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INDOOR AND OUTDOOR AIR POLLUTION

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The work carried out at the Institute for Medical Research and Industrial Hygiene in the field of indoor and outdoor air pollution is reviewed, with particular reference to the development of methods for the determination of very low concentrations of air pollutants, to the studies of the mechanism of reactions, and to the interferences of other atmospheric components in the quantitative analysis of air pollution. Personal protective devices developed at the Institute are described as well. The review also includes the results of the air pollution studies of the atmosphere of the City of Zagreb conducted in the last 6 years.

In studying the effect of environment on man the identification of environmental factors and their quantitative assessment is of first and foremost importance. For that, reliable identification methods and methods for the quantitative determination of main environmental factors are to be available.

The studies carried out at the Institute have primarily been concerned with the chemical factors of the atmosphere (air pollution), both in work environment and outdoor atmosphere.

The introduction of ever new, potentially harmful substances into industrial production and everyday use, as well as increasingly stricter criteria for the concentration of air pollutants that can still be considered harmless, call for the development of more and more sensitive methods and for their identification and determination. As the concentration of air pollutants is for the most part very low, and particularly so in the outdoor atmosphere, a number of problems are encountered in their analysis which in macro-analytical chemistry need not be considered at all. The samples get easily contaminated, in which way – owing to very low concentrations of the substances to be analysed – serious errors can easily slip in. A frequently unfavourable relation between the concentration of the pollutants to be determined and the far higher concentration of other air components often produces interferences in the determination of pollutants. Methods for the determination of air pollution should therefore be very specific or at least selective, or elimination should be secured of all interfering components from the reaction mixtures.

Furthermore, as the composition of industrial atmosphere, and particularly of the outdoor atmosphere, varies a great deal, a considerable number of measurements for a representative assessment of the air quality is necessary, and for this reason the measurement methods, in addition to being specific and selective, should also be simple, quick, and cheap, and air sampling should be just as simple, efficient, and representative.

For all these reasons most of the studies of the Institute's Environmental Hygiene Department have been focused on the methodological problems of air pollution. They have comprised:

– Studies of the efficiency of air pollution sampling under different conditions;

– Development and evaluation of methods for the determination of low concentrations of air pollutants;

– Studies of the reaction mechanisms and the interference of other substances simultaneously present in the atmosphere affecting the reliability of the air pollution determination;

– Application of developed and verified methods in the study of environment and its effect on people's health;

– Development of devices for personal and technical protection.

1. INORGANIC GASES

In the early phases of air pollution studies special attention was paid to the introduction of techniques for the preparation of gaseous mixtures of known concentrations and the use of these techniques for the checking of the sampling efficiency and determination of atmospheric gases (carbon monoxide, sulphur dioxide, hydrogen sulphide) (1). These techniques were later improved and used for a detailed study of new methods for the determination of nitrogen oxides and sulphur dioxide in the air (2).

A large experimental gas chamber (10 m^3) for the static preparation of the known concentration of gases and vapours in the air has been constructed (3, 4).

1.1 Nitrogen oxides

Though very sensitive and specific, the method for the determination of nitrogen dioxide in the air based on the formation of an azo-dye, has a basic drawback: the amount of nitrogen dioxide determined is in no accord with the theoretically expected amount (50%). Although this problem has been studied very extensively, no definite answer has as yet been given as to exactly how much of nitrogen

dioxide is transformed into nitrite which further takes part in the production of the coloured reaction, nor has a satisfactory theoretical explanation been proposed for such an irregular behaviour (5, 6, 7, 8). In our early work on the influence of light and temperature on the development and stability of the azo dye, the efficiency of various bubblers for the sampling of nitrogen dioxide, optimum air flow, the effect of temperature on absorption, and the use of three different reagents, about 60% of nitrogen dioxide in the calibration mixture were determined in most cases (2, 9). In the course of the experiments a certain relationship was observed between the concentration of nitrogen dioxide in the air and the relative amount of the nitrite determined. As the results were considerably scattered, efforts were made by the parallel determination of the part of nitrogen dioxide that in the absorption solution is transformed into non-active nitrate and the part that is present in the form of active nitrite, to get some information on the exact percentage of nitrogen dioxide that is transformed into nitrite at various concentrations, that is independently of the experimental error in the preparation of the gas mixture and sampling. Still, the results were more scattered than expected, which suggests that the relationship might be influenced by some more factors. Yet, the negative correlation between the absolute concentration of nitrogen dioxide in the air and the relative concentration of nitrite in the solution was quite obvious; it may be explained by the oxidation rate decrease with the decreasing concentration (10).

