Spatial Variability of Air Pollutants in the Basel Area and Carcinogenic and Non-Carcinogenic Health Risk

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Content

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Zusammenfassung

Ausgangslage

In der Umweltepidemiologie spielt die Expositionsabschätzung eine zentrale Rolle. Einerseits bestimmt ihre Qualität die Validität einer Studie, andererseits ist sie ein Schritt in der Risikoabschätzung zur Quantifizierung der Folgen von Umweltfaktoren. In Querschnittstudien werden üblicherweise die Aussenluftmessungen einer Messstation für die Expositionsabschätzung aller Personen des entsprechenden Wohnortes verwendet. Um die Validität dieses Vorgehens zu prüfen, wurde in der Region Basel die räumliche und zeitliche Variabilität von Luftschadstoffen systematisch untersucht. Weiter wurde das damit verbundene kanzerogene und nicht-kanzerogene Gesundheitsrisiko für die Bewohner der beiden Halbkantone Basel-Stadt und -Landschaft (Schweiz) quantifiziert.

Ziele

Messkonzept

1997 wurde eine breite Palette von Luftschadstoffen an sechs Messstationen mit einem mobilen Messwagen gemessen. Der Messwagen wechselte alle 14 Tage den Standort. Die sechs Stationen unterschieden sich hauptsächlich bezüglich der unmittelbaren Verkehrsexposition und repräsentierten typische Wohnlagen in der Stadt Basel. Gemessen wurden Schwebestaub (PM_4 , PM_{10} and TSP), klassische Luftschadstoffe (NO_x, SO_2, CO, O_3, schwarzer Kohlenstoff) und 61 verschiedene VOCs (volatile organic compounds), PAHs (polycyclic aromatic carbons) und Nitro-PAHs, darunter viele kanzerogene Schadstoffe. In die Analyse mit einbezogen wurden zusätzlich Schadstoffmessungen von weiteren städtischen und ländlichen Stationen sowie auch Messungen vom folgenden Jahr. Gleichzeitige Referenzmessungen ermöglichten eine statistische Auswertung der räumlichen Luftschadstoffverteilung an den sechs Messwagenstandorten.

Risikoabschätzungsmethoden

Um das Krebsrisiko durch die Luftbelastung zu quantifizieren, wurden zwei verschiedene methodische Ansätze gewählt. Einerseits wurde eine Methode basierend auf dem Prinzip eines Indikatorschadstoffes durchgeführt. Dabei wurde PM_{10} als Indikator für die Kanzerogenität der Luftbelastung ausgewählt. Dieser integrative Ansatz basierte nur auf epidemiologischen Studien an Menschen. Andererseits wurde eine analytische Methode basierend auf

'Unit-Risk'-Faktoren gewählt. 'Unit-Risk'-Faktoren quantifizieren das Krebsrisiko schadstoffspezifisch und werden traditionellerweise zur Krebsrisikoabschätzung verwendet. Sie basieren vorwiegend auf tier-experimentellen Studien, teilweise auch auf humanepidemiologischen Studien.

Räumliche Verteilung in der Stadt

Die Analysen der Immissionsmessungen ergaben, dass PM_4, PM_{10} und TSP innerhalb der Stadt Basel im allgemeinen homogen verteilt waren. Mit Ausnahme eines Standortes in einer Strassenschlucht neben einer Lichtsignalanlage hatte die Verkehrsmenge in unmittelbarer Nähe des Messstandortes nur einen geringen Effekt auf die mittleren Schwebestaubkonzentrationen. Die mittleren PM_{10}-Konzentrationen an den sechs Messwagenstandorten mit unterschiedlicher Verkehrsdichte waren in einem Bereich von ±10% des städtischen Mittelwertes. Höhere räumliche Unterschiede innerhalb der Stadt, bzw. eine stärkere Abhängigkeit von der Verkehrsdichte, wurde bei den meisten kanzerogenen Substanzen, bei der Partikelanzahl sowie bei denjenigen PM_{10}-Inhaltsstoffen, die typischerweise vom Verkehr emittiert werden, beobachtet.

Unterschied zwischen städtischen und ländlichen Stationen

Der Unterschied in der Schwebestaubbelastung zwischen städtischen und ländlichen Standorten war in der kalten Jahreszeit grösser als im Sommer, was mit dem häufigeren auftreten von Inversionslagen im Winterhalbjahr erklärt wird. Generell wurden die grössten Stadt-Land-Unterschiede für Substanzen festgestellt, die primär vom Verkehr emittiert werden, wie Kohlenstoffverbindungen und Blei, die geringsten Unterschiede für das sekundär entstehende Sulfat und Ammonium.

Zeitliche Variabilität

Im Mittel konnte ein klarer Wochengang in der Schwebestaubbelastung beobachtet werden mit den höchsten Werten am Mittwoch und Donnerstag und den tiefsten Werten während des Wochenendes. Das Werktag/Wochenende-Verhältnis war am ausgeprägtesten für elementaren Kohlenstoff sowie für typische mineralogische Substanzen wie Calcium, Titan und Eisen.

Kanzerogenes Gesundheitsrisiko

Mit einer Methode, die auf 'Unit Risk'-Faktoren basiert, wurde geschätzt, dass in den beiden Basler Halbkantonen jährlich 0.8 (Schätzbereich: 0.2-11.4) Krebsfälle pro 100'000 Einwohner auf den Einfluss der Luftbelastung

zurückzuführen sind. Mit der Berechnungsmethode, basierend auf PM_{10} als Indikatorschadstoff, wurden 6.7 pro 100'000 (95%-CI: -0.8 bis 14.2) jährliche Krebsfälle durch Luftschadstoffe geschätzt. Es zeigte sich, dass Schätzungen, die auf Studien an Menschen beruhen, generell höhere Risiken ergaben. Wenn das Krebsrisikos von Dieselruss in der Aussenluft nur auf der Basis von Arbeitsplatzstudien, ohne Berücksichtigung der Tierexperimente, quantifiziert wurde, ergaben sich 11.4 (95%-CI: 3.1-19.7) Krebsfälle pro 100'000 Personenjahre in der Studienregion.

Nicht-kanzerogene Gesundheits-risiken

Insgesamt wurde geschätzt, dass in den beiden Halbkantonen 59 (95%-CI: 36-81) vorzeitige Todesfälle pro 100'000 Personenjahre mit der Luftbelastung assoziiert sind. Weiter ergaben die Abschätzungen, dass eine signifikante Anzahl von Spitaleinweisungen wegen Atemwegs- oder Herz-/Kreislauferkrankungen, chronische und akute Bronchitis, Tage mit eingeschränkter Aktivität und Asthmaattacken auf den Einfluss der Luftbelastung zurückzuführen ist.

Schluss-folgerungen

Die bemerkenswerte räumliche Homogenität der mittleren Schwebestaubbelastung in einer städtischen Umgebung bedeutet, dass mit Hilfe einer fest installierten Messstation die Exposition aller Bewohner einer Ortschaft zuverlässig abgeschätzt werden kann. Die kleinräumige homogene Verteilung ist primär auf die lange Lebenszeit der Partikel und die Vielzahl von Schwebestaub Emissionsquellen zurückzuführen. Der Unterschied zwischen den Schwebestaubkonzentrationen am Werktag im Vergleich zum Wochenende kann als Indikator für den Einfluss von regionalen anthropogenen Emissionsquellen interpretiert werden. Der Unterschied zwischen Langzeit-Mittelwerten an ländlichen und städtischen Standorten scheint stärker durch Unterschiede in der Höhenlage als durch die Distanz bestimmt zu sein, was mit der grösseren Häufigkeit langandauernder Inversionslagen im Winterhalbjahr erklärt wird.

Aus der Interpretation der Resultate der Gesundheitsrisikoabschätzung ergaben sich Anhaltspunkte, dass ein beträchtlicher Teil der kanzerogenen Wirkung von Luftschadstoffen durch synergetische Wirkungen im Luftschadstoffgemisch zustande kommt und nicht Einzelsubstanzen zugeschrieben werden kann. Daher wird gefolgert, dass im Zentrum einer zukünftigen,

effizienten Luftreinhaltungspolitik sinnvollerweise die Luftqualität als ganzes steht. Das kann bedeuten, dass beim Vollzug und bei der Bestimmung der Höhe von Grenzwerten die Philosophie des 'Indikator-Konzeptes' der Umweltepidemiologie angewendet wird. Dabei werden Luftschadstoffe hauptsächlich als Vertreter für bestimmte Emissionsquellen interpretiert. In diesem Sinne spielt das kausale Wirken eines Einzelschadstoffes auf die menschliche Gesundheit eher eine untergeordnete Rolle. Durch Reduktion der Indikatorschadstoffe in der Umwelt werden gezielt alle Luftschadstoffe vermindert, so dass implizit auch die synergetische Wirkung des Schadstoffgemisches verkleinert wird. Die geplanten Massnahmen zur Reduktion PM_{10}-Immissionen zeigen, dass sich die schweizerische Luftreinhaltepolitik seit der Einführung des PM_{10}-Grenzwertes schon deutlich in diese Richtung entwickelt hat.

Summary

Initial situation

In environmental epidemiology, exposure assessment is a central topic as its quality determines in large part the validity of a study. Further, it is used in risk assessment to quantify the impact of environmental risk factors. Usually in cross-sectional studies the air pollution measurements, performed at one fixed site monitoring station, are used to assign exposure levels to all study subjects living in the corresponding city. In order to prove the validity of this technique, spatial and temporal variability of air pollutants in the Basel area was systematically investigated. Based on these findings a risk assessment was performed to quantify the carcinogenic and non-carcinogenic health risk that is associated with the occurring pollution levels in the cantons Basel-Stadt and Landschaft (Switzerland).

Objectives

Measurement concept

During 1997 a broad palette of air pollutants was measured at six temporary sites using a mobile monitoring station changing location every two weeks. The six sites differed mainly with respect to the road traffic density and represented typical residential place in the city of Basel. It was measured particulate matter (PM_4, PM_{10} and TSP), classical pollutants (NO_x, SO_2, CO, O_3, black carbon) and 61 different compounds belonging to VOCs (volatile organic compounds), PAHs (polycyclic aromatic carbons) and nitro-PAHs, thereof many carcinogens. Additional, air pollutant data from further urban and rural monitoring stations as well as from the following year were taken into account in the data analysis. Concurrently performed reference measurements allowed a statistical analysis of the spatial variability of air pollutants at the six temporary sites.

Methods in risk assessment

To quantify cancer risk from air pollution, two methodological approaches were performed. On the one hand cancer risk attributable to air pollution was quantified based on the indicator concept, taking PM_{10} as a surrogate of the cancerogenicity of air pollution. This integrative approach was based purely on human epidemiological data. On the other hand unit risk factors were used to quantify the cancer risk with an analytical approach. Unit risk factors express the cancer risk of each single agent and have been traditionally

applied in cancer risk assessment. They are based mainly on animal, partly also on human data.

Spatial variability in an urban environment

The analyses of the air pollutant measurements yielded generally a homogeneous distribution of PM_4, PM_{10}, and TSP in the city of Basel. With the exception of one site in a street canyon next to a traffic light, proximity to road traffic had only a weak tendency to increase the levels of PM. Mean PM_{10} concentration at the six temporary sites with different traffic densities was in the range of less than ±10% of the average urban PM_{10} level. Higher spatial variability in the urban environment and a stronger association to the traffic density, respectively, was found for the particle number, some traffic related components in PM, and for most carcinogens.

Differences between urban and rural sites

Differences in the PM levels between urban and more elevated rural sites were larger during the cold season than during the warm season due to days with persistent surface inversion in winter. Generally, largest urban-rural-differences were observed for primary traffic related substances such as carbonaceous compounds and Pb, smallest differences for the secondary produced sulfate and ammonium.

Temporal variability

Comparing mean PM values by day of the week showed a clear trend with highest values on Wednesday and Thursday and lowest values on weekend. The workday/weekend was most pronounced for mineralogical elements and elemental carbon.

Carcinogenic health risk

The quantification of the cancer risk attributable to air pollution yielded 0.8 (range: 0.2-11.4) annual lung cancer cases per 100,000 persons in the cantons Basel-Stadt and Basel-Land when an unit risk-based approach was used. By means of an indicator-based approach resulted 6.7 (95%-CI: -0.8 to 14.2) cases. Generally estimates based on human data yielded a higher risk. Estimating the cancer risk from diesel exhaust in ambient air based only on human occupational studies without considering animal studies resulted in 11.4 (95%-CI: 3.1-19.7) lung cancer cases per 100,000 person years in the study area.

Non-carcinogenic health risk

In total 59 (95%-CI: 36-81) premature deaths per 100,000 person years were estimated to be associated with air pollution in the both cantons. Further a

significant number of respiratory and cardiovascular hospitality admissions, chronic bronchitis, acute bronchitis, restricted activity days, and asthma attacks had been quantified.

Conclusions

The remarkable spatial homogeneity of long-term mean PM levels reduces the error of assigning data from one fixed monitoring site to all study subjects living in one city as usually done in cross-sectional studies. The homogeneous distribution is mainly caused by the long residence time of particle in the atmosphere and the plurality of emissions source types for PM. The difference between mean PM concentration on workday as compared to the one on weekend can be interpreted as an indicator of the influence from regional anthropogenic emission sources. Difference between long-term PM levels at urban and rural sites seems more to be affected by differences in the altitude than by the distance to the city, due to occurring persistent surface inversions in the cold season.

The health risk assessment generated evidence that a large part of the cancerogenicity of air pollution is caused by synergistic effects of various pollutants in the ambient air mixture and cannot be ascribed to single agents. Thus, it is propagated that in future an efficient air pollution regulatory strategy is rather focussed on the whole air quality than on single agents. This can mean that setting and controlling of air pollutant standards is done based on the philosophy of the 'indicator-perspective' of the environmental epidemiology. If so, air pollutants are mainly comprehended as a proxy of certain emission sources. The causal acting on human health of a single agent is of minor importance. However, reducing the indicator pollutants, decrease strategically all pollutants so that implicitly synergistic effects of the air pollution mixture are diminished. The planned sanctions to reduce PM_{10} levels in Switzerland show that regulatory policy has developed in this direction, yet.

PART I INTRODUCTION

1 Open Issues in Ambient Air pollution Research

Health effects of particles

A body of epidemiologic evidence has emerged which demonstrates a range of health effects due to both long-term and short-term exposure to air pollution at concentrations commonly occurring in ambient air (e.g. Abbey et al., 1993, Ackermann-Liebrich et al., 1997, Braun-Fahrländer et al., 1997, Dockery et al., 1993, Katsouyanni et al., 1997, Pope et al., 1995, Zemp et al., 1999). Whereas in the past the discussion about health effects of ambient air pollution was mainly focused on the classical pollutants such as nitrogen dioxide, sulfur dioxide or ozone, recently several studies found that fine particles are strongest associated to health effects (Schwartz and Neas, 2000, Schwartz et al., 1996, Pope et al., 1999b, Fairley, 1999). Consequently there was an increasing discussion in regulatory decision about new standards for fine particulates in the USA and in Europe. As a result in Switzerland the air quality standard for TSP was replaced by a new standard for PM_{10} in 1998 (LRV, 1985, revised 1998), which aimed to give a better protection of the population from the effects of ambient air pollution.

