Correlations between Mutagenic Activity of Organic Extracts of Airborne Particulate Matter, NO\textsubscript{x}, and Sulphur Dioxide in Southern Germany

Results of a Two-Year Study

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Abstract

Goals, Scope and Background. Among other substances, sulphur dioxide (SO\textsubscript{2}), nitric oxide (NO) and nitrogen dioxide (NO\textsubscript{2}) are parameters which are routinely measured to describe basic air quality. Organic extracts of airborne particulate matter contain mutagenic chemical compounds of different origins. The aim of the study was to find correlations between routine monitoring data and mutagenic activity of organic extracts of simultaneously drawn samples.

Methods. Specimens were collected over a period of two years at 8 sampling sites in south-west Germany. Simultaneously, concentrations of NO, NO\textsubscript{2}, and SO\textsubscript{2} were measured on-line within the framework of the official air monitoring network of Baden-Württemberg, Germany. Dust samples were collected for biotesting using high volume air samplers equipped with glass fibre filters. After sampling was completed, filters were extracted and samples were prepared for biological testing. Mutagenic activity was tested by means of the plate incorporation assay (Ames test) using \textit{S. typhimurium} TA98 and TA100 tester strains. During the first year of the study, all tests have been performed with and without metabolic activation. Additionally, a series of tests has been performed in parallel with TA98 and TA98NR.

Results and Discussion. Comparison of Ames test data obtained with and without metabolic activation indicates no statistically significant difference between both methods. Therefore, during the second year of the study, all tests have been performed without metabolic activation. Average yearly activities at the sampling sites were between 1 and 27 Revertants per m\textsuperscript{3} (Rev/m\textsuperscript{3}). High activities were preferentially found at congested sites (Karlsruhe, up to 95 Rev/m\textsuperscript{3}). However, peak values of over 100 Rev/m\textsuperscript{3} were found in other places where pollution by traffic is significantly lower. The reason for these high level values is not evident. Tests performed using TA98NR tester strain indicate a significant share (average 31\%) of compounds requiring activation by nitroreductase for mutagenic activity. Average mutagenic activity can be correlated to routine monitoring parameters. Comparison of averaged data for particular sampling sites indicates significant correlation between nitric oxide and mutagenic activity in TA98 (r\textsuperscript{2}=0.90), while correlation between nitrogen dioxide (0.84) or sulphur dioxide (0.52) and mutagenic activity is weaker. For TA100, correlations are generally weaker than for TA98. Comparison of data for mutagenic activity and routine monitoring data of distant sites being sampled simultaneously shows parallel behaviour.

Conclusions. Results from this study show that mutagenic activity can be compared to seasonal and local variations of gaseous indicator air pollutants. Tester strain TA98 generally shows the best correlations. Although pollution by particle-bound mutagenic substances is significantly higher during the cold season than during summer on average, mutagenic activity of airborne dust is not a continuous effect. During winter, peak levels as well as low pollution periods can occur. Even during winter time mutagenic activity can reach very low levels typical for summertime. Comparison of results for distant sampling sites where samples have been collected simultaneously indicate that ‘classical’ indicators of air pollution and bacterial mutagenicity of organic extracts from airborne particulate matter are influenced by connected effects. Seasonal trend of mutagenic activity, in particular, is similar to the concentrations of nitrogen oxide. NO is a strong indicator for vehicle exhaust gases. It is concluded that the average mutagenic activity at particular sites can be estimated using NO concentrations as an indicator.

Keywords: Air pollutants; airborne particulate matter (APM); Ames test; extractable organic matter (EOM); monitoring; mutagenic activity; nitrogen dioxide; routine parameters; smog; sulphur dioxide; \textit{S. typhimurium} TA98 and TA100 tester strains

Introduction

Air monitoring programmes are the basis of estimations of the public health impact of air pollution exposure and are therefore performed in many countries on a routine basis. Measurements are mostly carried out at fixed site monitoring stations. Data are collected for certain air pollutants like CO, Volatile Organic Compounds (VOC), SO\textsubscript{2}, NO\textsubscript{x}, O\textsubscript{3}, dust, and others. Carbon monoxide, VOCs as well as nitrogen oxides are mainly contributed by on-road vehicles, while SO\textsubscript{2} emissions can be allocated mostly to fuel combustion at electric utilities. Paths for particle emissions include various sources like different kinds of fuel combustion as well as natural sources (Finlayson-Pitts and Pitts 2000). Local concentrations of these compounds are variable and depend on sources of emissions as well as on atmospheric mixing and transport processes. Pollutants are removed from the atmosphere by processes like wet and dry deposition. During severe air pollution episodes, however, meteorological conditions are such that the pollutants are effectively contained in relatively small volumes, leading to high pollutant concentrations. During
winter, smog situations can be initiated by temperature inversions in the atmosphere, thus leading to an increase in airborne pollutants as indicated mainly by increasing sulphur dioxide concentrations, while summer, ‘Los Angeles smog’ episodes are connected to high levels of ozone being formed via nitrogen oxides by photolytic processes.