The problem of an efficient oxidation of nitrous oxide into nitrogen dioxide for the determination of nitrous oxide by the same method was studied as well (11).

In the course of this work a study of the reliability of visual colorimetry was undertaken, and a logarithmic scale of standard solutions for the visual comparison of colour intensity proposed (12).

Although the method for the determination of nitrogen dioxide, based on the azo dye formation, is specific, the reaction may be interfered by the presence of sulphur dioxide. The extent, mechanism, and prevention of this interference were investigated and the results have shown that the interference occurs only when the samples are stored before measurement for a long time. Consequently, it was concluded that sulphur dioxide affected the already formed colored compound (13).

1.2 Suphur dioxide

A very sensitive, specific method for the determination of sulphur dioxide in the air is based on the formation of a coloured compound by the reaction between sulphur dioxide and reduced para-rosaniline in the presence of formaldehyde (14).

A thorough study of the reaction included the development of the dye at various temperature and light conditions, the effect of the origin of the reagent on the efficiency of the method, the sampling efficiency with various collectors, the influence of air flow on sampling efficiency, the optimum interval between sampling and analysis, and the interference of nitrogen dioxide in the determination of sulphur dioxide and the prevention of this interference. It has been proved that contradictory literature data stem from the fact that the interference does not occur during sampling and that nitrogen dioxide affects only the already formed coloured compound, and for this reason the interference depends on the amount of nitrogen dioxide retained in the absorption solution during sampling. As the trapping efficiency of nitrogen dioxide greatly depends on the type of the collector and is comparatively low with the common bubblers successfully used for sulphur dioxide sampling, the effect of nitrogen dioxide is considerably less significant than one would expect if concluding on the basis of the experiments with pure solutions and taking into account only the concentration of sulphur dioxide in the air. The results of the experiments with controlled gas-air samples of known concentrations confirmed this hypothesis (15).

1.3 Cyanides

The correlation has been studied between low concentrations of cyanides in the air and the amount of the thiocyanates produced by the detoxication of cyanides in the organism of exposed persons and excreted in the urine. A modification of the method for the determination of thiocyanates after *Bruce, Howard* and *Hanzal* has been proposed (16).

By determining the normal values of thiocyanates in the urine of smokers and non-smokers the excretion in smokers was found significantly higher. In view of the difficulties regularly encountered in the field collection of 24-hour urine samples, the possibility of the use of the morning urine instead of the 24-hour sample was investigated; it was found that there was no significant difference in the concentration of thiocyanates between the morning and the 24-hour urine samples (17, 18).

1.4 Carbon monoxide

Within the studies of the effect of low carbon monoxide concentrations, normal carbon monoxide values in the blood of smokers and non-smokers were determined. It was found that the normal carbon monoxide values in the blood of smokers were significantly higher than those in the blood of non-smokers but that there was no significant difference between carbon monoxide values in the blood of urban and rural populations, or between men and women (19).

2. ORGANIC POLLUTANTS

2.1 Organic solvents

The basic problem in the determination of the concentration of organic vapours in the air is the efficient collection of representative samples. Very extensive investigations have been conducted of the optimum air flow, temperature, type of collectors, and the number of collectors connected in series for the maximum, reliable sampling efficiency of a number of chlorinated hydrocarbons. To this end a procedure has been elaborated and verified for the production of mixtures of chlorinated hydrocarbons and the air of exactly known concentrations (20). A method has been introduced for the analysis of chlorinated hydrocarbons based on the thermal decomposition of the organic compound and the determination of the chlorine released in the form of chloride ions (21). The method was checked in the field. Parallel with this work a system of local ventilation was designed for the protection of persons exposed to chlorinated hydrocarbons, and clinical investigations of exposed workers were conducted as well (22).

A method has been introduced for the quantitative determination of benzene, toluene, and xylene in the air (23).

2.2 Tetranitromethane

A method for the efficient sampling and reliable determination of the very toxic tetranitromethane in the air has been developed. The method is based on the formation of a coloured complex by the reaction of tetranitromethane and benzidine, the colour intensity of which is proportional to the tetranitromethane concentration. A detailed study has been made of the effect of light, the reagent concentration, the ageing of the solution, and other pollutants on the results of the analysis. Special attention was paid to the sampling method (24).

2.3 Combustible hydrocarbons

Sources were studied of very serious errors that may arise when in the analysis of combustible air pollutants metal collectors for air sampling are used. It has been shown that the production of hydrogen, formed by the effect of zinc on the water in the collector, may lead to wrong results when determining methane by means of explosimeters. The concentrations determined in this way may be many times as high as those actually existing in the air (25).