Gap of knowledge in air quality monitoring

In the mid nineties, however, little was known about the levels of fine particles in Switzerland, as $PM_{2.5}$ and PM_{10} were hardly ever routinely measured. Moreover, few data about the spatial distribution of particulate matter and its chemical composition were available, making it difficult to develop a measurement concept to monitor the PM_{10} standard and to propose reduction strategies.

Gap of knowledge in environmental epidemiology

On the other side in most environmental epidemiologic studies, investigating the effects of long term exposure to particulate matter on health, information on health outcomes and important covariates are collected on an individual level, whereas exposure is assigned on a group level, commonly based on data from one fixed monitoring station per study area. The appropriateness of this method depends to a large part on the spatial distribution of the particles. A heterogeneous spatial distribution would lead to a large exposure misclassification of the study subjects affecting the result of a study. Additionally, there is an ongoing debate in environmental epidemiology

whether the mass concentration of fine particulate matter, the particle numbers or some specific compounds of PM, respectively, are best used in air pollution studies (Peters and Wichmann, 1996, Pekkanen et al., 1997). Thus, information about the chemical composition of PM_{10} may help to clarify this issue.

Carcinogenic health risk assessment

Another topic in air pollution research which has been controversly discussed over the last decades is the possible cancerogeneity of air pollution (Speizer and Samet, 1994). Although outdoor air has been shown to be contaminated by carcinogens, the extent of cancer risk attributable to ambient air pollution has remained unclear. Cancer risk has generally been estimated based on toxicological data (e.g. Törnqvist and Ehrenberg, 1994, Kappos and Schmitt, 1993, Yetergil, 1998, Morello-Frosch et al., 2000) or by comparing regions with different air pollution levels (e.g. Katsouyanni and Pershagen, 1997, Hemminki and Pershagen, 1994). Both approaches have its inherent weaknesses. Recent studies found an association between fine particles and lung cancer (Abbey et al., 1999, Dockery et al., 1993, Pope et al., 1995). Even if this association may not be causal, it opens the possibility to assess cancer risk by interpreting PM_{10} as an indicator of the carcinogenic effect of ambient air pollution. Such an approach was successfully applied to assess non-cancer health risk from ambient air pollution (Künzli et al., 2000).

Carcinogenic air pollutants

To compare different approaches for cancer risk assessment information about the levels of cancerogens in a given study area is required. In Switzerland, air quality standards for carcinogenic pollutants do not exist. Thus, carcinogens are not measured routinely and the available data is very limited.

BRISKA

To fill these gaps the Institute for Social and Preventive Medicine of the University of Basel and the Air Quality Management Agency of Basel were both interested to investigate the spatial distribution and the chemical composition of fine particles as well as the levels of carcinogenic air pollutants and the associated carcinogenic health risk. Thus, the BRISKA (Basler Risikostudie Aussenluft) study, which this thesis is based on, was started in 1997.

2 Goal and Objectives

2.1 Goal

The goal of this thesis is to evaluate the spatial variability of air pollutants in an urban environment with a particular focus on particulate matter and to estimate the associated carcinogenic and non-carcinogenic health risk.

2.2 Main objectives

Air pollution analysis

1. Evaluating the temporal and spatial variability of air pollutants.

Specifically to:

- characterize temporal variations of particulate matter concentrations on a daily and a seasonal scale.
- characterize spatial variability of particulate matter and carcinogenic pollutants within an urban environment (Basel) and between urban and rural sites.
- characterize the chemical composition of particulate matter and its temporal and spatial variability.
- interpret influences from meteorology and emission sources on the temporal and spatial pattern of particulate matter concentration.

Risk assessment

2. Assessing health risk of air pollution.

Specifically to:

- quantify carcinogenic health risk from air pollutants in the Basel area using an integrative approach with PM_{10} as an indicator of the cancerogenicity of air pollution and an analytical approach based on unit risk factors.
- compare the both applied methods.
- compare carcinogenic risk with non-carcinogenic health risk from air pollutants in the Basel area.
- study methodological issues concerning pooling of epidemiological data.

PART II BACKGROUND

3 Environmental Epidemiology

3.1 Domain of environmental epidemiology

Begin of environmental epidemiology

One of the earliest published environmental epidemiology studies was the report on "the endemial colic of Devonshire" Baker, 1767. For over half a century, physicians had noted that those who drank cider in Devonshire became seriously ill and sometimes died whereas elsewhere cider drinkers were not suffering. Baker concluded that "the cause of this Colic is not to be sought for in the pure Cyder; but in some either fraudulent, or accidental adulteration". Investigations of the apparatus in various counties revealed that lead was used in Devonshire but not elsewhere resulting in detectable lead concentration only in the cider from Devon. In this study the epidemiologic method was simple: regional differences in disease rates were observed and a necessary cause was identified (Hertz-Picciotto, 1998).

Environmental factors

Often environment refers to all nongenetic factors. It includes physical, chemical, and biological agents as well as social, political, cultural and engineering or architectural factors (Hertz-Picciotto, 1998). In the recent period, the focus of environmental epidemiology has put great emphasis on chemical and physical agents such as volatile organic compounds, metals, particulate matter, pesticides, and radiation. With this focus on chemical and physical agents, there is a strong link to occupational epidemiology. Where adverse effects are observed at high exposure levels commonly found in work settings, the possibility of similar effects at lower exposures in the community has generated concern from the lay public and health scientist.

3.2 Causation in epidemiology

Cause-effect association

A primary objective of epidemiological investigations is to describe an exposure-response association that is unlikely to be explained by extraneous differences between the study groups, thereby elucidating cause-effect relationships. However, random errors or bias are possible non-causal reasons, which can affect the study result (Marsh, 1995).

Random error

Random error is a result of the fact that in epidemiological studies a sub-sample of the entire source population is investigated. Therefore, it is possible that this sample is randomly not representative for the whole source population. Random errors are not systematic or differential. A measure of it is for instance the confidence interval: the wider it is, the less accurate is the study.

Bias

Bias is a systematical error resulting from methodological features of study design and analysis and can be classified in the following primary three types.

Confounding Bias

First, confounding bias, or confounding, arises from the failure to account for (or control for) the effects of other factors related to both, the exposure and health outcome. A confounding variable must be a risk factor for the disease, it is associated with the exposure in the study population, and it is not an intermediate step in the causal pathway between exposure and disease. Confounding can be minimized by controlling for it in the study design and the statistical analysis. Unfortunately, it is never possible to know the effects of all potential confounders. Confounding is inevitable in all studies, because it is a basic characteristic of nature (Dockery, 1993).

Selection bias

Secondly, selection bias refers to a distortion in the estimate of effect resulting from a systematic bias in the manner in which subjects are selected for the study population. Selection must be related to both exposure and disease for selection bias to occur. It can enlarge or decrease the true exposure-response effect.

Information bias

Finally, information bias, or misclassification is related to the instruments and techniques used to collect information on exposure, health outcomes, or other study factors. Nondifferential information bias occurs when the likelihood of misclassification is the same for both groups being compared. When an effect exists, bias from nondifferential misclassification is always in the direction of no effect, and thus is of particular concern in studies that show no association between exposure and disease. Differential information bias occurs when the likelihood of misclassification is different for each comparison group. It can bias the observed effect estimate either toward or away from the null value.

3.3 Overview of study designs and analytic tools in environmental epidemiology

Experimental studies

Experimental studies are the methodological easiest way to investigate the causal effect of an environmental factor. One distinguishes between clinical and field trials. A clinical trial is an experiment with patients as subjects whereas a field trial deals with subjects who have not yet gotten the disease. Usually such trials are conducted blinded, ideally double blind, i.e. the individual who makes the assignment, the study subject, and the assessor of the outcome should all be ignorant of the treatment assignment. Blinding prevents certain biases that could affect assignment, assessment or compliance.

Non-experimental studies

Experimental studies are often not very feasible in environmental epidemiologic. On the one hand it is unethical for an investigator to expose a person to a potential cause of disease simply to learn about etiology. On the other hand experimental trials are mostly not eligible to study long-term effects.

Cohort studies

The most intuitive study design in epidemiology is the classic cohort study. Two or more groups of people are observed that are free of disease and that differ according to the extent of their exposure to a potential cause of disease. This study design allows investigating many health endpoints, both in absolute and relative metrics. However, it is expensive and time consuming, especially for rare diseases.

Case-control studies

In case-control studies, cases of the disease of interest are identified and their exposure status is determined. Further a control group from the entire source population is chosen independently of their exposure status. This permits to estimate the relative size of the incidence rates among exposed and unexposed. Case-control studies are more cost-efficient than cohort studies especially for rare diseases. It can be studied only one disease but many different exposures. However, only relative risks can be straightforwardly estimated. The estimate of absolute measures of risks uses further information about the distribution of disease in the source population.

Case-crossover studies

In recent years many variants of case-control studies have been described. One that is compelling in its simplicity is the case-crossover study (Maclure, 1991). Analogous to the design of crossover studies two (or more) interventions are compared, with each study participant acting as his or her own control. It has been shown that this study design is also feasible in environmental epidemiology to investigate short-term effects of air pollution (Navidi, 1998).

Ecologic studies

A third type of study is the ecologic study (sometimes known as geographical correlation study). In contrast to the cohort and the case-control studies, which deal with individual data, the unit of observation in an ecologic study is a group. The only requirement is that information on populations such as schools, factories, and cities is available for the exposure and disease distribution in each group. However, group aggregated data are insufficient to control for confounding in the analysis and moreover, the degree of association between exposure and disease need not reflect individual-level associations (Greenland and Robins, 1994). Despite such problems ecologic studies can be useful for detecting associations, which may be worthy for further investigations.

Prospective vs. retrospective studies

Studies can be classified as either prospective or retrospective. In a prospective study exposure and covariate measurements are made before the cases of illness occur, in a retrospective study these measurements are made after the cases have already occurred. Early writers referred to cohort studies as prospective studies and case-control studies as retrospective studies because cohort studies usually begin with identification of the exposure status and then measure disease occurrence, whereas case-control studies usually begin by identifying cases and controls and measure exposure status. The term prospective and retrospective, however, are more useful to describe the timing of disease occurrence with respect to exposure measurement (Rothman and Greenland, 1998a). However, the prospective/retrospective distinction is sometimes also used to refer to the timing of subject identification, rather than measurement of exposure and covariates. With this usage, a retrospective (or historical) cohort study involves the identification and follow-up of subjects after their follow-up period has ended.

Cross-sectional vs. longitudinal studies

Another distinction can be made between cross-sectional and longitudinal studies. Traditionally an epidemiologic study is considered to be longitudinal if the information obtained pertains to more than one point in time, whereas in a cross-sectional study persons are classified relative to disease and exposure status at the same point in time (Rothman and Greenland, 1998a). In this sense a cross-sectional study (sometimes known as prevalence study) is comprehended as a survey and is not appropriate to establish an exposure-response association. In environmental epidemiology the terms of cross-sectional and longitudinal studies were often used less restricted. If so, cross-sectional studies are meant to compare data on a spatial scale, i.e. comparing communities with higher pollutant levels to ones with lower levels. Indeed, often the exposure measurements are performed at one point (or interval) in time. However, presuming that the exposure situation remains qualitatively unchanged over a longer period, which may be often true for the air pollution situation in communities, the cross-sectional studies allow investigating long-term effects of air pollution. In contrast, in environmental epidemiology longitudinal studies are often apprehended as time-series studies, i.e. the short term influence of a time varying exposure situation is analyzed within one community using the statistical time-series techniques.

3.4 Short history of air pollution studies

First generation studies

The literature on the health effects of air pollution can be divided into three periods (Hertz-Picciotto, 1998). The first generation of studies was concerned with the health impact of incidents with extremely high levels of air pollution triggered by meteorologic inversions, e.g. in the Meuse valley of Belgium in 1930 (Firket, 1931), in Donora, Pennsylvania, 1948 (Schrenk et al., 1949) and in the London winter of 1952 (Logan, 1953). This first generation of studies was based on a methodological before-and-after design, ideally to investigate effects that have a short latency.

Second generation studies

The second generation of studies, conducted in the 1950s and 1960s, was concerned with extreme levels of persistent air pollution, frequently followed a design that compared communities with different pollutant levels (cross-

sectional). The greatest problem facing these between-community studies was the strong correlation between socioeconomic level and air pollution level. Though controlling for socioeconomic level in the data analysis, the possibility of residual confounding due to inadequate characterization of socioeconomic status or other unmeasured factors remained. Despite weakness, the second generation of air pollution studies contributed to the establishment of air standards.

Third generation studies

The third generation of studies addresses even lower levels of air pollutants and more subtle effects. On the one hand these recent studies have relied heavily on within-community time-series analyses that examine the effects of fluctuations in pollutant levels over time in a single region (e.g. Samet et al., 2000a, Moolgavkar, 2000, Katsouyanni et al., 1997, Samet et al., 2000b, Samet et al., 2000c, Daniels et al., 2000). On the other hand between-community comparisons were performed by means of cohort studies, which allowed a better handling of possible confounders such as socioeconomic level or smoking than other study types (e.g. Dockery et al., 1993 Pope et al., 1995 Abbey et al., 1999).

3.5 Air pollution and health

3.5.1 Non-carcinogenic health effects of particulate matter

Introduction

In the following, today's state of knowledge is presented by means of selected key studies and an overview about the actual objectives in this research field is given.

Long term effect on mortality

Two landmark prospective cohort studies reported that chronic exposure to particulate matter pollution increases the risk of premature mortality. The Harvard Six-Cities study (Dockery et al., 1993) estimated a 26 percent increased risk of premature mortality between highest and lowest polluted city corresponding to a difference in the PM_{10} concentrations of 28.3 µg/m³. The American Cancer Society study (Pope et al., 1995) reported a 17 percent increased mortality risk for a $PM_{2.5}$ concentration difference of 24.5 µg/m³. Some critics had argued that these long-term associations were a result of

confounding. However, recent reanalysis of these study data could not confirm this objection (Krewski et al., 2000).

Short term effect on mortality

A short-term influence of daily increases in particulate pollution on daily death rates could be found in many time-series analysis (e.g. Samet et al., 2000a, Moolgavkar, 2000, Katsouyanni et al., 1997, Samet et al., 2000b, Samet et al., 2000c). From over 20 daily time series studies Levy et al., 2000 estimated by means of a quantitative meta-analysis an increase in the mortality rate of approximately 0.7 percent per 10 µg/m³ increase in PM_{10} concentrations.

Lag

The time-series studies were using different assumptions about the number of days following exposure to air pollution that effects will occur. Some studies have assumed that effects occur the day after exposures (1-day lag). However, toxicological evidence suggests that effects of exposure may be observed over several subsequent days. Schwartz, 2000 demonstrated by means of statistical models that assuming effects continue over several days roughly doubles the relative risk of premature mortality compared to a 1-day lag.

Harvesting effect

Some have argued that the association between day to day variations in mortality and air pollution represents a "harvesting" effect, that is, the advancement of death by a few days in people already about to die from other causes. If air pollution advances death of the very frail by only a few days, then you would expect that an increase in daily deaths would be followed by a decrease in deaths within a few days, which could not be confirmed by several analyses (Zeger et al., 1999, Schwartz, 2000b).

Reduction of life expectancy

However, the effect of air pollution on the reduction of life expectancy is still controversial discussed, though some studies found a notable influence (Nevalainen and Pekkanen, 1998). Brunekreef, 1997 concluded from life table calculation a reduction of life expectancy of more than one year due to air pollution. Pope, 2000 calculated 2.5 to 3.1 years reduction of life expectancy according to the used model. Instead of estimating the effect on life expectancy, Sommer et al., 1999 developed a method to assess the number of years lost per premature death. It yielded approximately 10 years lost per victim.

