** The method for identification and quantitation of PAH and nitro-PAH is based on fine-structure luminescence spectra of frozen n-alkane solutions (77°K) under selective excitation. Quantitation is based on the principle of the addition of the standard. The error ± 10%. Sensitivity of the method (with regard to B[a]P and pyrene) is 0.5 × 10⁻¹⁰ g/ml. Luminescence (fluorescence and phosphorescence) spectra of several PAH, which occur most frequently in urban air, have been studied. The spectra were obtained using a Perkin-Elmer luminescent spectrometer with a low-temperature accessory for phosphorescence. Specificity and selectivity of spectra for the most of the PAH is so high that it makes possible their quantitation after benzencyclohexane ultrasonic extraction and a single chromatographic fractionation in a thin layer of aluminum oxide.

In Figure 2, photographs of the first and second fractions after chromatography of the coke plant emission extract are presented. The structure of pyrene is clearly seen in the first fraction, and that of the B[a]P, in the second fraction under selective excitation.

Wavelengths chosen as analytical in excitation and luminescence spectra for 17 PAH under study are presented in Table 2.

![Figure 1. Carcinogenic risk assessment by the analysis of carcinogens in air pollution (research directions).](image)

![Figure 2. Fluorescence spectra by selective excitation of fractions after thin-layer chromatography of exhaust extract from coke batteries: α₁, α₂, spectra of pyrene and benzo[a]pyrene standard solutions, respectively; β₁ and β₂ spectra of the first and second fractions, respectively.](image)

### Table 2. Spectral characteristics of polycyclic aromatic hydrocarbons studied.

| PAH     | Excitation  | Emission  |
|---------|-------------|-----------|
| P       | 338.0       | 372.0     |
| Fl      | 362.0       | 406.5     |
| B[a]A   | 292.0       | 384.5     |
| Chr     | 272.0       | 361.0     |
| B[fl]   | 304.0       | 396.0     |
| B[a]Fl  | 310.0       | 403.0     |
| P       | 414.5       | 449.3     |
| B[a]P   | 298.0       | 403.0     |
| B[e]P   | 334.0       | 388.0     |
| DB[a,h]A| 301.0       | 394.2     |
| DB[a,c]A| 291.0       | 375.0     |
| Bl(g,h,| 313.0       | 449.0     |
| P       | 303.0       | 419.6     |
| DB[a,h]P| 398.0       | 431.7     |
| DB[a,c]P| 313.0       | 449.0     |
| Cor     | 307.0       | 443.8     |
| Fnt     | 255.0       | 347.0     |
| IndP    | 362.0       | 463.0     |

**Abbreviations:** PAH, polycyclic aromatic hydrocarbons; P, pyrene; Fl, fluoranthene; B[a]A, benzo[a]anthracene; Chr, chrysene; B[fl], benzo[fl]fluoranthene; B[a]Fl, benzo[k]fluoranthene; P, perylene; B[a]P, benzo[a]pyrene, B[e]P, benzo[e]pyrene, DB[a,h]A, dibenz[a,h]anthracene; DB[a,c]A, dibenz[a,c]anthracene; B[g,h,j]P, benzo[g,h,j]perylene; DB[a,j]P, dibenz[a,j]pyrene; Cor, coronene; Fnt, phenanthrene; IndP, ineno[1,2,3-c-d]pyrene.
Table 3. Conditions for nitro-polyacyclic aromatic hydrocarbons analysis.

| Nitro-PAH, (Internal standard) | Excitation, nm | Phosphorescence, nm | Sensitivity, ng/ml |
|-------------------------------|---------------|-------------------|-------------------|
| 1-Nitropyrene                 | 417           | 632               | 5.00              |
| (B[e]P)                       | 334           | 537               | 0.50              |
| 1,3-Dinitropyrene             | 418           | 640, 656          | 1.00              |
| (B[e]P)                       | 334           | 537               | 0.50              |
| 2-Nitrofluorene               | 540           | 510               | 0.25              |
| (1,3-Dinitropyrene)           | 418           | 640               | 1.00              |

Abbreviations: PAH, polycyclic aromatic hydrocarbon; (B[e]P), benzo[e]pyrene.