2.4 Suspended particulates

Most suspendend organic particulates in the air occur in the form of smoke: a mixture of carbon, incompletely oxidized organic compounds, and ash, with waste gases and air. The problem of the determination of suspended organic particulates is most frequently encountered in the measurement of smoke in the outdoor atmosphere. As an

index of the outdoor air pollution by suspended particulates, the optical density or reflection of smoke samples on the filter paper is often used. The corresponding gravimetric concentration of atmospheric particulates is obtained by specially prepared calibration curves. Detailed studies have been performed of the relation between optical density and reflection, respectively, and the gravimetric concentration of parallel samples (26). The work on quite a new method for the determination of the total particulate concentration in the atmosphere based on fluorescence quenching is being completed. It has been shown that the particles deposited on the filter paper in the course of sampling quench the fluorescence of certain fluorescent substances with which the filter paper is previously impregnated (rodamine B, berberine-sulphate, leukofor BB, fluorescein sodium). A linear relation has been found between the relative degree of fluorescence quenching and the concentration of total atmospheric particulates when predominantly consisting of black smoke particulates or soot. In this way the concentration of total atmospheric particulates may be determined by measuring flourescence quenching. Studying the same relation in the case of white particles no such favourable results have been obtained (27).

The work on the development of a simple new method for the determination of polynuclear aromatic cancerogenic hydrocarbons in the atmosphere by the ring oven technique using standard smoke samples is in progress.

In view of the difficulties encountered in the sampling of the particles of vegetable origin by the standard electrostatic precipitator, where the particles in some cases deposit also on the outer side of the collecting electrode, two modified procedures have been designed: in one the electrostatic field is shortened by shortening the central electrode, and in the other a flange is fixed onto the collecting electrode changing the air flow patern and preventing, in this way, the undesirable depositing of particles on the outer side of the electrode (28).

A method has been worked out for the fractionation of coal dust by a modified sedimentation technique. Equations have been proposed for the determination of the number of repeated sedimentations necessary for obtaining the adequate degree of the homogeneity of fractions. A series of 6 fractions in the 1–20 μ particle size range can be obtained by this method (29). The relation between the projected and actual surface of particles has been studied too (30). So has the theory of the extinction cross-section coefficient of large spherical particles (31).

3. METALS

Occupational exposure to toxic metals had for years been one of the top problems of industrial toxicology in Yugoslavia, and much of the Institute's activity was taken up by the studies of the effect of metals on the health of industrial workers. Lately more attention has been paid to the effect of metals in the outdoor atmosphere on the gen-

eral population. As there is in the same issue a separate review (32) dealing with the problems of the effect of heavy metals, mention will here be made only of some work from this field.

3.1 Mercury

One of the first problems studied in connection with mercury was the problem of miners' exposure to this metal (33). A photocolorimetric dithizone method for the determination of total mercury in the air was introduced. A Beckman spectral photometer was adapted for direct atmospheric mercury vapour measurements by measuring absorption in the ultraviolet portion of the spectrum. A respirator for the personal protection of workers against mercury vapour and aerosols was designed. In it the retention of mercury aerosols is attained by a mechanical filter and that of mercury vapour by iodized activated charcoal (34). The equipments for the static and dynamic production of the known concentrations of mercury vapour in the air were constructed by means of which the calibration of analytical methods was carried out and the efficacy of the respirator designed evaluated. The respirator proved to quantitatively retain mercury vapour even after more than 300-hour exposures to the mercury concentration 100 times as high as the maximum allowable concentration. A prototype of the respirator was produced and checked in a mercury mine and a mercury smelter (35). A chamber was constructed for the exposure of experimental animals with a dynamic production of known concentrations of mercury in the air. The analytical methods developed and the set-up for introducing mercury vapour into the air have been used for the study of hematological changes in experimental animals exposed to mercury vapour (36). A new iodine-iodide method for the determination of total atmospheric mercury has been developed, paying special attention to the interpretation of the results of the determination of total mercury and the determination of only mercury vapour in the air (37). Absorption and excretion of mercury in miners exposed to different concentrations of mercury have also been studied (38). Special studies were devoted to the effect of various levels of mercury exposure on the activity of some enzymatic systems by which it has been shown that mild mercury absorption produces an increase in the activity of alkaline phosphatase, glutamine-oxalate transaminase, and glutamine-pyruvate transaminase in the human serum (39).