Non-mortality symptoms

Numerous studies have focused on mortality because it is an easy to measure effect for which data is readily available. It is important to note that early deaths represent just the tip of the iceberg of particulate related health effects. It has been shown that for each death there are many more people admitted to the hospital (Schwartz, 1999), and for each hospital admission, many more visits to emergency departments and doctors offices (Delfino et al., 1997, Medina et al., 1997). Similarly, for each patient who visits an emergency clinic, many more experience uncomfortable respiratory symptoms or days when they must restrict their activity, increase their use of medication, or remain indoors (e.g. Ackermann-Liebrich et al., 1997, Zemp et al., 1999).

Cardiovascular symptoms

Recent studies have focused on the effect of particulate matter on the cardiovascular system. Pope et al., 1999a found little evidence of pollution effects on the oxygen carrying capacity of the blood, but observed that a small elevation in pulse rate was associated with a rise in PM_{10} levels. Other studies found that elevated concentrations of fine particulate matter were associated with lower heart rate variability (Pope et al., 1999c, Liao et al., 1999, Gold et al., 2000), the frequency of arrhythmic events (Peters et al., 2000), and higher concentration of fibrinogen (Ghio et al., 2000). All of these parameters are generally changed when suffering from a heart attack or cardiac illness. Further, increases in plasma viscosity were observed during an air pollution episode in Central Europe with elevated concentrations of sulfur dioxide, total suspended particulates and carbon monoxide (Peters et al., 1997). Similar effects of particles on cardiac functions were found in animal experimental studies (Costa and Dreher, 1997, Godleski et al., 2000). The results of these studies may help to better understand the mechanisms of the association between particulate air pollution and cardiovascular mortality, which was found in a number of studies (e.g. Gouveia and Fletcher, 2000, Daniels et al., 2000, Mar et al., 2000).

Children

A growing body of literature demonstrates that infants and children are especially sensitive to the effects of fine particle pollution. Associations between particulate matter concentrations and post-neonatal infant mortality were found (Bobak and Leon, 1999, Loomis et al., 1999, Woodruff et al.,

1997). Further it was observed that birth weight, the most important predictor for neonatal mortality, was influenced by the maternal exposure to sulfur dioxide and total suspended particles during the third trimester of pregnancy (Wang et al., 1997). Increased particle concentrations have been associated with acute reductions in lung function, and increased symptoms such as cough, phlegm production, and sore throat (Braun-Fahrländer et al., 1997, Vedal et al., 1998). Children with asthma were found to be more susceptible to these effects than other children. Further, some studies found increased pediatric emergency room visits for acute asthma associated with particulate matter concentrations (Norris et al., 1999, Tolbert et al., 2000).

Assessment of non-carcinogenic health risk in Basel

Non-carcinogenic health risk for the population living in the Swiss cantons Basel-Stadt and Basel-Land were estimated in the study BRISKA (Röösli et al., 2000b).

Health outcome	Lower 95%-CI	Point estimation	Upper 95%-CI
Total mortality (adults = 30 years)	36	59	81
Respiratory hospital admissions (all ages)	2	23	43
Cardiovascular hospital admissions (all ages)	30	53	80
Chronic Bronchitis Incidence (adults = 25 years)	8	75	133
Bronchitis (children < 15 years)	430	800	1100
Restricted activity days (adults =20 years)	41'900	49'000	55'700
Asthmatics: Asthma attacks (children < 15 years)	260	420	570
Asthmatics: Asthma attacks (adults = 15 years)	560	1120	1650

Table 3-1: Estimates of the non-carcinogenic health risk of air pollution in the Basel area (numbers per 100,000 person years).

For this non-cancer health risk assessment the method and the exposure-response data from Künzli et al., 2000 were used and applied on the exposure

situation of the study area (see Table 3-1). It was estimated that an increase of 10 µg/m³ in the average PM_{10} concentration is associated with 59 premature deaths per 100,000 persons. Further, 76 hospital admissions, and effects on chronic and acute bronchitis, as well as on restricted activity days and asthmatics attacks are related with ambient particulate matter levels.

3.5.2 Carcinogenic effects from air pollution

Introduction

Already in the 18th century, there were some scattered, and largely ignored, reports establishing an association between environmental factors and cancer, for instance cigarette smoking and lip cancer (Sommering, 1795) or soot exposure of chimney sweeps and cancer of the scrotum (Pott, 1775). However, the systematic investigation of cancer risk began earnest in the 20th century.

Biological mechanism

The biological mechanism of the cancerogenesis is complex and still not fully understood. A detailed overview is given in Franks and Teich, 1986 or Farber, 1987. Cancer is a multifactorial disease. Strictly required prerequisite to develop a cancer is a mutation in a somatic cell by a carcinogenic substance or a metabolite of it. Substances acting in this stage are called initiators. The time between initiation and tumor manifestation is called latency. The length of this period is influenced by the promoters (LAI, 1992). The last step in cancerogenesis, the tumor growing is called progression.

Types of cancer studies

Cancer risk of suspicious substances is evaluated using chemical structure analyses, bioassay, toxicological studies, and/or epidemiological studies (Higginson et al., 1992). Often the chemical structure analysis is the first step to evaluate cancer risk of a substance. The chemical structure of a suspicious substance is compared with the chemical structure of known carcinogenic substances. Short-term bioassays are performed in-vitro and in-vivo using different types of cells ranging from prokaryote to human ones. A substance, which causes DNA-damages in bioassays, is designated genotoxic. Toxicological studies allow testing a suspicious substance in a complex organism under controlled exposure conditions. However, the significance of the result for humans is often uncertain due to interspecies differences. Finally, only epidemiological studies on humans can prove doubtless the cancerogeneity of a substance on humans.

Types of epidemiologic cancer studies

There are several lines of epidemiologic research providing evidence about the association of ambient air pollution with lung cancer in the general population: a) migrant studies; b) urban-rural comparisons; c) studies of populations residing near specific point sources; d) case-control and cohort studies; and e) biologic markers study (Cohen, 2000).

Migrant studies

Studies of migrants provide limited evidence in support of the hypothesis that air pollution is associated with lung cancer due to incomplete control for the effects of smoking and occupational exposure. Migrants from countries with higher lung cancer rates and higher levels of air pollution tend to develop lung cancer at rates higher than those of the new country of residence, suggesting that prior exposure was a risk factor (Speizer and Samet, 1994).

Urban-rural comparisons

Urban-rural studies are the first type of study to evaluate the effect of air pollution on development of lung cancer. Most studies found an overall excess risk in the order of 30-50% in the urban areas and larger relative excess risk among nonsmokers (e.g. Katsouyanni and Pershagen, 1997, Hemminki and Pershagen, 1994). The attribution of these results to the effect of ambient air pollution was strengthened by evidence of urban-rural differences in ambient levels of carcinogens such as benzo[a]pyrene or urban-rural gradients in the mutagenicity of airborne particulate matter across the United States (Hannigan et al., 1997, Pedersen et al., 1999). However, the influence of smoking on these findings is controversial (Doll and Peto, 1981). The urban factor may reflect influences instead of, or in addition to, outdoor air pollution. These could include indoor air pollution, patterns of migration occupational exposure or factors related to population density.

Point source studies

In ecologic studies of residential proximity to diverse industrial sources (e.g. petrochemical plants and steel mills) generally an increased rate of lung cancer was observed but in these studies it was not possible to control for confounders at the individual level (Pershagen, 1990). However, Archer, 1990 took advantage of a "natural experiment" by comparing two counties in Utah, similar in many respects: low smoking rates as well as low and nearly equal respiratory cancer mortality rates until a steel mill was constructed during World War II. The subsequent differences in incidences of lung cancer cases

were substantial within about 15 years after the increase in air pollution and have persisted.

Cohort cancer studies

Three American cohort studies quantified the association between lung cancer mortality and air pollution (Abbey et al., 1999, Dockery et al., 1993, Pope et al., 1995). Using PM_{10} as an indicator of air pollution the Adventists and the Six-Cities study found an effect of air pollution on lung cancer mortality, but the Cancer Society study did not. In this latter study lung cancer was associated with air pollution only when sulfate was used as the index of air pollution. These studies are discussed more extensively in part IV of this thesis as they build the base of the cancer risk quantification.

Case-control cancer studies

Most published case-control studies found relative increases of lung cancer risks after adjustment for age, smoking, and occupational exposure similar to those observed in the urban-rural and ecologic studies (Pike et al., 1979, Vena, 1982, Jedrychowsky et al., 1990, Katsouyanni et al., 1990, Barbone et al., 1995, Nyberg et al., 2000).

Biomarkers studies

Biomarkers offer a new approach to quantifying the lung cancer risk associated with air pollution. Potential biomarkers for lung cancer include actual levels of the putative carcinogen in biologic materials, DNA adducts of potential carcinogens or metabolites, and antibodies against such adducts (Schulte and Perera, 1993). This type of study has focused mainly on the carcinogenic effects of PAHs using benzo[a]pyrene as a prototype.

Occupational studies

In order to evaluate the cancer risk from one single agent occupational studies are popular in the field of cancer research. For most substances an occupational situation can be found where the exposure to the specific substance is substantially high. Unfortunately, in the corresponding profession field there are often other pollutants or factors occurring possibly biasing the result. In this context is the 'Healthy Worker Effect' often discussed (Ahlbohm and Norell, 1991). It refers to the fact that a working collective is usually healthier than the general population resulting in a bias to null when exposed workers are compared with the unexposed general population.

Classification of the IARC and EPA

There exists an immense literature of cancer research. As part of the World Health Organization (WHO), the International Agency for Research on Cancer

(IARC) evaluates the cancer risk of pollutants. So far 833 suspicious substances are systematically investigated and classified. The IARC distinguished five categories, whereby agent can mean a single agent, a mixture or exposure circumstance (IARC, 2000):

- Group 1: The agent is carcinogenic to humans. The exposure circumstance entails exposures that are carcinogenic to humans.
- Group 2A: The agent is probably carcinogenic to humans. The exposure circumstance entails exposures that are probably carcinogenic to humans. *(Definite evidence from toxicological study, pronounced evidence from human studies)*
- Group 2B: The agent is possibly carcinogenic to humans. The exposure circumstance entails exposures that are possibly carcinogenic to humans. *(Sufficient evidence from toxicological study, uncertain evidence from human studies)*
- Group 3: The agent is unclassifiable as to carcinogenicity in humans.
- Group 4: The agent is probably not carcinogenic to humans.

EPA's classification system

A similar classification system is employed of the United States Environmental Protection Agency (U.S. EPA). It comprehends the category A ('human carcinogen'), B1 ('probable human carcinogen, limited human data are available'), B2 ('probable human carcinogen, sufficient evidence in animals and inadequate or no evidence in humans'), C ('possible human carcinogen'), D ('not classifiable as to human carcinogenicity') and E ('evidence of non-cancerogeneity for humans') (U.S.EPA, 2000). Table 3-2 lists the IARC and EPA classification of most important substances, which can be measured in relevant concentrations in ambient air.

Agent/mixture/exposure circumstances	IARC class	EPA Class
Arsenic (inorganic)	1	A
Asbestos	1	A
Benzene	1	A
Benzo[a]pyrene	2A	B2
Beryllium	1	B2
1,3-Butadiene	2A	B2
Cadmium (and compounds)	1	B1
Chromium (hexavalent)	1	A
Coal tars / Coal tar pitches	1	not class.
1,4-Dichlorbenzene	not class.	2B
1,2-Dichlorethan	not class.	B2
Diesel engine exhaust	2A	not class.
Formaldehyde	2A	B1
Lead (and compounds)	2B	B2
Nickel (and compounds)	1, 2B	A, B2
Radon	1	not class.
Soots	1	not class.
Tetrachlorethene	2A	not class.
1,1,2-Trichlorethan (Vinyl Trichlorid)	3	C
Trichlorethene	2A	not class.
Trichlormethane (Chloroform)	2B	B2
Tobacco smoke	1	A
Wood dust	1	not class

Table 3-2: Classification of cancer risk of some selected air pollutants.

NOAEL, LOAEL

Whereas the noncancer health effects are often based on the NOAEL- (highest concentration that did not produce any adverse effect) and LOAEL- (lowest concentration that did produce an adverse effect) concept, respectively, cancer risk is generally quantified based on the unit risk concept. The unit risk assumes a linear cumulative exposure-response association without a threshold. It refers to a lifelong exposure (i.e. 70 years) to 1 µg/m³ of the substance of interest. Unit risks of selected substances are estimated and published from the IARC (IARC, 2000), U.S.EPA (U.S.EPA, 1999b), OEEHA, (Office of Environmental Health Hazard Assessment, California) (OEEHA, 1999), and LAI (Länderausschuss für Immissionsschutz des Landes Nordrhein-Westfalen) (LAI, 1992). For some substances (e.g. diesel exhaust) the range of estimated unit risk factors is larger than an order of magnitude.

Unit risk

3.6 Exposure assessment in air pollution studies

Exposure on binary and continuous scale

On the one hand exposure assessment is part of each environmental study, on the other hand it is a step in risk assessment (see chapter 3.7). The validity of an environmental epidemiology study is in large part determined by the quality of the exposure measurements. A crude binary variable is often not adequate in environmental epidemiology because exposure may occur in a large range. Quantification on a continuous scale of exposure will endow a study with greater sensitivity, allow researchers to assess comparability across studies, and can provide the basis for regulatory decision making (Hertz-Picciotto, 1998).

Personal vs. outdoor measurements

In the last years there was an increasing discussion which measure can be considered as the gold standard for exposure to air pollution (Zeger et al., 2000, Kunzli et al., 1997, Zartarian et al., 1997, Ozkaynak et al., 1996, Wallace, 1996, Thatcher and Layton, 1995, Oglesby et al., 2000, Wilson et al., 2000): personal measurements or outdoor measurements or a mixture of both, outdoor measurements which are complemented with further personal information obtained from different sources such as diaries, sample measurements, etc. Intuitively, one tends to consider personal measurements as best exposure measure because people spend a large portion of their day indoor. However, personal measurements are not feasible in large study populations, which are generally needed to find small effects from general ambient air pollution. Moreover, under the perspective of causal mechanism it may not be sufficient, when study subjects would carry the measurement device with them all the time, because these estimated individual measures could be a poor surrogate for individual absorbed doses due to the variability introduced by the human activity patterns and physiologic characteristics (Hertz-Picciotto, 1998). Moreover, such measurements can only be performed during a limited time interval and for a limited number of pollutants. However, the effect of pollutants might be caused by previous exposure. Finally, many people may be concerned about air pollution originating from anthropogenic activities such as road traffic or industrial sources. Mage et al., 1999 demonstrated that from this point of view, outdoor measurements may be a good proxy because

particulate matter concentrations measured at monitoring stations within the community are highly correlated in time to the human exposure to fine particles of ambient origin. A similar high correlation was found by Janssen et al., 1999 between time series measured in classrooms and at outdoor locations. However, generally a low correlation was found between long-term ambient particulate matter concentration and mean personal exposure from a cross-sectional point of view (Linn et al., 1996, Spengler et al., 1985, Ozkaynak et al., 1996). Though, considering substances in particulate matter which are solely emitted from outdoor sources such as lead or sulfate resulted in a high correlation between long-term indoor and outdoor mean values (Oglesby et al., 2000, Mathys et al., 2000).