Table 4. Moscow air pollution with polycyclic aromatic hydrocarbons (one of ecologically unsafe districts).

| Characteristics of the sampling site | B[a]P concentration, ng/m³ | Maximal permissible B[a]P concentration, ng/m³ |
|--------------------------------------|----------------------------|-----------------------------------------------|
| Regular traffic                      | 5.4 ± 1.1                  | 1.0                                           |
| Crossroads with heavy traffic        | 20.0 ± 1.4                 |                                               |
| Area of the oil processing plant     | 13.4 ± 0.6                 |                                               |

Analysis of nitro-PAH is rather complicated since it extraction, enrichment and fractionation require a large number of steps aimed. However, our modification of the nitro-PAH analysis method may facilitate this procedure. After a single fractionation by chromatography, nitro-PAH may be identified and quantified by the phosphorescence spectra of frozen polycrystalline solutions. Phosphorescence spectra of 1-nitropyrene, 2-nitrofluorene, and 1,3-dinitropyrene are shown in Figure 3. The spectral structure is not as fine as that of PAH. (To compare these spectra, the phosphorescence spectrum of benzo[e]pyrene (B[e]P) is used as an internal standard in nitro-PAH quantitation. Table 3 shows the analytical lines in the excitation and phosphorescence spectra for the nitro-PAH under study, as well as the internal standard and the sensitivity of the suggested method.

Study of PAH Profiles in Vehicle and Industrial Emissions and Ambient Air. A long-term study of PAH emissions by gasoline-powered automobiles in the Europa test and their effect on urban air pollution was carried out with the assistance of Abgasprüfelle (East Berlin) (71). The samples were taken out on a test bench with a rotating drum. The content of 12 major PAH constituents was determined by the luminescence spectra in frozen polycrystalline solutions. Absolute exhaust values depended upon the engine type, but the PAH profile obtained was rather similar to that of various European automobile exhausts (4). Stability of the B[a]P profile makes it possible to use one of the constituents, B[a]P in particular, for rough quantitation of the other PAH and of the total PAH content. Mean B[a]P emission is 1 μg/km for Russian gasoline automobiles, as it was determined in Europa test; this value may increase 10-fold and more if the engine does not work properly. So care should be taken in the control of this parameter.

Experimental study of East Berlin air pollution has shown relatively high concentrations of PAH (and B[a]P in particular) in the streets with heavy traffic. PAH concentration values are the mean of 11 measurements; B[a]P concentration values are the mean of 240 measurements. PAH profiles in the areas with heavy traffic are quite similar to those of automobile exhausts which reveals the major source of pollution in the area. In the summer, vehicular contribution to urban air pollution increased 58.6 ± 6.9% (in the industrial pollution background).

In Moscow, mean B[a]P concentration was elevated to 22 MPC at crossroads with heavy traffic (Table 4). At the highway in the same district they amounted to 6 MPC, while in the area adjacent to the oil-processing plant they amounted to 14 MPC (mean daily MPC for B[a]P is 1 ng/m³).

Complex studies carried out at the Rustavy (12), Kemorovo, and Altay coke plants (13) are examples of our studies on PAH content in industrial emissions, aimed, as said above, at the development of MPE for these carcinogens and the reduction of PAH emissions. Gas exhausts from coke batteries were shown to be the major sources of PAH. Quantitation of PAH emitted by different sources has shown the PAH profile (the ratio of individual PAH concentrations to that of B[a]P) to be practically independent of the emission sources of all the coke plants. Figure 4 presents PAH profiles for five sources at the Rustavy coke plant, for the dust on coke battery surface, as well as for the oven topside at the Kemorovo and Altay coke plants. Similar PAH profiles were
observed for the air in the residential area adjacent to coke plants (Kemerovo), which points to the major source of pollution. Stability of PAH allows organizing the relative concentrations of the PAH constituents (related to B[a]P), into a system of coefficients (Table 5, column 5). Each coefficient may be employed to calculate the concentration of PAH using the measured B[a]P concentration. The following formula is used: C(PAH) = K(PAH) × C(B[a]P). (Absolute concentration values for coke-chemical plant emissions are presented in Table 5).

Concurrent Studies of PAH Content and Genotoxicity. Concurrent studies of PAH content and genotoxicity of industrial and vehicle emissions may be presented by the study of the coke plant emissions (I4). The total PAH and B[a]P content in the samples from the exhausts of two coke batteries have been checked along with the genotoxicity of these extracts, for bacterial tester strains. The strains tested for were Salmonella typhimurium TA98, TA100, and a hybrid strain, AG276, bearing two types of point mutations (a base-pair substitution mutation, his G46, and a frameshift mutation arg-2) and a transposon TN10 (I2). Considerable mutagenic effect of the tested extracts was observed with metabolic activation by S-9 mixture, which is characteristic of the PAH (Table 5, column 2). A double increase in the yield of frameshift revertants was noted in the absence of S-9, probably due to direct-acting mutagens, the nitroarenes. A 3- to 5-fold increase in the rate of TN10 precise excision may be attributed to SOS-induction. Comparison of the rapid spectral analysis of PAH contents to genotoxicity data allowed the assessment of potential carcinogenicity of coke plant exhausts.