Recent concern about health effects of air pollution has focussed on particulate matter. While neither the exact damaging agents of environmental aerosol particles have been identified nor are their pathophysiological mechanisms leading to disease fully understood, the health impact seems to be enormous. Particles can act directly or by way of toxic effects caused by compounds adsorbed on their surface. So far, studies performed at various locations worldwide demonstrate that ‘mutagenic activity’ is a common effect present in organic extracts of airborne particulate matter (APM) (Finlayson-Pitts & Pitts 2000). Most of these studies show that mutagenic activity is higher during the cold season than in summertime, and that urban areas are more involved than rural zones (e.g. Binkova et al. 2003). Other studies have shown that mutagenic activity using the Ames test as a tool (e.g. Zwodzianik et al. 2001) or mutagenicity assays based on human cells behaves in a similar way (Pedersen et al. 1999). A study performed over 18 years in Sapporo, Japan suggested that mutagenic activity remained nearly unchanged from 1974 to 1992 (Matsumoto et al. 1998). Traffic is often addressed as being one of the main contributors to mutagenic activity because of the PAH content of exhaust gases. Because traffic flow is more or less constant over the course of the year, increased heating during the cold season is made responsible for the increased mutagenic activity of APM during this season. Therefore, efforts have been made to correct mutagenic activity of APM by using either dust concentration in the air or organic content to calculate mutagenicity per unit. Nardini (1992) found that mutagenicity of organic extracts of APM during winter was increased twofold compared with summer activity, expressed as mutagenicity per milligram. On the other hand, highly variable PAH/mutagenicity ratios suggest that formation and degradation of mutagens in the atmosphere play an important role. Feilberg et al. (2003) showed that air masses from Central Europe are highly enriched in mutagens, and that mutagenic activity is even elevated when these air masses are mixed with local urban air. Additionally, mutagenicity has been attributed to different particle size fractions. Massolo et al. (2002), for example, showed that particles with a mean diameter below 0.49 µm were about one order of magnitude more active than particles in the range of 3.0 to 0.49 µm.

Using conventional parameters as indicators, increasing air pollution is indicated by higher concentrations of certain gaseous pollutants. As mentioned above, it is well known that concentrations of these indicator compounds on average are higher during the cold season as compared to the summer. Alfheim suggested that meteorological conditions might play a role for mutagenic activity of airborne particulate matter too (Alfheim et al. 1983).

Up to now, however, there is little knowledge as to whether these routine monitoring parameters correlate with mutagenic activity, or whether they can be used to estimate or to assess mutagenic activity of airborne particulate matter. Harrison et al. (1997) found a strong correlation between NOx concentrations in the air and particulate matter concentrations during winter time, indicating that road traffic is the main common source for both. Airborne particulate matter consists of particles with different size fractions and airborne particles up to diameters of about 10 µm in a first approach will behave like gases in the atmosphere (Roedel 1994). The aim of our study, therefore, was to evaluate the correlation between mutagenic activity of organic extracts of APM and routinely monitored air pollution indicators.

To this end, data from the highly developed air monitoring programme in southern Germany (Baden-Württemberg), operated by UMEG (Karlsruhe), were compared to the results of laboratory tests for mutagenicity. Samples were taken over two years at eight sampling sites at different locations in Baden-Württemberg and examined using the Salmonella typhimurium plate incorporation assay with TA98 and TA100. During the first year, all analyses have been performed with and without metabolic activation by rat liver S9. Additionally, the share of nitro-group containing compounds in total mutagenicity was estimated for one sampling site using S. typhimurium TA98NR.

1 Materials and Methods

1.1 Sampling location

Samples were collected from January 1993 to December 1994 at eight locations in Baden-Württemberg within the state-wide air monitoring network maintained by UMEG (Zentrum für Umweltmessungen, Umwelterhebungen und Gerätesicherheit Baden-Württemberg). This agency operates a network of 62 monitoring stations all over Baden-Württemberg. Information about the monitoring network is available at <http://www.umeg.de>.

In general, monitoring stations are located in urban areas and reflect a mixture of commercial and residential neighbourhoods. Of the eight sites included in this study, Källbelescheuer (located in the Black Forest) exemplifies a non-urban environment and serves as a ‘background’ monitoring station not being polluted by nearby anthropogenic emission sources. All sampling stations are equipped with systems for the automatic detection of at least NO, NO2 and SO2. All routine data are measured as half hour mean values. A description of the basic localisation and the main influences on air quality at the peculiar station is given in Table 1.

Geographical distribution of sampling sites is depicted in Fig. 1. Mannheim is situated in northern Baden-Württemberg, while Weil upon Rhine is located about 200 km southward at the border to Switzerland. West of the Black Forest, Freiburg was included, while sampling stations Freudenstadt and Källbelescheuer are located in the Black Forest. Reutlingen and Stuttgart are sampling sites east of the Black Forest. The west-east distance of the sampling area was about 80 km. All urban monitoring stations show different traffic loads indicated by average local NOx pollution. Additionally, pollution by SO2 indicates industrial pollution.