Berkson model vs. classical error model

Due to these reasons as well as due to feasibility there are many instances in air pollution studies, in which a single measured exposure level is assigned to a large number of people who are all in the same category, for example to all inhabitants of a city. This semi-individual study design (i.e. individual health data and collectively sampled exposure data (Künzli and Tager, 1997)) can be considered an individual-level analysis with measurement error in the exposure variable (Hertz-Picciotto, 1998). The underlying error model is called Berkson model in contrast to the classical error model, where erroneous exposure measurements were performed individually for each study subject (Steenland and Savitz, 1997). In both the classical and Berkson models, one usually assumes a specific distribution for the error term in the model, for example a normal distribution with mean 0 and variance σ_e^2 corresponding to a nondifferential misclassification. If so, it is well known that in ordinary linear regression with the classical error model the estimated exposure response relation is biased to the null value. This bias to the null will generally also hold for log linear regression models. However, if the assumptions of the classic error model are violated because the measurement error is correlated with the true value (differential misclassification), then the linear regression coefficient can be biased in every direction. In contrast, it is less well appreciated that if the Berkson model holds, then the estimate of exposure effect obtained by ordinary linear regression is in fact unbiased when the error is not related to the exposure level (Berkson, 1950). However the standard error is

increased, resulting in less power or precision. On the other hand, when the variance of the errors is not constant, measurement error can lead to bias away from the null value in log linear regression (e.g. logistic, Cox, Poisson regression) when the Berkson error model holds. This would occur if the errors were greater for larger values of the observed variable, a situation that may occur in environmental studies. Of course the study result is also biased when the mean distribution of errors in each site is not equal to 0.

Single agent vs. complex mixture

It is useful to distinguish in exposure assessment between an exposure setting, a complex mixture, and a single agent. Exposure setting is possible in experimental studies and allows studying the health effect under controlled exposure situation. There is evidence from these studies of acute health effects following exposure to sulfur dioxide, nitrogen dioxide, carbon monoxide, ozone, and particulate matter (Hertz-Picciotto, 1998). As a result the emission of these pollutants are regulated in most countries. Epidemiological studies have supported the findings of the controlled exposure studies. However, in real exposure situations, pollutants are mostly highly correlated with respect to both spatial and temporal variability. Thus, it is often somehow tricky to distinguish between the exposure effects of different single agent, because the possibility of confounding by other pollutants than considered cannot be absolutely eliminated. Though efforts have been made to separate the effects of closely associated pollutants by using sophisticated statistics and by identifying unusual areas in which levels of one pollutant are considerably higher than the others (e.g. Samet et al., 2000a), epidemiologic studies are more effective at assessing the effects of combined exposure to a mixture.

This exposure situation corresponds to the exposure of the general population and may be therefore particular relevant to consider.

The exposure proxy concept

Hence, in recent years the use of a proxy for exposure to air pollution has become more and more popular in epidemiologic air pollution studies. In many studies particulate matter has been considered as best proxy to investigate the adverse health effect of anthropogenic air pollution. On the one hand acute air pollution episodes such as the London smog is consisting primarily of particulate matter and sulfur air pollution yielding an anchor of certainty

that particulate air pollution causes adverse health effects in the high-dose region of the exposure-response curve (Dockery and Pope, 1997). On the other hand particulate matter is a mixture which is emitted by all anthropogenic activities relevant for air pollution such as road traffic, oil burning, industrial processes and so on; and a causal association of particulate is supported by plausible biological effect mechanism models (Pope, 2000).

3.7 Risk assessment

Objectives in risk assessment

Environmental risk assessment is the quantification of potential adverse health effects of human exposure to environmental hazards. In this sense quantitative risk assessment plays a major role in the setting of occupational and environmental standards or exposure limits (Hertz-Picciotto, 1995a). Moreover, one of the important products of risk assessment, in addition to the quantitative results, is the identification of critical gaps in our knowledge (Wright et al., 1997). The definitiveness of a risk assessment is determined by the assumptions, which has to be made based on the current knowledge. Unawareness of the causal and biological mechanism results in unexplainable differences between studies, uncertainty about the form of the exposure-response association, uncertainty about the relevant pollutants, and so on.

Steps in risk assessment

Risk assessment comprises four steps (Gold et al., 1992). First, hazard identification evaluates whether previous research indicates that the exposure may harm human. Second, exposure assessment identifies the specific agents, determines the route of human exposure, and quantifies the amount and duration of exposure. Third, exposure-response assessment uses published data and then extrapolates to a (usually) lower environmental exposure. Fourth, risk characterization combines exposure assessment with exposure-response assessment to quantify, for a defined population, the predicted risk.

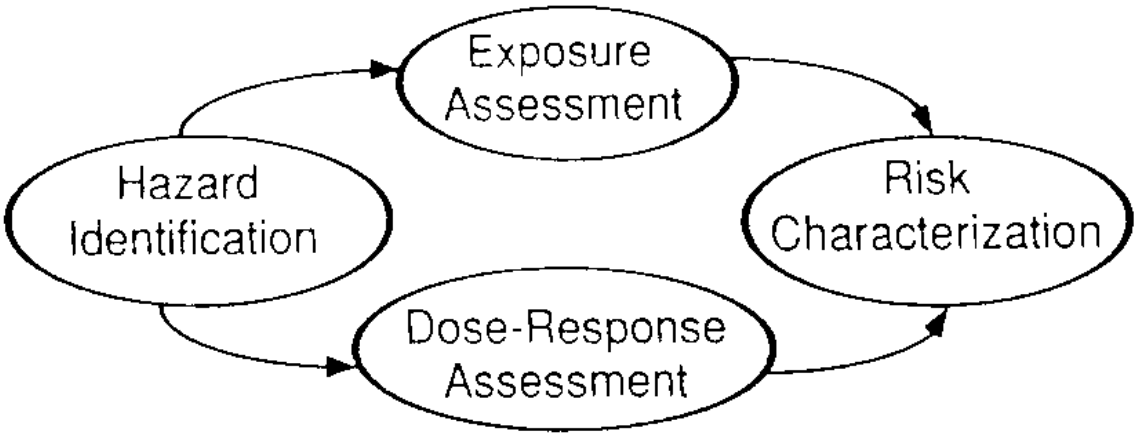

Figure 3-1: Schematic diagram for the four stages of risk assessment (from Hertz-Picciotto, 1995a)

Animal vs. epidemiological data

Risk assessment can be done with experimental animal or epidemiologic human data. Although epidemiologic data are considered the most convincing evidence of human risk, risk assessments are often performed based on animal data because adequate human data to quantify the exposure-response association are often lacking. There are a number of advantages and disadvantages in conducting risk assessment based on either type of data (Smith, 1995, Hertz-Picciotto, 1995b, Wright et al., 1997, WHO, 2000). Briefly, animal studies are conducted under well-controlled exposure situations, which allow estimating the exposure-response association quantitatively. However, the exposure dose is often orders of magnitude higher than typical human exposure, the intake pathway is often different (injection instead of inhalation), and it is not known whether exposure to a single agent is representative for human exposure to a complex mixture of air pollutants.

Interspecies differences

Moreover it is impossible to quantify the uncertainties from interspecies difference when results are transferred to humans. For instance, for site specific carcinogenic response between rats and mice was found only a correlation of 37% analyzing 379 long-term experimental studies (Haseman and Lockhart, 1993). In fact, no significant interspecies correlations were observed for lung tumors, which are a common concern in health risk assessment of air pollution. Extrapolating causation even between rodent species is difficult, so it is expected that extrapolating actual cancer risk from

rodents to humans would involve larger degrees of uncertainty.

Epidemiologic studies

The advantage of epidemiologic studies is that they provide direct evidence for carcinogenic effects in humans, thus avoiding the uncertainty of interspecies extrapolation. However, they do not show cause- and-effects relationships with the same ease, as do experimental studies in animals, because they are subject to a number of biases and potential confounders. Moreover, most studies are retrospective making it difficult to identify and quantify with certainty the critical exposure that may have occurred 20 to 40 years in the past (Wright et al., 1997). Thus, exposure-response association is often estimated only qualitatively, i.e. high vs. low exposed.

Concept of the attributable cases

Every risk assessment constructs an association between a cause and an effect and is therefore based on assumption about the underlying causal model. In epidemiologic research the concept of attributable cases has became popular (Rothman and S., 1998b). In summary, it says that a given case (e.g. lung cancer) is caused by one sufficient cause. This sufficient cause can consist of many causal components, for instance smoking, air pollution, diet, genetic factors, etc which are cooperating altogether to cause one specific cancer case. If one causal component had been eliminated, this specific lung cancer cases had not occurred. However, it is impossible to prove this on an individual level, but applying statistical methods allows investigating the effect of the component cause on a population level. When dealing with attributable cases one must be aware of the fact that the sum of all attributable proportions for all causal components is higher than 100 percent for a multi-factorial disease, because competing risks of the various causal components can not be taken into account. Concretely, according to the concept of attributable cases each cancer case could be prevented several times by eliminating one of several component causes. Statistical approaches which are able to allow for competing risk need to be developed first.

Meta-analysis

Meta-analysis is a tool, often used in risk assessment to combine results across several studies with the goal of estimating measures of association with improved precision. It can be used at the hazard identification stage as well as in the third stage to assess an exposure-response association (Greenland, 1998).

Steps in meta-analysis

There are four steps in a meta-analysis (Petitti, 1994a). First, studies with relevant data are identified. Second, eligibility criteria for inclusion and exclusion of the studies are defined. Third, data are abstracted. Fourth, the abstracted data are analyzed statistically. Generally the result is summarized by a method of weighting by precision of the studies taking the inverse variance.

Analyzing heterogeneity between study results

An important aspect in the last step is to qualitatively and quantitatively analyze any heterogeneity that may be present. This can be done using Cochran's Q-statistic (DerSimonian and Laird, 1986). Under the null hypothesis of no heterogeneity, Q is approximately a χ^2 statistic with k-1 degrees of freedom. If a large amount of unexplained heterogeneity remains, one may consider turning from 'fixed-effect' models to 'random-effect' models. With this approach, both random variation within studies and heterogeneity between studies is taken into account. When heterogeneity is present, the estimated confidence interval is more conservative (i.e. wider) than it would have been on the fixed-effect assumption.

Problems in meta-analysis

Publication bias

One problem in meta-analysis is its tendency to combine studies including a wide range with respect to their quality. A second problem is publication bias, meaning that studies, which did not find an effect, tend not to be published and thus are unavailable. There are many approaches to deal with these problems according to the study subjects; however, there is no ideal, general solution (Wright et al., 1997).

Observational studies in meta-analysis

There exist a controversy whether observational studies are eligible for meta-analysis (Shapiro, 1994b, Greenland, 1994, Petitti, 1994b, Thompson, 1994). A major problem is that purely synthetic meta-analysis can give a false impression of consistency across study results, "offers the Holy Grail of attaining statistically stable estimates of effects of low magnitude" (Shapiro,

1994a). However, Greenland, 1994 believes the solution is to adopt a comparative approach in which meta-analysis is used as an aid in comparing studies and identifying patterns among study results, e.g. analyzing heterogeneity. Hypotheses and biases can be tested without trapping within the framework of one study design. However, one should always address the possibility that all studies have suffered a common systematic error.

4 Particulate Matter

4.1 Overview

Size fractions

Microscopic particles are ubiquitous in our environment. They are produced by natural processes and by human activities. Particulate air pollution is a mixture of solid and liquid particles suspended in the air. The particle size ranges from molecular clusters of 0.001 µm to fog droplets and dust particles as large as 100 µm (Figure 4-1). The size fraction >2µm is usually referred to as the coarse mode, while the fraction below this size is the fine mode. The latter mode can be further divided into the accumulation mode (d ˜ 0.1-2 µm) and the nucleation mode (d < 0.1 µm).

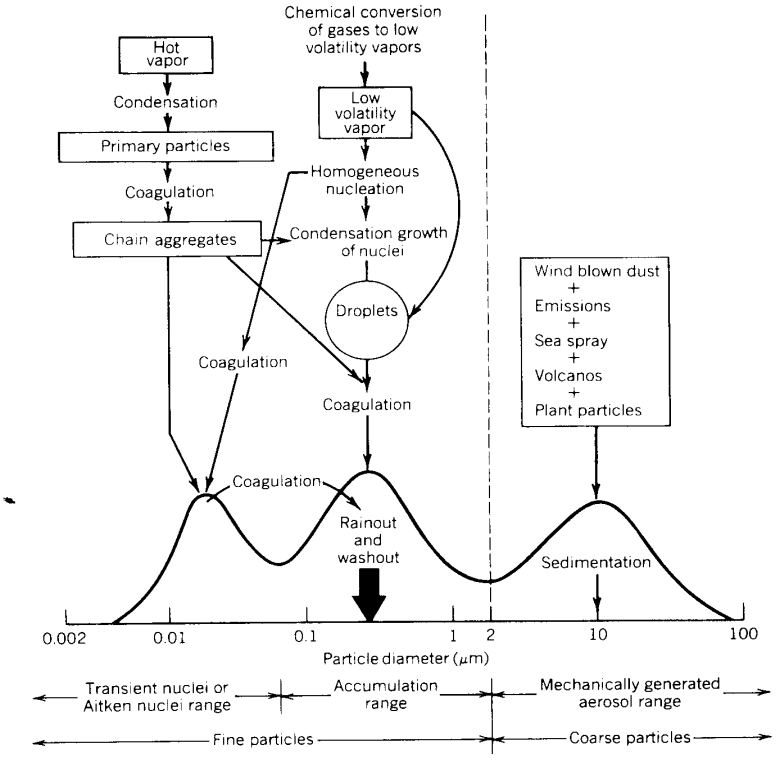

Figure 4-1: Typical size range of atmospheric aerosols (from Whitby and Baron, 1980)

TSP, PM_{10}, $PM_{2.5}$

The aerodynamic particle size is the most important characteristic influencing deposition in the respiratory system. In summary, the smaller the deeper particles are penetrating into the lung (Figure 4.2). A variety of size measures

of particles are used. Analogous to the EPA, Switzerland's initial reference measure for particles was total suspended particulate (TSP) with an upper size limit between 25 and 45 µm depending on the used measurement device. With the time PM_{10} (d < 10 µm) had became more popular, because only particles smaller than 10 µm penetrates into the airways of the lung. In 1998 the ambient air quality standard for TSP was replaced by a standard for PM_{10} in Switzerland (LRV, 1985, revised 1998). A potentially more specific estimate of the etiologically relevant exposure is given by particles smaller than 2.5 µm because they enter into the alveolar region (Dockery and Pope, 1997).