Concurrent studies of the PAH content and mutagenicity of the vehicle emissions were carried out for two types of diesel-powered engines (one shaft gas-turbine GTE-5 engine with a power of 30 kilowatts and a standard D-54A diesel with a power of 40 kilowatts) (Table 6). The extracts of soot from the two engines induced reversions in Salmonella both with and without metabolic activation. Furthermore, the extracts possessed higher mutagenic activity (I5). The direct mutagenic effect of the exhausts depended neither on the presence of B[a]P nor of any of the other PAH. The culpable agent was most likely nitro-PAH.

Conclusions

Urban air carcinogens (polynuclears and their nitrated derivatives, in particular) are not the only factors to be used in determining carcinogenic hazard.

Methods for identification and quantitation of carcinogenic substances in urban air and industrial and vehicle emissions necessary for realization of program aimed for the hazard reduction were developed at our laboratory. The standards require limita-

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**Table 5.** Polycyclic aromatic hydrocarbons content in the exhaust of coke batteries and in the air pollution (M ± m).

| PAH     | Coke batteries exhausts | Air pollution |
|---------|-------------------------|--------------|
|         | Absolute concentration, ng/m³ | Mean      | Workplace area | Adjacent area |
| B[a]P   | 78500 ± 6000            | 24 ± 0.2    | 2.9 ± 0.7      |              |
| Relative PAH concentrations, % |
| P       | 2.5 ± 0.2               | 2.2 ± 0.3   | 2.4 ± 0.2      | 2.9 ± 0.7    |
| Fl      | 4.1 ± 0.3               | 4.7 ± 0.8   | 4.6 ± 0.8      | 5.2 ± 1.7    |
| B[a]A   | 2.6 ± 0.1               | 1.3 ± 0.4   | 1.9 ± 0.3      | 1.7 ± 0.3    |
| Chr     | 3.1 ± 0.4               | 3.6 ± 0.7   | 2.9 ± 0.5      | 4.1 ± 1.5    |
| B[k]Fl  | 5.9 ± 0.5               | 4.3 ± 0.8   | 4.8 ± 0.8      | 4.7 ± 1.2    |
| B[b]Fl  | 1.1 ± 0.2               | 1.3 ± 0.1   | 1.5 ± 0.2      | 1.7 ± 0.5    |
| Fl      | 0.5 ± 0.1               | 0.5 ± 0.1   | 0.6 ± 0.1      | 0.7 ± 0.1    |
| B[a]P   | 1                      |              | 1              |              |
| B[e]P   | 0.8 ± 0.1               | 0.9 ± 0.2   | 0.9 ± 0.2      | 1.0 ± 0.3    |
| DB[a, h]A| 0.8 ± 0.2              | 1.3 ± 0.2   | 1.0 ± 0.2      | 0.8 ± 0.3    |
| DB[a, c]A| 1.1 ± 0.1              | 1.0 ± 0.2   | 1.0 ± 0.2      | 0.8 ± 0.2    |
| DB[g, h]Pi| 2.1 ± 0.2              | 1.7 ± 0.3   | 1.9 ± 0.3      | 2.4 ± 0.7    |
| DB[a, h]P| 0.03 ± 0.01            | 0.09 ± 0.01 | 0.07 ± 0.01    | 0.08 ± 0.02  |
| DB[a, h]P| 0.03 ± 0.01            | 0.05 ± 0.01 | 0.04 ± 0.01    | 0.04 ± 0.01  |
| Cor     | 0.8 ± 0.1               | 0.8 ± 0.1   | 0.8 ± 0.1      | 0.8 ± 0.2    |
| Total   | 26.5 ± 2.3              | 25.2 ± 4.5  | 24.8 ± 4.2     | 26.4 ± 3.7   |

Abbreviations: PAH, polycyclic aromatic hydrocarbons; B[a]P, benzo[a]pyrene; P, pyrene; Fl, fluoranthene; B[a]A, benzo[a]anthracene; Chr, chrysene; B[k]Fl, benzo[k]fluoranthene; B[b]Fl, benzo[b]fluoranthene; Fl, perylene; B[e]P, benzo[e]pyrene; DB[a, h]A, dibenzo[a, h]anthracene; DB[a, c]A, dibenzo[a, c]anthracene; DB[g, h]Pi, dibenzo[g, h]pierylene; DB[a, h]P, dibenzo[a, h]pyrene; DB[a, h]P, dibenzo[a, h]pyrene; Cor, coronene.
Table 6. Mean values of concentration ratio of individual polycyclic aromatic hydrocarbons and benzo[a]pyrene in soot extracts of GTD-5 and D-54A diesel engines.