3.2 Lead

Extensive analyses of lead exposure in lead mines and smelters and lead battery plants have been conducted. In this connection various methods for the sampling and determination of lead have been devised. A detailed description of this work is given in the review on the industrial hygiene and pathophysiological aspects of poisoning with heavy metals (32).

3.3 Manganese

A method for the determination of atmospheric manganese has been developed, and studies have been conducted on the exposure of workers in a manganese mine. The work is reported in the above quoted review on the industrial hygiene and pathophysiological aspects of poisoning with heavy metals (32).

3.4 Metals in outdoor atmosphere

With regard to the still unsolved question of the effect of very low concentrations of metals in the outdoor atmosphere on the general population, work has started on the introduction of the ring oven techniques for a quick and cheap determination of metals in atmospheric particulates. For the determination of these very low concentrations of metals expensive instruments, such as spectrographs, spectrophotometers, atomic absorption spectrophotometers, etc., are commonly used. These instruments are, however, far too expensive for a wider network of sampling stations necessary to be set up for the study of the effect of metals in the outdoor atmosphere on large population samples. For this reason work has started to explore the possibility of the use of the cheap ring oven technique for the identification and quantitative determination of very low concentrations of metals. So far methods for the identification of ten metals have been introduced. At the same time work is continuing on a new method for the determination of atmospheric lead by means of the ring oven technique. The method is based on the relative insolubility of lead sulphate in diluted sulphuric acid for its separation from soluble sulphates of some metals and the specific solubility of lead sulphate in the solution of ammonium acetate for its separation from non-soluble sulphates of some other metals. For the quantitative determination lead is transformed into the yellow lead chromate, the colour intensity of which is proportional to the lead concentration (40).

The ring oven technique allows a direct analysis of particle samples on the filter paper, without previous digestion in acids, which represents a considerable saving in time and material. The development and verification of methods for the determination of other metals by the ring oven technique is in progress.

4. PROTECTION AGAINST AIR POLLUTION

In addition to a series of ventilation system projects, and some local exhaust ventilation systems in particular, aiming at the reduction of air concentrations in work environment, attention has also been given to the development of personal protective devices against the effect of harmful gaseous and solid air pollutants.

A hose mask with compressed air has been constructed for the protection of railway engine drivers against smoke in tunnels; the com-

pressed air is obtained from the compressed air reservoir used for train braking (41). The respirator proved successful for the protection against smoke. Later it was modified and examined to serve also as protection against a simultaneous exposure to carbon monoxide up to 2500 ppm, suphur dioxide up to about 10 ppm, and hydrogen sulphide up to 20 ppm. The respirator has proved to give full protection to engine drivers and stokers in tunnels (42).

The respirators »Nebojša« and »Dunav« were adapted to be used as hose masks (43).

The respirator for the protection against mercury vapour and aerosols has already been mentioned.

5. STUDIES OF AIR POLLUTION IN THE TERRITORY OF THE CITY OF ZAGREB

Air pollution in the territory of Zagreb has been measured over the last 6 years. Some measurements have at the same time been used as a verification of laboratory methods in the field.

In the course of a year the amount and composition of deposited matter at 11 sampling stations in the greater territory of Zagreb were followed (44). In the course of 5 years the spatial and seasonal distribution of sulphur dioxide and smoke concentrations in the atmosphere were studied by a continuous collection of 24-hour air samples. The effect has also been studied of meteorological conditions on the diffusion and accumulation of air pollution in the territory of Zagreb (45).

From time to time a continuous registration of sulphur dioxide and smoke fluctuations during day was also conducted. The analysis of these results has allowed the assumption that well-planned individual measurements through shorter time intervals may be an indicator of the average air pollution level (46). This hypothesis is being checked. Sporadic collection was made of the gravimetric samples of suspended particulates which was also used for the determination of metals in the atmosphere. Preliminary measurements were conducted of the concentration of sulphur dioxide, carbon monoxide, nitrogen dioxide, formaldehyde, and smoke in a few busy town crossings.

The analysis of all these data has shown that in winter the main sources of the Zagreb air pollution by sulphur dioxide and smoke are domestic space heating systems. The highest sulphur dioxide concentrations were found in densely populated residential areas. A particularly rapid increase in the concentration of sulphur dioxide of an episodic character was recorded in January 1964 when these concentrations surpassed those found in London during the great smogs in 1952 and 1962. Industrial sources of air pollution prevail in summer, but for an adequate assessment of their share in the air pollution of the city of Zagreb, specific emissions of individual industries should be recorded and analysed (26).

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