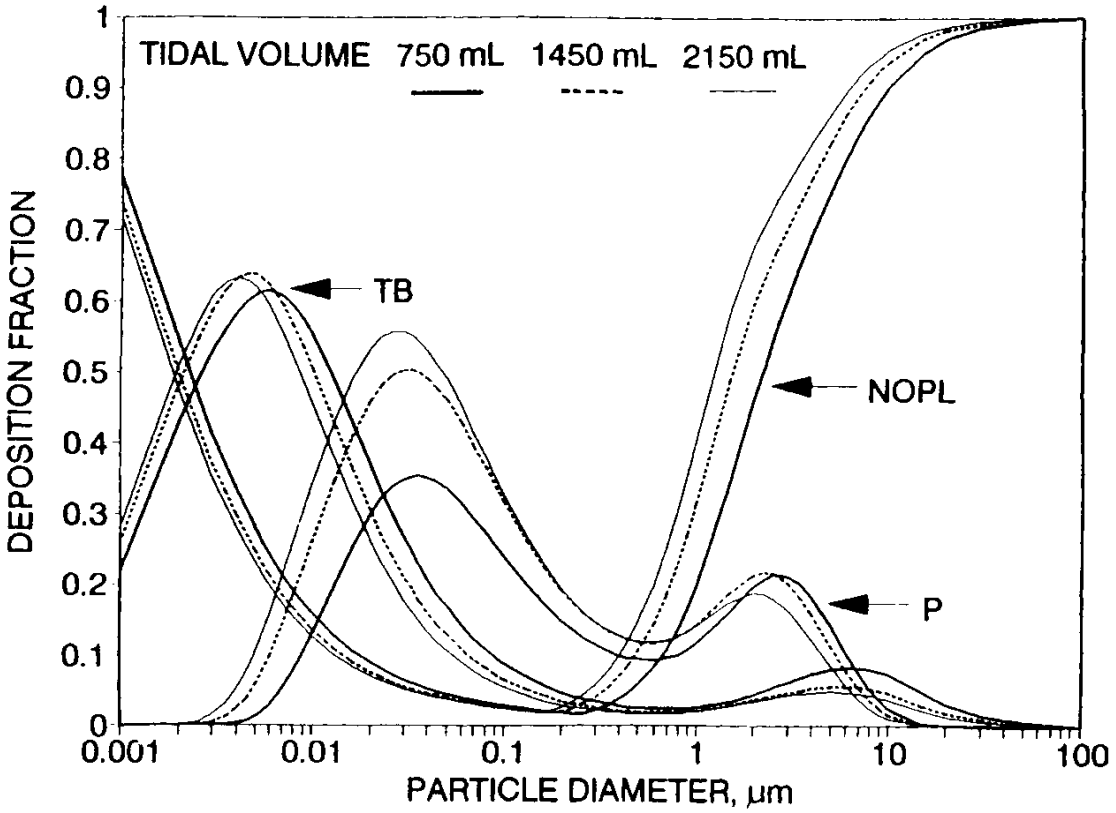

Figure 4-2: Disposition efficiencies in different parts of the respiratory system. "TB" means tracheo-bronchial (alveolar region), "NOPL" means naso oropharynx larynx (region of nose and mouse), and "P" refers to the pulmonary region (airways) (after Phalen et al., 1991, from Dockery and Pope, 1997).

Atmospheric residence time

The particle size determines not only the deposition rate in the lung but also the residence time in the atmosphere. Generally, particle with an aerodynamic diameter of approximately 1 µm has longest residence time, in the order of

one week. Larger particles are faster removed from the atmosphere due to sedimentation; smaller particles are coagulating and are thus present in another size fraction (Figure 4-3).

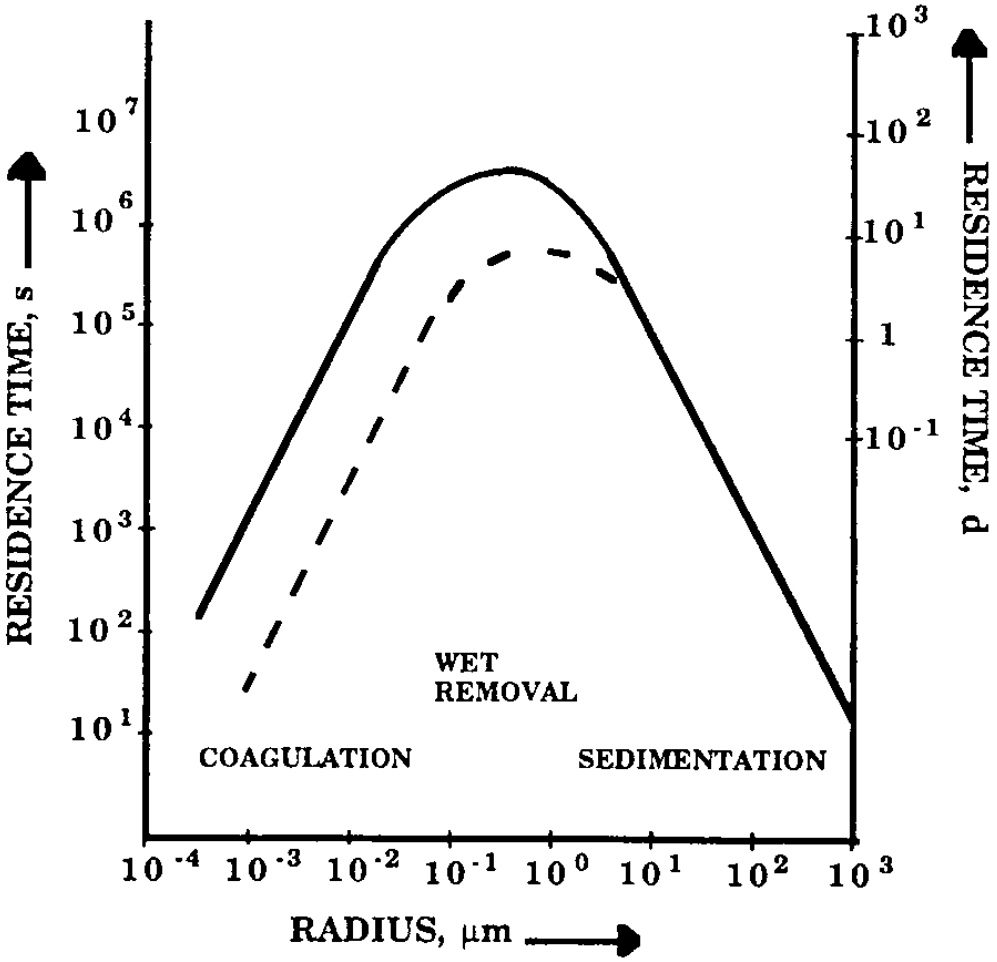

Figure 4-3: Calculated residence time of typical aerosols over continental regions (———) and areas with very clean air (----) (after Jaenicke, 1988 from Hering, 1996)

4.2 Formation and sources of particles

Primary and secondary particles

Ambient airborne particulate matter can be either of primary or secondary origin. Primary particles are directly released into the atmosphere. Secondary particles are formed within the atmosphere by homogeneous or heterogeneous gas-to-particle conversion from gaseous precursor substances such as sulfur dioxide (SO_2), nitrogen dioxide (NO_2), ammonia (NH_3) and volatile organic

carbon compounds (VOC). Most important source of SO_2 is coal and oil combustion, NO_2 originates mainly from road traffic, NH_3 from agriculture as well as partly from cars with catalysator and VOCs are mainly emitted by industrial sources and to a small extent by traffic. Secondary particles are mainly small particles (Baltensperger and Nyeki, 1998).

Sources of primary emitted particles

Particles originating from combustion processes are also small. Combustion is an important source of particulate matter in industrialized regions. Mechanically generated particles are generally larger. Table 4-1 gives an overview about the most important emission sources, their most important emitted substances and the typical size range summarized from Kouimtzis and Samara, 1995, Baltensperger and Nyeki, 1998, Hüglin et al., 2000.

Sources	Typical substance	Typical size range
Secondary produced	SO_4^{2-}, NO_3^-, NH_4^+, OM	nucleation mode
TRAFFIC		
Exhaust	EC, OM, Pb, Br, Pt, Pd, Rh, PAH	nucleation mode
Wire abrasion	Zn, EC, OC	coarse mode
Brake line abrasion	Fe, OC	accumulation mode
Road resuspension	Al, Ca, Fe, Mn, Ti, Ba, Si	coarse mode
Railroad	Fe	accumulation mode
STATIONARY COMBUSTION		
Coal combustion	EC, OM, S, As, Se, Cl	nucleation mode
Wood combustion	EC, OM, K	nucleation mode
Oil combustion	EC, OM, S, V, Ni	nucleation mode
Incineration	EC, OM, Cd, Zn, Cu, Cl	nucleation mode
INDUSTRIAL SOURCES		
Cement/building industry	Ca, Mg, Ti	accumulation/coarse
Steel and metal industry	Fe, Mn, Cd, Cu, As	accumulation/coarse
NATURAL SOURCES		
Mineral dust	Al, Ca, Fe, Mn, Ti, Ba, Si	coarse mode
Biogenic aerosols	K, protein	coarse mode
Sea-salt	Cl	accumulation mode
Volcanic	EC, OM, S	accumulation mode

Table 4-1: EC means elemental carbon, OM, organic matter, and the remaining acronyms are conventional elements.

Natural vs. anthropogenic sources

The distinction between natural and anthropogenic sources of particulate matter is sometimes ambiguous. For instance, smoke aerosol can either arise from wild fires or from manmade combustion processes. Or, mineral dust can be entrained into the atmosphere by wind blown dust from agriculturally eroded regions.

Meteorological influences

The daily measurements of airborne particle matter are not only influenced by the emission source but also to a large extent by the predominating meteorological conditions. On the one hand meteorological conditions affect the production of aerosols like gas-to-particle conversion or wind blown dust as well as the removing from the atmosphere due to aerosol scavenging. On the other hand meteorology influences the import or dilution of polluted air. The efficiency of air dilution is highly depending on the vertical stability of the atmosphere, which is on its part a function of many meteorological parameters such as vertical temperature gradient, wind velocity, radiation conditions, etc. Our analyses showed that the daily variability of particulate matter concentration is much more influenced by meteorological parameters acting on dilution than by meteorological parameters playing a role in forming and removing of the aerosol such as rain and radiation (Röösli et al., 1999). In Figure 4-4 is pictured the percentage change of modeled PM_{10} concentration by an interquartal change of the according parameter.

Deliquescence

Deliquescence, meaning the uptake of water of salty aerosols, is influenced by the atmospheric humidity. In this process hysterese effects plays an important role. This process act on the particulate matter measurements by altering the size distribution of particulate matter as well as the proportion of water.

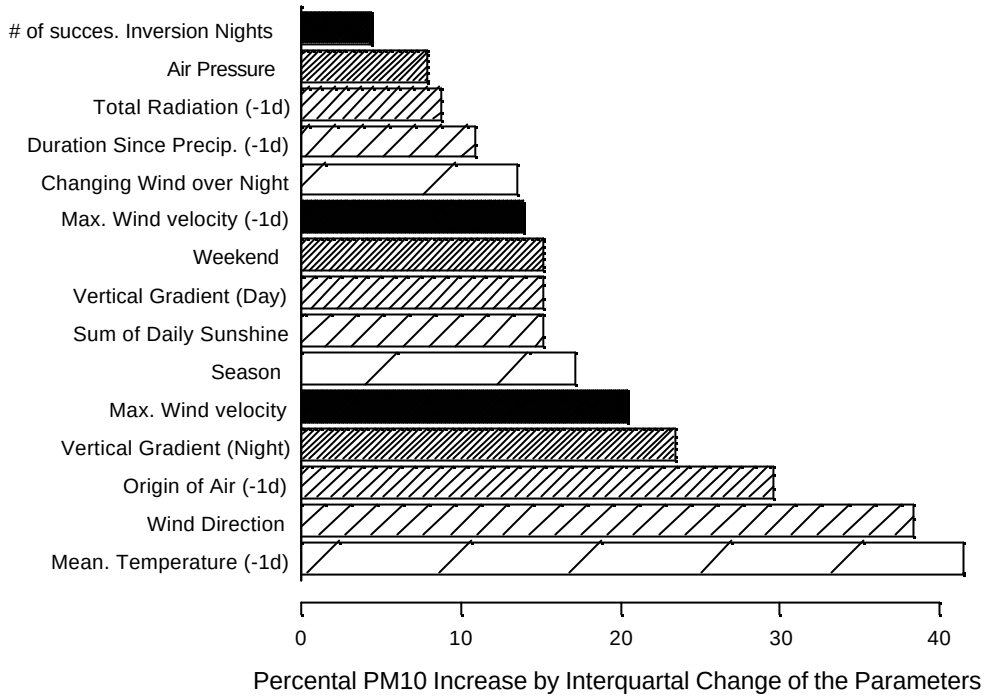

Figure 4-4: Percentage change of modeled PM10 concentration by an interquartal change of the according parameter. (-1d) means that the parameter measured one day before was more significantly associated than the one from measurement day (adapted from Röösli et al., 1999).

PART III GENERAL METHODS

5 Methods

5.1 Air pollution measurements

1997 data

Mobile monitoring measure-ments

The air pollution measurements and the used chemical analytical methods are described in detail in the method sections of Article 1 and 3. An overview is pictured in Figure 5-1. Briefly, during 1997 PM_4 , PM_{10} and TSP was measured at six temporary sites using a mobile monitoring station changing location every two weeks. The six sites were chosen to be representative for the place of residence of the population living in Basel. They differed mainly with respect to the road traffic density. Two sites were typical residential sites with low traffic density, one site was located in the center of Basel with moderate traffic, and three sites were exposed to high traffic with different proportion of heavy-duty vehicles (details see Table 6-1). Further, at the six sites the classical pollutants (NO_x, SO_2, CO, O_3, black carbon) were measured and 61 different compounds belonging to VOCs (volatile organic compounds), PAHs (polycyclic aromatic carbons) and nitro-PAHs, thereof many carcinogens.

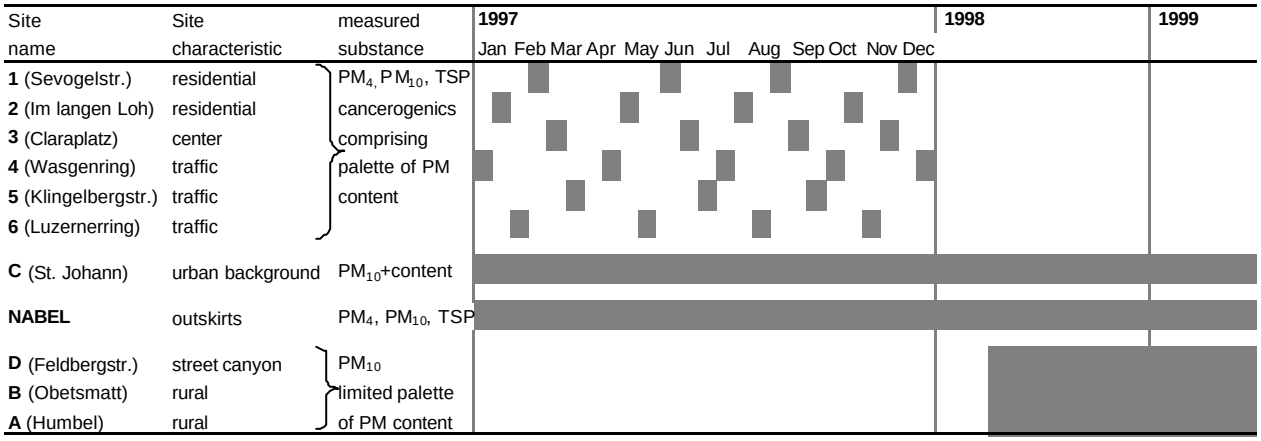

Figure 5-1: Overview of the measurement concept.