| PAH               | GTD-5 Gas exhaust | Soot | t  | GTD-5 Gas exhaust | Soot | t  |
|------------------|-------------------|------|----|-------------------|------|----|
| P                | 15.7 ± 0.56       | 15.0 ± 0.72 | 0.77 | 6.76 ± 0.11       | 6.9 ± 0.32 | 0.56 |
| Chr              | 15.1 ± 0.54       | 14.5 ± 0.23 | 1.05 | 2.5 ± 0.34        | 2.4 ± 0.16 | 0.26 |
| Fl               | 34.4 ± 1.32       | 32.8 ± 2.00 | 0.67 | 7.62 ± 0.30       | 7.7 ± 0.24 | 0.24 |
| B[a]A            | 2.7 ± 0.16        | 2.4 ± 0.22  | 1.00 | 2.2 ± 0.15        | 2.0 ± 0.21 | 0.77 |
| B[b]F            | 9.8 ± 0.22        | 10.2 ± 0.44 | 0.88 | 3.0 ± 0.22        | 2.7 ± 0.34 | 0.55 |
| B[ghi]P           | 0.18 ± 0.01       | 0.16 ± 0.02 | 1.00 | 0.23 ± 0.03       | 0.26 ± 0.02 | 0.85 |
| Pt               | 0.23 ± 0.01       | 0.21 ± 0.02  | 0.91 | 0.07 ± 0.01       | 0.06 ± 0.01 | 0.71 |
| B[a]P             | 1.0 ± 0.07        | 1.00 ± 0.00 | 0.00 | 1.0 ± 0.00        | 1.0 ± 0.00 | 0.00 |
| B[b]P             | 4.3 ± 0.26        | 4.83 ± 0.54  | 0.88 | 2.33 ± 0.15       | 2.85 ± 0.32 | 1.37 |
| B[ghi]P           | 2.8 ± 0.12        | 2.76 ± 0.21  | 0.18 | 0.67 ± 0.07       | 0.58 ± 0.08 | 0.82 |
| DB[a,c]A          | 0.31 ± 0.03       | 0.42 ± 0.05  | 0.20 | 0.08 ± 0.01       | 0.09 ± 0.01 | 0.36 |
| DB[a,h]A          | 0.85 ± 0.03       | 0.70 ± 0.02  | 1.25 | 0.39 ± 0.10       | 0.43 ± 0.14 | 0.23 |
| Cor               | 0.44 ± 0.03       | 0.46 ± 0.03  | 0.50 | 0.17 ± 0.02       | 0.15 ± 0.02 | 0.71 |
| DB[a,IP]          | 0.06 ± 0.01       | 0.05 ± 0.01  | 0.71 | 0.05 ± 0.01       | 0.05 ± 0.02 | 0.00 |
| DB[a,h]P          | 0.21 ± 0.01       | 0.18 ± 0.02  | 1.36 | 0.11 ± 0.02       | 0.13 ± 0.01 | 0.91 |

Abbreviations: P, pyrene; Chr, chrysene; Fl, fluoranthene; B[a]A, benzo[a]anthracene; B[b]F, benzo[b]fluoranthene; B[ghi]F, benzo[k]fluoranthene; Pt, perylene; B[a]P, benzo[a]pyrene; B[b]P, benzo[b]pyrene; B[ghi]P, benzo[k]pyrene; DB[a,c]A, dibenzo[a,c]anthracene; DB[a,h]A, dibenzo[a,h]anthracene; Cor, coronene; DB[a,IP], dibenzo[a,IP]pyrene; DB[a,IP], dibenzo[a,h]pyrene; t, Student's t test; a–d are absolute values of PAH concentrations. 4 0.18–1.00 μg/m² at Pe = 0–30 kg (Pe: GTD-5 power). 4 0.3 μg/g soot. 4 1.4–2.25 μg/m² at Pe = 0–30 kg (Pe: D-54A power). 6 0.8 μg/g soot.

tion of the emissions. Such work is being carried out by the Cancer Research Center (Moscow) with Zaporozyhe (Ukraine), Alma-Ata (Kazakhstan), Kemerovo, Orenburg (Russia), and Moscow enterprises. However, the lack of standards for vehicular emission of carcinogens does not aid regular work aimed at their reduction. Setting standards, both for carcinogenic and genotoxic content in emissions, and use of physical/chemical studies in conjunction with bio-assays, as well as epidemiological studies are all necessary for the evaluation and reduction of carcinogenic emissions.

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