Fixed site measure-ments

Additional to the mobile monitoring measurements, between 1997 and May 1999 PM_4, PM_{10}, and TSP was permanently measured in the outskirts of Basel at the NABEL station (Nationales Beobachtungsnetz für Luftfremdstoff) (BUWAL, 1998) as well as PM_{10} in Basel at the fix monitoring station St. Johann of the air quality management agency of Basel (see Figure 5-2).

1998/99 data

Moreover, PM_{10} was daily measured at two rural sites (A and B) and another urban site (D) between April 1998 and May 1999. A limited number of substances contained in PM_{10} were analyzed from these measurements.

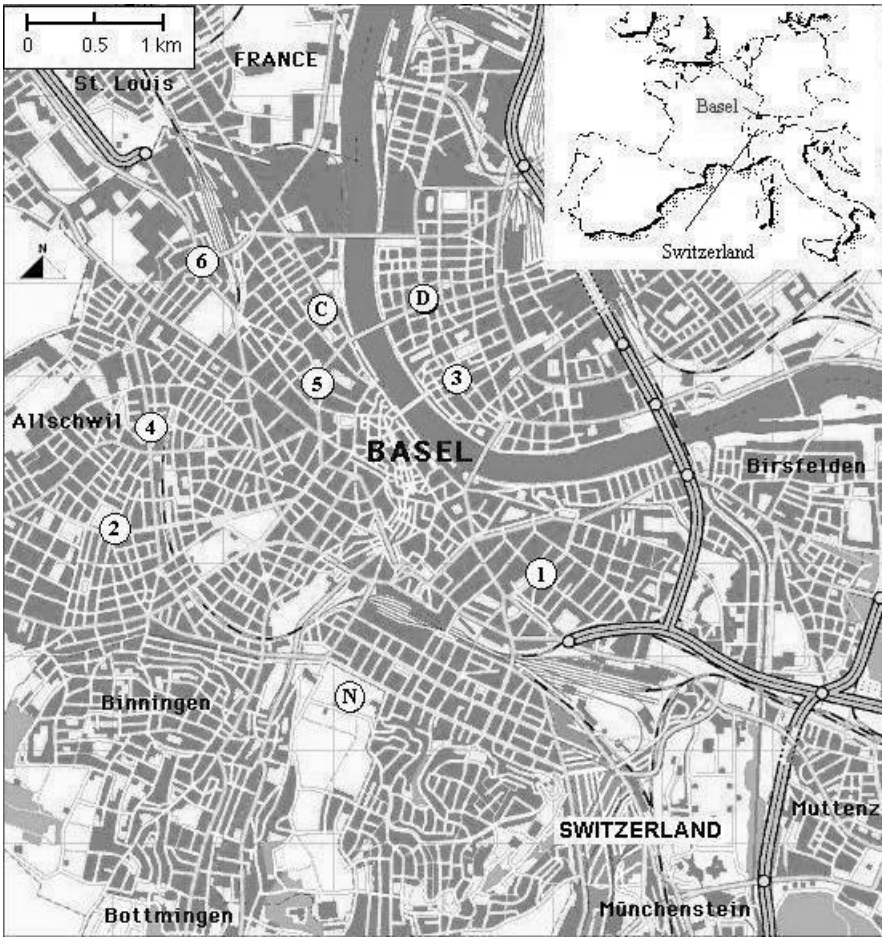

Figure 5-2: Map of the monitoring sites in Basel. Site 1-6 refers to the six temporary sites with the mobile monitoring station. C and D denote St. Johann and Feldbergstrasse, respectively; and N means the NABEL site. (The rural sites A and B are not shown, they are located 20 and 20 km southeast of Basel, respectively.)

5.2 Analysis of spatial variability

Study design

The ideal study design to analyze spatial variability of air pollutants is to place several well-calibrated measurement devices at a number of locations and to measure concurrently. However, for a large palette of pollutants this needs a lot of devices and financial support, respectively. Statistical methods can be used to gain a better cost effectiveness ratio for restricted financial resources. Thus, we applied various statistical approaches to gain a maximum of

information about spatial variability of air pollutants from restricted measurements using a mobile monitoring station and a concurrently performed reference measurement at a fixed site.

Particulate matter within Basel

The particulate matter measurements at the six temporary sites in Basel, performed during 1997 by the mobile monitoring station, could not be directly used to calculate annual means and to assess the spatial variability, as they had not been performed concurrently. Comparing raw particulate matter levels between the sites could have yielded differences in the concentrations of particulate matter simply owing to differing meteorological conditions prevailing during the sampling period, or to temporal varying source strengths such as traffic density, or both. In order to deal with such influences the daily particulate matter measurements from the temporary station were regressed against the respective daily particulate matter measurements from the NABEL station as well as meteorological parameters. Thus, we obtained for each mobile monitoring site and particulate matter size fraction a complete annual time series consisting of measured and modeled values, which were used to calculate comparable annual mean values. The detailed procedure and results are described in Article 1, page 45.

Chemical composition of PM_{10} within Basel

The data about the chemical composition were rather more limited than particulate mass measurements as we were not able to chemically analyze all collected particulate matter filters due to financial restrictions. From 41 days, however, we analyzed PM_{10} filters from the same day, measured concurrently at the fixed urban monitoring site 'C' and one of the temporary monitoring sites. This data were too limited to analyze differences in the chemical composition between the six temporary sites. However, we used them to assess the small-scale spatial variability within Basel. For each substance, we calculated the Pearson correlation coefficients between concurrently performed measurements at the fixed urban monitoring site 'C' and one of the temporary monitoring sites. A high correlation had been interpreted as an indicator for homogenous small-scale spatial variability within the city of Basel. Results are presented in Article 3, page 84.

PM_{10} between urban and rural sites

In 1998 and 1999, 24h hours mean values of PM_{10} were measured at two urban and two rural sites (but not PM_4 and TSP). Spatial variability of these concurrently performed measurements could be directly analyzed. Results are presented in Article 1, page 45.

Chemical composition of PM_{10} between urban and rural sites

From these 1998/99 measurements, a limited number of substances from every fourth filters were chemically analyzed (SO_4^{2-}, NO_3^-, NH_4^+, Cl^-, Pb, Zn, Cd). Based on this sample monthly mean values were determined taking into account the ratio of the mean PM_{10} concentration during the sampling days compared to the monthly mean PM_{10} concentration. These values could directly be compared across the four sites. Results are presented in Article 3, page 84.

'Classical pollutants' within Basel

The analyzes of the spatial variability of the 'classical pollutants' (SO_2, NO, NO_2, NO_X, O_3 and CO) within Basel were conducted in the same way like particulate matter within Basel, i.e. taking into account continuously performed reference measurements from site 'C' (St. Johann) and the NABEL site. These results had not been used for the risk assessment. They are presented in Röösli et al., 1999.

Particle numbers and pPAH.

The spatial variability of particle numbers and particle-bound polycyclic aromatic hydrocarbons (pPAH) were assessed by qualitatively comparing spot measurements from two traffic sites and one typical residential site. In each case measurement were performed during about 26 hours with high time resolution (~5min). Results are presented in Article 2, page 65.

Carcinogens within Basel

The carcinogens were measured on the mobile monitoring car on the base of 13 days value. Reference measurement from site 'C' were available for most VOCs, but not for PAH, nitro-PAH and 1,3-butadiene. For substance with available reference measurements, mean values for each temporary site were estimated based on regression models. Spatial variability of substances without available reference measurements was analyzed only qualitatively. Results are presented in Röösli et al., 1999.

5.3 Risk assessment

Indicator-based vs. unit risk based approach

Two methodological approaches were performed to quantify cancer risk from air pollution. First, cancer risk of air pollution was quantified based on the proxy concept, taking PM_{10} as a surrogate of the cancerogenicity of air pollution. This integrative approach was based purely on human epidemiological data. The second risk assessment approach was based on the unit risk concept. For this analytical approach all relevant carcinogenic air pollutants were identified and their potencies were quantified with unit risk factors. Unit risk factors are based mainly on animal, partly also on human data.

5.3.1 Indicator-based risk assessment

Hazard identification

First step in risk assessment is the identification of potential hazards. In the indicator-based approach air pollution was comprehended as one hazard. This approach is based on the key assumption that PM_{10} is an appropriate surrogate of the carcinogenic potency of air pollution. On the one hand PM_{10} contains some carcinogenic compounds such as diesel exhaust, chromium VI, nickel, PAH, etc. On the other hand most pollutants are correlated in time and space. Thus it may suffice to choose one proxy, though a large part contained in PM_{10} are not expected to be carcinogenic and certain cancerogens are known to be gaseous such as benzene and 1,3-butadiene.

Quantification of the cancerogenicity

Applying the indicator-based method, the cancerogenicity of air pollution was quantified based on the association between PM_{10} and lung cancer investigated in three American long-term cohort studies (Abbey et al., 1999, Dockery et al., 1993, Pope et al., 1995). Based on the concept of the attributable cases, excess rate per 10 µg/m³ increase in average PM_{10} levels was estimated from these three studies using a meta-analytic approach, described in detail in Article 4, page 106.

Exposure assessment

Step two and three in risk assessment comprehends the identification and quantification of the relevant exposure. It was assumed that the relevant exposure of the 450,900 persons (Bundesamt für Statistik, 1997) living in the

Cantons of Basel-Stadt and Basel-Landschaft is equal to the annual mean concentration at their living place.

PM_{10} exposure

PM_{10} population exposure was assessed by using a dispersion model (Heldstab et al., 1999) developed in the tri-national study (Künzli et al., 2000), which is described in detail in (Filliger et al., 1999). The model was based on emission data taking into account primary and secondary particulate (see Figure 5-3).

Quantification of the health risk

Last step in risk assessment comprise the quantification of the health risk. The effect measure expressed as excess rate was multiplied by the population weighted average PM_{10} exposure (see Article 5, page 116).

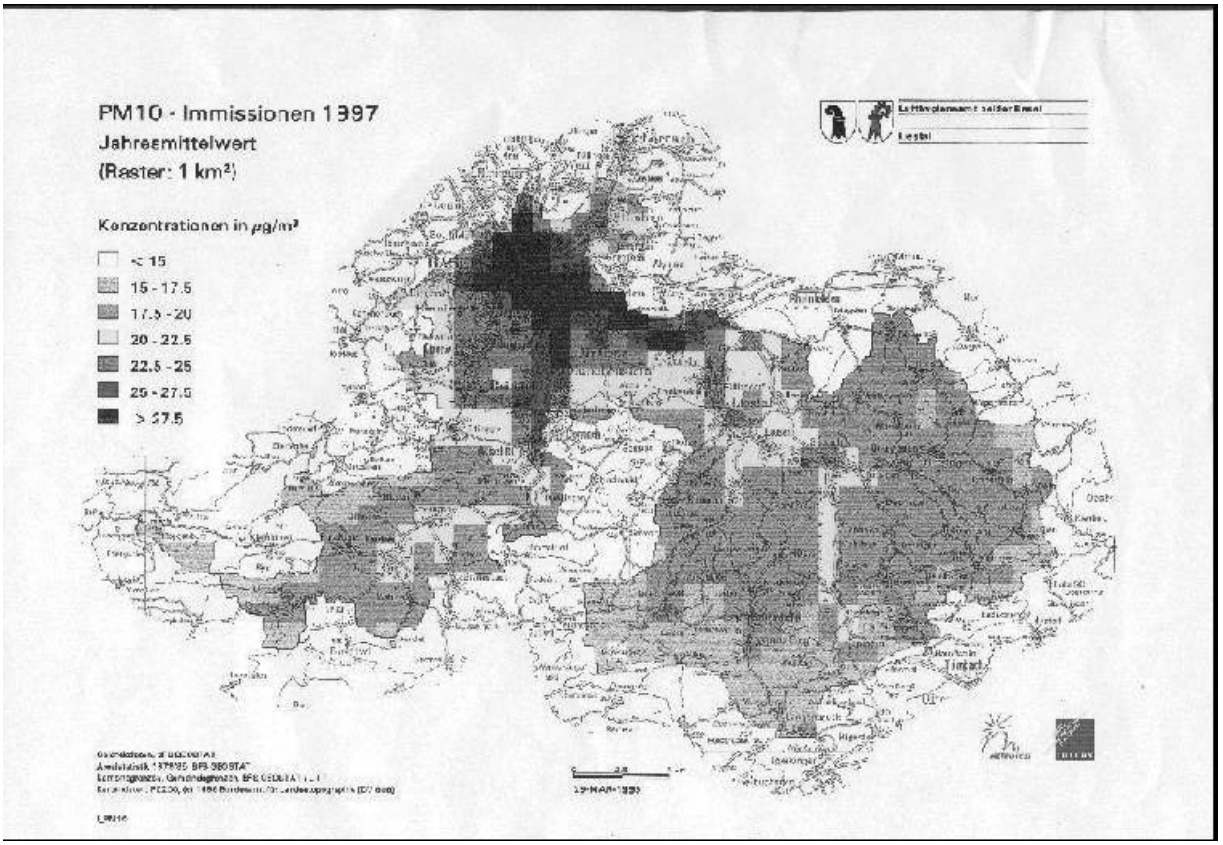

Figure 5-3: Map of modeled PM_{10} concentration in the cantons Basel-Stadt and Basel-Landschaft (from Heldstab et al., 1999).

5.3.2 Unit risk-based risk assessment

Including criteria for carcinogens

In the unit risk-based approach all carcinogens which risk had to be taken into account had to be identified. The following criteria were applied to choose the relevant carcinogens: (i) the substance had to be listed as at least 'possibly

carcinogenic' from the IARC, U.S.EPA or LRV; (ii) it had to be possible to measure it in outdoor air with our devices; and (iii) the carcinogenic potency had to be quantifiable, i.e. at least one unit risk estimate had to be published. Thus, we included the following substances in our risk assessment: arsene, benzene, 1,3-butadiene, cadmium, chromium (VI), 1,4-dichlorobenzene, diesel exhaust, lead, nickel, PAH (benzo[a]pyrene), trichloromethane, trichloroethene, tetrachloroethene.

Quantification of the cancerogenicity of single agents

The carcinogenic potency of these substances was evaluated based on the current scientific literature. For each substance an unit risk factor was estimated based on publication from IARC, U.S.EPA, OEEHA, and LAI (IARC, 2000, U.S.EPA, 1999b, OEEHA, 1999, LAI, 1992). The result is detailed described in Röösli et al., 2000b.

Exposure to carcinogens

Exposure to carcinogenic substances was assessed based on three exposure levels: heavily polluted, average polluted, weakly polluted.

Heavily, weakly, and average polluted

Heavy pollution level of each carcinogen was calculated from the mean value of the six temporary sites representing an average urban value. Within the study BRISKA only a few measurements of carcinogens were performed at rural sites, because priorities were set to estimate their levels in the most polluted areas. Concentration levels at rural sites, often in the borderline of the detection limit, were more problematic to measure and less relevant. Nevertheless, in the risk assessment we had to make some assumptions about their concentration to quantify the health risk. We assumed that the lowest 13-day value in Basel might be a valuable estimate of the background concentration, because these values had to be measured during an effective dilution situation when the emission from the city could be neglected. Comparisons with rural measurements in Switzerland exterior of our study showed generally a good agreement (Röösli et al., 2000b). Average pollution was determined as the arithmetic average of heavily and weakly polluted levels.

Population distribution

The assignment of the population exposure distribution in the three exposure levels was based on the PM_{10}-model. Persons living in areas with average PM_{10} higher than 25 µg/m³ were determined to be exposed to heavy pollution levels (243'500 persons). Persons exposed to an average PM_{10} concentration

between 20 and 25 µg/m³ were assigned to average exposed (157'800 persons) and persons living in areas with PM_{10} levels below 20 µg/m³ were determined to be weakly exposed (49'600 persons).

Quantification of the health risk

Last step in risk assessment comprise the quantification of the health risk. For each carcinogen the unit risk factor was multiplied with the respective population exposure. Cancerogenicity of the total air pollution was obtained by adding the risks of all single substances. The results are presented in Article 5, page 116.

PART IV

AIR QUALITY ANALYSIS

Chapter 6

Spatial Variability of Different Fractions of Particulate Matter Within an Urban Environment and Between Urban and Rural Sites

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Abstract

The spatial variability of different fractions of particulate matter (PM) was investigated in the city of Basel, Switzerland, based on measurements performed throughout 1997 with a mobile monitoring station at six sites and permanently recorded measurements from a fixed site. Additionally, PM_{10} measurements from the following year, which were concurrently recorded at two urban and two rural sites, were compared.

Generally, the spatial variability of PM_4, PM_{10} and total suspended particulates (TSP) within this Swiss urban environment (area=36 km²) was rather limited. With the exception of one site in a street canyon next to a traffic light, traffic density had only a weak tendency to increase the levels of PM. Mean PM_{10} concentration at six sites with different traffic densities was in a range of less than ±10% of the mean urban PM_{10} level. However, comparing the mean PM levels on workdays to that on weekends indicated that the impact of human activities, including traffic, on ambient PM levels may be considerable.

Differences in the daily PM_{10} concentrations between urban and more elevated rural sites were strongly influenced by the stability of the atmosphere. In summer, when no persistent surface inversions exist, differences between urban and rural sites were rather small. It can therefore be concluded that spatial variability of annual mean PM concentration between urban and rural sites in the Basel area may more likely be caused by varying altitude than by distance to the city center.

Introduction

In the past decade, a body of epidemiologic evidence has emerged which demonstrates a range of health effects due to both long-term and short-term exposure to particulates at concentrations commonly occurring in ambient air. In epidemiologic studies investigating long-term effects of PM on health, exposure is usually assigned to a group or population level based on fixed-site monitoring data (Ackermann-Liebrich et al., 1997, Braun-Fahrländer et al., 1997, Zemp et al., 1999, Dockery et al., 1993, Pope et al., 1995). Several Swiss studies comparing the health of children (Braun-Fahrländer et al., 1997) and adults (Ackermann-Liebrich et al., 1997, Zemp et al., 1999) living in different areas of the country also based the exposure assessment on one fixed-site monitor in each study area. However, since these studies were cross-sectional in design, little is known about the small-scale spatial variability

of PM within the study areas although it may affect the observed results. A higher spatial variability leads to a higher expected misclassification error associated with exposure, generally biasing the regression coefficient of the association between air pollution and health towards the null.

Several studies in the United States and Great Britain found long-term mean $PM_{2.5}$ and PM_{10} concentrations to be uniformly distributed within an urban environment (scale: 1-20 km), whereas a higher within-city variability was observed for the coarse fraction ($2.5 < d < 10$ µm) (Burton et al., 1996, Chow et al., 1992, Harrison, 1997, Wilson and Suh, 1997). However, two recent studies reported within-city spatial variability for both $PM_{2.5}$ and PM_{10} in Canada and California (Brook et al., 1999, VanCuren, 1999), but neither of these studies explicitly compared urban sites with different exposures to traffic.

A recent study in the Netherlands comparing daytime $PM_{2.5}$ and PM_{10} concentrations at urban background sites and sites heavily exposed to traffic showed that PM levels were approximately 30% higher at the sites exposed to traffic, mainly due to increased concentrations of black smoke. In addition, the elemental concentrations of Fe and Si in the PM_{10} fraction were significantly higher at the traffic-exposed sites (Janssen et al., 1997). In Switzerland, measurements of vertical and horizontal PM_{10} concentration profiles orthogonal to a road in the city of Zurich showed that mean ground-level measurements of PM_{10} directly at the roadside were 30 µg/m³. Roadside measurements 20 m above ground and ground-level measurements at a distance of 80 m from the road were approximately 4 µg/m³ (13%) lower (Monn et al., 1997).

However, until now, no systematic assessment of the spatial variability in a Swiss urban environment and its association with traffic density has been carried out. Results of studies from the United States and the Netherlands may be difficult to transfer to the Swiss situation because of the existing differences in topography. In particular, it is not known whether the surrounding hills of Basel may affect the small-scale and meso-scale (20-30 km) spatial variability of PM.

Thus, in the present study, concentrations of different fractions of particulate matter (PM_4, PM_{10}, TSP, and coarse fractions) were measured during 1997 at six locations within the city of Basel that differed in traffic density. Between April 1998 and April 1999 additional PM_{10} measurements were concurrently performed at two urban and two rural sites in the Basel area.

The main purpose of the present study was to evaluate whether it is appropriate to use one monitoring site per city for PM exposure assessment in a cross-sectional health study. We quantified the spatial variability of different fractions of PM in an urban environment and between urban and rural locations. Moreover, we were interested in factors which might influence this spatial variability.

Methods

Air pollutant measurements

For the present analyses, two sets of data were used, both of which were recorded in the Basel area. Basel is a city of about 200.000 inhabitants located in the northwestern part of Switzerland at 250 m above sea level, and it is surrounded by suburbs and more elevated rural areas.

The first data set included measurements performed between January 8 and December 22, 1997, as part of the Basel Risk Assessment Study of Ambient Air Pollutants (BRISKA). Twenty-four hour mean concentrations of TSP, PM_{10}, and PM_4 were recorded at six temporary sites within the city of Basel by means of a mobile monitoring station rotating every two weeks. (Due to technical problems, PM_4 was not measured until March 1.) This resulted in a 13-day sampling period from each site in each season (generally a total of 52 measurements per site). The six temporary sites differed in traffic density and were assumed to be representative of the traffic exposure of the Basel population (see Table 6-1). Two sites (1 and 2) were situated in typical residential areas with low road traffic density, one site (3) was located on a square in the very center of Basel, approximately 150 m away from a road with moderate traffic, and three sites (4-6) were located close to roads with a high traffic volume (16.300 - 29.500 passenger cars daily; 1.130 - 4.920 heavy duty vehicles daily). The maximum distance between these six sites was 3.3 km. PM_{10} was also measured permanently at a fixed monitoring station of the Air Quality Management Agency of Basel (site C), to represent the urban background. In addition, permanent measurements of PM_4, PM_{10} and TSP recorded throughout 1997 at a fixed site monitoring station located at the outskirts of Basel (NABEL station) (BUWAL, 1998) were used as reference for the present analyses.

The second set of data was recorded from April 1, 1998, to April 30, 1999, at two rural and two urban sites (see Table 6-1). In addition to urban site C (urban background) a monitoring station was located in a street canyon with heavy traffic close to a traffic light (site D). Rural sites A and B were 25 and 20 km to the southeast of the city at altitudes of 900 and 600 m,

respectively (Basel = 250 m). At all four stations simultaneous measurements of 24-hr mean concentrations of PM_{10} were performed.

Table 6-1: Overview of the database and the characteristics of the measurement sites. Traffic counts originate from the traffic inventory of the city of Basel (Baudepartement and Basel-Stadt, 1994).

Date	PM	Sampling Period	Type	Site Name	Passenger Cars/24h	Heavy Duty Vehicles/24hr	Comments
Jan. 8 - Dec. 22, 1997	PM_4, PM_{10}, TSP	permanent		NABEL	no traffic		outskirt of Basel (reference site)
		temporary sites, 13 days per season at each site using mobile monitoring station	urban	1	5.000	260	residential
				2	5.200	370	residential
				3	6.200	500	center of Basel
				4	16.300	3.410	high truck quota
				5	27.100	1.130	sloped lane
				6	29.500	4.920	cross-way
4/1/98 -4/30/99	PM_{10}	permanent	urban	D	17.000	890	street canyon, traffic light
				C	11.500	950	PM_{10} also measured during 1997
			rural	B	no traffic		Altitude=600 m, 20 km from Basel
				A			Altitude=900 m, 25 km from Basel

All PM concentrations (PM_4, PM_{10} and TSP) were measured with High Volume Samplers (HVS) (Digitel DHA 80). The samples were collected on quartz filters (QF 20 Schleicher & Schuell), changed automatically at 12:00 a.m. local time (GMT+1h, wintertime). The air was sampled 2.40 m above ground. Particle concentrations were determined using pre- and post-sampling filter weights taking into account the sampled air volume. The filters were equilibrated under controlled temperature (15-24°C) and relative humidity (47-60%) 48 hr prior to weighing, in order to ensure consistent values for particle mass.

As a quality control measurement, an additional 21 blank filters were placed in the HVS device at the monitoring site for 14 days but were not exposed. Weight differences between

pre- and post-sampling weighing of these blank filters were, on average, associated with a concentration of -0.04 µg/m³ (standard error = 0.80 µg/m³).

The air-flow volume of the HVS was calibrated at 500 L/min at all monitoring sites. The flow was kept constant using a light barrier. The volume was corrected for temperature. The applied procedure is described in EMPA, 1998 (EMPA, 1998).

Data quality control of the measurements revealed that PM_4 had actually been recorded instead of $PM_{2.5}$. This was due to errors in the calculation of the geometry of the device's head which had led to that larger cut-off. To assess how comparable the PM_4 measurements were with the more common measurement of $PM_{2.5}$, the EPA-WINS Impactor used for the Air Pollution Exposure Distribution among Adult Urban Populations in Europe (EXPOLIS-Basel) study (Oglesby et al., 2000) was repeatedly installed at the fixed site NABEL monitoring station throughout 1997. The measurements indicated that PM_4 levels are highly correlated with $PM_{2.5}$ levels, though on average 11% higher than the latter (25.2 vs. 22.3 µg/m³). The coarse fractions were determined by calculating the differences between the three size fractions ($PM_{(10-4)}$, $PM_{(TSP-10)}$, $PM_{(TSP-4)}$).

Statistical analyses

The measurements at sites 1-6 in Basel, performed during 1997 by the mobile monitoring station, could not be directly used to calculate annual means and to assess the spatial variability, as they had not been performed concurrently. Comparing raw PM levels between the sites could have yielded differences in the concentrations of PM, simply owing to differing meteorological conditions prevailing during the sampling period, or to temporal varying source strengths such as traffic density, or both. Two methods were used to deal with such influences on the calculation of mean values for each temporary site. First, the 52 measurements from each temporary site were compared with the concurrent measurements from the permanent NABEL site. The Pearson correlation coefficients and the mean ratios between the measurements from each temporary site and the corresponding measurements from the NABEL site were calculated. As the distribution of ratios is asymmetrical, mean values were obtained by taking the logarithm of the ratios and then exponentiating that value.

Second, to adjust for meteorological conditions, a multiple regression model was designed for each site and each particulate size fraction. The daily PM measurements from the temporary station were regressed against the respective daily PM measurements from the NABEL

station and against meteorological parameters. The following mathematical expression describes the model:

$$C_{temp.site} = a + b \cdot C_{NABEL} + \sum_{i=1}^{n} k_i \cdot m_i \qquad \textbf{(Equation 6-1)}$$

where C = concentration of PM_4, PM_{10}, TSP, or coarse fractions at the six temporary sites (temp. site) and at the reference site (NABEL), m_i = meteorological parameters and a, b, and k_i are estimated coefficients.

Meteorological parameters that were significant in at least one model were considered as covariates of the final model that was used for all sites. The following meteorological parameters were included in the preliminary models: season (binary: oct-mar=1), daily means of temperature and humidity, daily sum of global radiation, daily minimum and maximum of the temperature gradient between ground level and a tower measurement 243 m above ground and the daily vector sum of all wind-direction measurements taken every 10 min.

In general, each model was built with 52 pairs of daily PM measurements (temporary and NABEL) and meteorological parameters. Based on these models daily PM concentrations for the remaining days were predicted. Annual and seasonal means were then calculated on the basis of all observed and predicted PM values. Because of the missing PM_4 values in January and February models for PM_{10} and TSP were calculated for both the January to December and March to December periods.

The models were validated with the PM_{10} measurements from site 'C' which had been measured permanently throughout the year. The measurement periods 1-6 were defined to correspond to the four time intervals during which the mobile monitoring station was located at the respective sites 1-6. Each measurement period consisted of 52 days with PM measurements (13 days in each season). First, the mean PM_{10} concentration at site C was calculated for each measurement period yielding 22.7 µg/m³, 24.3 µg/m³, 27.8 µg/m³, 30.0 µg/m³, 23.3 µg/m³, and 32.2 µg/m³, respectively, for the measurement periods 1 to 6. The annual mean at site 'C' computed from the daily measurements throughout the year was 27.9 µg/m³. Thus, estimates of annual means based on averages of the available PM measurements during one measurement period would give rather variable results compared to the 'true' mean of 27.9 µg/m³. Second, the PM_{10} readings of site C in each measurement period were used as dependent variable for the regression models described above (i.e. including the PM_{10} values

of the reference station and meteorological parameters as covariates). Based on these models PM_{10} concentrations for the remaining days were predicted and annual means calculated. The model-based annual means for site C could then be compared to the measured annual mean PM_{10} concentration at site C. The six annual means derived from the regression models were 26.9 µg/m³, 27.8 µg/m³, 26.4 µg/m³, 27.6 µg/m³, 28.4 µg/m³, and 30.4 µg/m³, respectively, for the measurement periods 1-6. These results indicate that the annual means computed from regression models are indeed close to the measured mean concentration of 27.9 µg/m³ at site C, although some inaccuracy could not be eliminated.

In order to compare the spatial variability of the different size fractions of PM across the six temporary sites, coefficients of spatial variation were calculated. For each size fraction, the overall mean of the six sites and the corresponding standard deviation were computed using the measurements from March to December to avoid undue influence from the missing PM_4 measurements in January and February. The coefficient of spatial variation (cv) for each particulate size fraction was obtained by dividing the standard deviation by the overall mean.

To evaluate the hypothesis that proximity to traffic might influence the local PM measurements, the annual mean PM concentrations of each of the six temporary sites were plotted against the respective daily mean number of passenger cars and heavy-duty vehicles, and linear regression models were calculated to estimate the regression coefficients of this association. The information on traffic density was provided by the traffic inventory of the canton Basel-Stadt (Baudepartement and Basel-Stadt, 1994). This inventory contains information on average numbers of cars and trucks passing a given address each hour and is updated by traffic counts every 3-4 years. Again, these analyses were based on the measurements from March to December 1997.

The differences between mean PM concentrations on workdays and weekends may reflect the influence of regional anthropogenic sources since many human activities such as industrial processes, construction work, or road traffic are reduced on weekends. Thus, the mean concentration for each PM fraction was calculated for each day of the week. In addition, the ratio between mean concentrations on workdays and weekends and the corresponding two-sided t-test were computed. These analyses were based on the data provided by the fixed monitoring station (NABEL) recorded between March and December 1997.

The second data set consisting of simultaneously performed measurements at four fixed monitoring sites (two urban and two rural) was used to assess differences in annual mean

PM_{10} concentrations between urban and rural sites. In addition, monthly mean values at the two rural and the two urban sites were compared to assess whether seasonal factors such as the number of days with thermal inversion may influence urban–rural differences.

Temperature gradients between the urban and the rural sites were computed by calculating the differences between the daily maximum temperature at the urban (NABEL) and the rural station A. A positive gradient is indicative of an inversion layer.

Data analyses

Data were analyzed using S-Plus 4.5 Professional Release 2 for Windows.

Results

Spatial variability of different fractions of particulate matter within an urban environment

Overall mean PM concentrations across the six temporary stations for the measurements performed between March and December were 33.1 µg/m³ (TSP), 24.7 µg/m³ (PM_{10}) and 19.9 µg/m³ (PM_4). The annual mean TSP and PM_{10} concentrations (January-December) were 3.8 and 2.4 µg/m³ higher, respectively, than mean values for the period from March to December, due to an episode of high air pollution in January (PM_4 was not measured in January and February). The ratio of the mean PM_4 versus PM_{10} concentrations (March-December) was 81%, and 75% for the mean PM_{10} versus TSP concentrations.

Comparisons of the 52 measurements of each temporary site with the concurrent measurements from the NABEL site are shown in Table 6-2. In each size fraction the mean ratios of the concurrent measurements were generally lowest at the two residential sites '1' and '2'. Only $PM_{(10-4)}$ did not show this pattern. For the coarse fractions a wide range of ratios was found between the six sites ,whereas for PM_4, PM_{10} and TSP a smaller range was found, indicating more spatial uniformity. The correlation coefficients (R^2) of the measurement were highest for PM_{10} but also mostly above 0.9 for PM_4 and TSP. The R^2 of all coarse fractions ($PM_{(TSP-10)}$, $PM_{(TSP-4)}$, $PM_{(10-4)}$) was below 0.8 at all six sites. The R^2 of the measurements was always lower than the corresponding R^2 from the meteorology-based regression models, even taking into account the lower number of degrees of freedom in the regression models. The standard errors of the regression coefficient of the NABEL measurements indicate that the t-value was lower for the coarse fraction models than for the PM_4, PM_{10} and TSP models.

Table 6-2: **Mean ratio (ratio measured) and R^2 (R^2 measured) between concurrently measured PM concentrations at the six temporary sites and the permanent NABEL site, as well as R^2 (R^2 model) and regression coefficients b ± standard error (b±SE model) of the regression models. The latter included the measurements from the temporary sites as dependent variables and the permanent NABEL measurements and meteorological parameters as explanatory variables. (b is the coefficient of the permanent NABEL measurements. Model coefficients of the meteorological parameters are not shown as they had a considerably weaker influence than the measurements from the NABEL site.)**

		Residential 1 [µg/m³]	Residential 2 [µg/m³]	Center 3 [µg/m³]	Traffic 4 [µg/m³]	Traffic 5 [µg/m³]	Traffic 6 [µg/m³]
PM_{10} (Jan-Dec)	ratio (measured)	0.90	0.94	0.99	0.97	0.96	1.04
	R^2 (measured)	0.97	0.97	0.97	0.98	0.96	0.98
	R^2 (model)	0.99	0.99	0.99	0.99	0.99	0.99
	β (± SE) model	0.94 (±0.02)	0.97 (±0.03)	0.88 (±0.03)	0.90 (±0.03)	1.03 (±0.03)	1.03 (±0.04)
TSP (Jan-Dec)	ratio (measured)	1.06	1.02	1.18	1.19	1.12	1.34
	R^2 (measured)	0.84	0.94	0.95	0.93	0.93	0.92
	R^2 (model)	0.93	0.97	0.97	0.94	0.95	0.94
	β (± SE) model	0.93 (±0.06)	1.20 (±0.06)	1.05 (±0.07)	1.20 (±0.07)	1.14 (±0.06)	1.06 (±0.07)
PM_4 (Mar-Dec)	ratio (measured)	0.75	0.82	0.92	0.88	0.99	0.98
	R^2 (measured)	0.95	0.78	0.98	0.91	0.97	0.98
	R^2 (model)	0.98	0.93	0.99	0.98	0.99	0.99
	β (± SE) model	0.85 (±0.04)	0.91 (±0.07)	0.93 (±0.03)	0.97 (±0.04)	1.06 (±0.03)	0.94 (±0.05)
PM_{10} (Mar-Dec)	ratio (measured)	0.88	0.93	0.99	0.99	0.96	1.03
	R^2 (measured)	0.97	0.88	0.97	0.93	0.96	0.98
	R^2 (model)	0.99	0.96	0.99	0.97	0.99	0.99
	β (± SE) model	0.91 (±0.03)	0.92 (±0.06)	0.88 (±0.03)	0.98 (±0.05)	1.03 (±0.03)	1.00 (±0.05)
TSP (Mar-Dec)	ratio (measured)	1.00	0.97	1.18	1.20	1.12	1.32
	R^2 (measured)	0.94	0.85	0.95	0.79	0.93	0.92
	R^2 (model)	0.97	0.96	0.97	0.86	0.95	0.95
	β (± SE) model	0.92 (±0.05)	1.00 (±0.07)	1.05 (±0.07)	1.14 (±0.16)	1.14 (±0.06)	1.06 (±0.10)
$PM_{(TSP-10)}$ (Mar-Dec)	ratio (measured)	1.92	1.01	3.52	2.66	4.25	3.51
	R^2 (measured)	0.73	0.56	0.43	0.44	0.46	0.63
	R^2 (model)	0.86	0.87	0.53	0.69	0.63	0.83
	β (± SE) model	0.85 (±0.26)	1.09 (±0.21)	0.65 (±0.46)	1.28 (±0.39)	1.35 (±0.36)	1.93 (±0.35)
$PM_{(TSP-4)}$ (Mar-Dec)	ratio (measured)	1.61	1.40	2.04	1.93	1.40	2.24
	R^2 (measured)	0.68	0.60	0.66	0.57	0.61	0.63
	R^2 (model)	0.87	0.84	0.72	0.76	0.74	0.80
	β (± SE) model	0.87 (±0.16)	0.90 (±0.16)	1.56 (±0.35)	1.33 (±0.29)	1.33 (±0.21)	1.57 (±0.23)
$PM_{(10-4)}$ (Mar-Dec)	ratio (measured)	1.35	1.45	1.26	1.43	0.80	1.37
	R^2 (measured)	0.30	0.31	0.44	0.66	0.11	0.76
	R^2 (model)	0.59	0.90	0.66	0.79	0.44	0.92
	β (± SE) model	0.75 (±0.16)	0.71 (±0.15)	0.50 (±0.11)	0.80 (±0.12)	0.55 (±0.20)	1.04 (±0.13)

Table 6-3 shows the mean concentrations of the different fractions of PM at the six temporary urban sites, which were calculated based on the regression models. Again lowest mean concentrations of PM_4, PM_{10} and TSP occurred at the two residential sites (1 and 2) and had a weak tendency to increase with increasing road traffic density. The biggest differences between the residential sites and more traffic-exposed sites were detected for $PM_{(TSP-10)}$ and $PM_{(TSP-4)}$ but not for $PM_{(10-4)}$. The calculated coefficients of spatial variation, as well as the ratio of the highest to the lowest mean concentration at the six sites, were clearly higher for the coarse fractions ($PM_{(10-4)}$, $PM_{(TSP-10)}$, $PM_{(TSP-4)}$) than for PM_4, PM_{10}, and TSP. The lowest spatial variability was found for the PM_{10} fraction.

To evaluate whether there was an association between the traffic density at a given site and the respective concentrations of the different size fractions of PM, linear regression models were calculated. The results are given in Table 6-4, and Figure 6-1 illustrates the association. Overall, only a very weak association between local PM concentrations and the traffic density was observed, and it reached statistical significance only for PM_4 and passenger cars.

Table 5-3. Calculated mean concentration of different fractions of particulate matter for the periods of January-December and March-December as well as coefficient of spatial variability (cv) and the ratio of the highest and lowest concentration of each PM fraction.

	Residential 1 [µg/m³]	Residential 2 [µg/m³]	Center 3 [µg/m³]	Traffic 4 [µg/m³]	Traffic 5 [µg/m³]	Traffic 6 [µg/m³]	cv[a] [%]	PM Ratio highest/lowest
PM_{10} ‡	27.6	28.4	30.1	28.6	29.6	32.0	5.2	1.16
(CI)*	(26.1;29.2)	(26.4;30.4)	(27.9;32.3)	(26.1;31.0)	(27.7;31.5)	(28.7;35.2)		
TSP ‡	34.5	36.4	41.5	40.8	38.9	45.4	9.9	1.32
(CI)	(30.1;38.8)	(32.6;40.1)	(36.7;46.3)	(33.7;47.8)	(35.2;42.7)	(39.2;51.5)		
PM_4 §	17.4	18.0	20.5	19.7	21.4	22.1	9.4	1.27
(CI)	(15.3;19.5)	(16.0;19.9)	(18.6;22.5)	(18.2;21.1)	(19.6;23.1)	(18.7;25.4)		
PM_{10} §	23.0	23.7	26.0	24.8	24.3	28.0	7.2	1.21
(CI)	(21.2;24.9)	(21.7;25.7)	(23.8;28.2)	(23.0;26.7)	(22.4;26.2)	(23.8;32.1)		
TSP §	30.6	29.4	36.9	34.0	34.2	42.1	13.3	1.43
(CI)	(27.3;34.0)	(27.0;31.8)	(32.1;41.8)	(26.3;41.7)	(30.4;37.9)	(34.5;49.7)		
$PM_{(TSP-10)}$ §	7.3	5.1	10.4	8.2	9.5	14.1	33.8	2.78
(CI)	(4.7;9.8)	(3.0;7.1)	(5.4;15.4)	(1.2;15.3)	(5.5;13.5)	(9.9;18.3)		
$PM_{(TSP-4)}$ §	12.9	10.8	16.1	13.6	12.6	19.9	22.4	1.84
(CI)	(10.1;15.8)	(8.7;12.9)	(10.8;21.3)	(6.5;20.7)	(9.1;16.1)	(14.8;24.9)		
$PM_{(10-4)}$ §	5.4	5.4	5.6	4.9	2.6	6.3	25.2	2.40
(CI)	(3.8;7.0)	(4.4;6.5)	(4.0;7.2)	(3.7;6.2)	(1.0;4.2)	(4.4;8.1)		

‡ January-December § March-December * CI=confidence interval [a] cv=standard deviation/mean

Table 6-4: Results from the linear regression analyses of mean PM concentration at the six urban sites and mean number of cars and trucks passing the site per day. b **refers to an increase of 10.000 passenger cars and 1000 trucks.**

	Passenger Car Model				Truck Model			
	intercept	β	(CI of β)	R^2	intercept	β	(CI of β)	R^2
PM_4	17.8	1.38	(0.10;2.65)	0.69	18.8	0.59	(-0.46;1.63)	0.38
PM_{10}	23.6	0.91	(-0.91;2.73)	0.33	23.7	0.69	(-0.15;1.54)	0.56
TSP	30.5	2.73	(-1.48;6.95)	0.45	31.4	1.76	(-0.42;3.94)	0.56
$PM_{(TSP-10)}$	6.2	1.95	(-0.71;4.62)	0.51	7.1	1.11	(-0.47;2.68)	0.49
$PM_{(TSP-4)}$	12.2	1.45	(-1.97;4.87)	0.26	12.2	1.18	(-0.43;2.79)	0.51
$PM_{(10-4)}$	5.6	-0.39	(-1.86;1.09)	0.12	4.7	0.18	(-0.69;1.05)	0.08

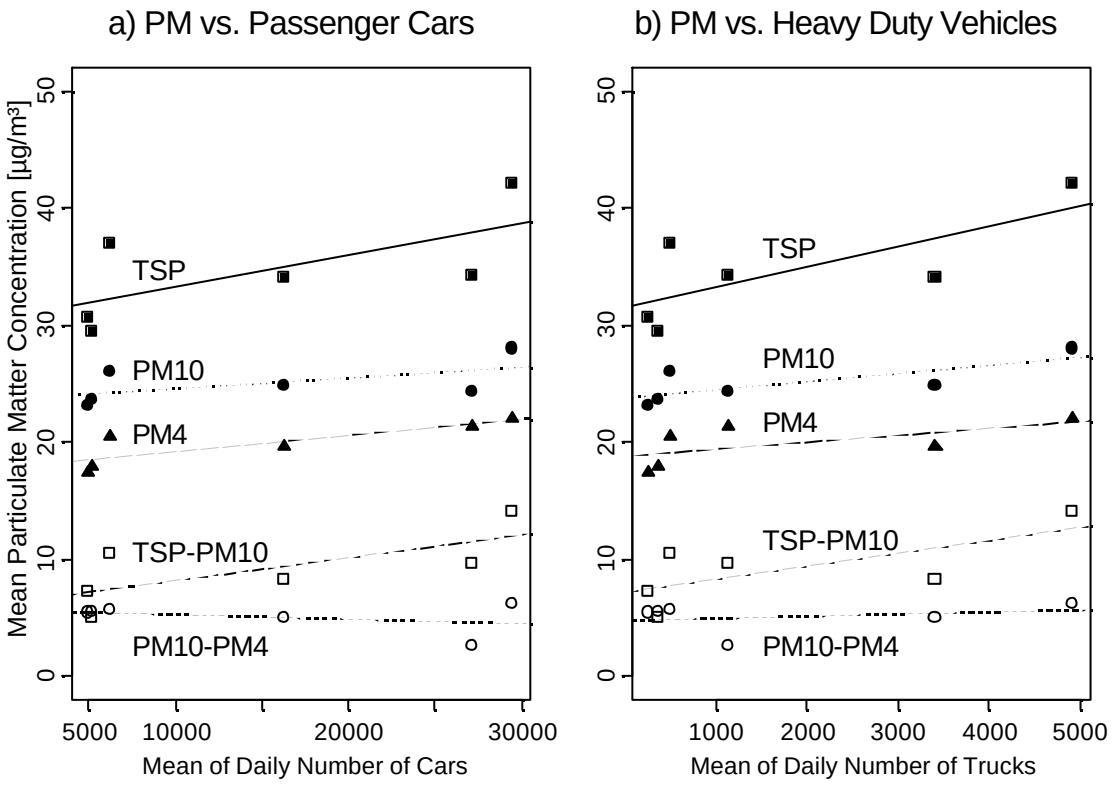

Figure 6-1: Mean calculated PM concentration of the different size fractions of PM for March 1 to December 31, 1997, plotted against the mean daily counts of passenger cars (a) and heavy duty vehicles (b).

Figure 6-2 depicts the mean concentrations of the different PM size fractions for each day of the week. The concentrations of all size fractions were lowest on Sunday, increased from Monday to Wednesday, and decreased again on Friday and Saturday. The ratios between the mean concentrations on workdays and weekends increased with increasing particle sizes. The concentration of $PM_{(TSP-10)}$ was 51% higher on workdays as compared to weekends, and $PM_{(10-4)}$ was 32% higher. On workdays, TSP, PM_{10} and PM_4 were elevated by 20, 17, and 14%, respectively. These differences were statistically significant for PM_{10} ($p=0.045$), TSP ($p=0.012$), $PM_{(10-4)}$ ($p=0.0007$) and $PM_{(TSP-10)}$ ($p=0.0028$). For PM_4, the difference was of borderline significance ($p=0.107$).

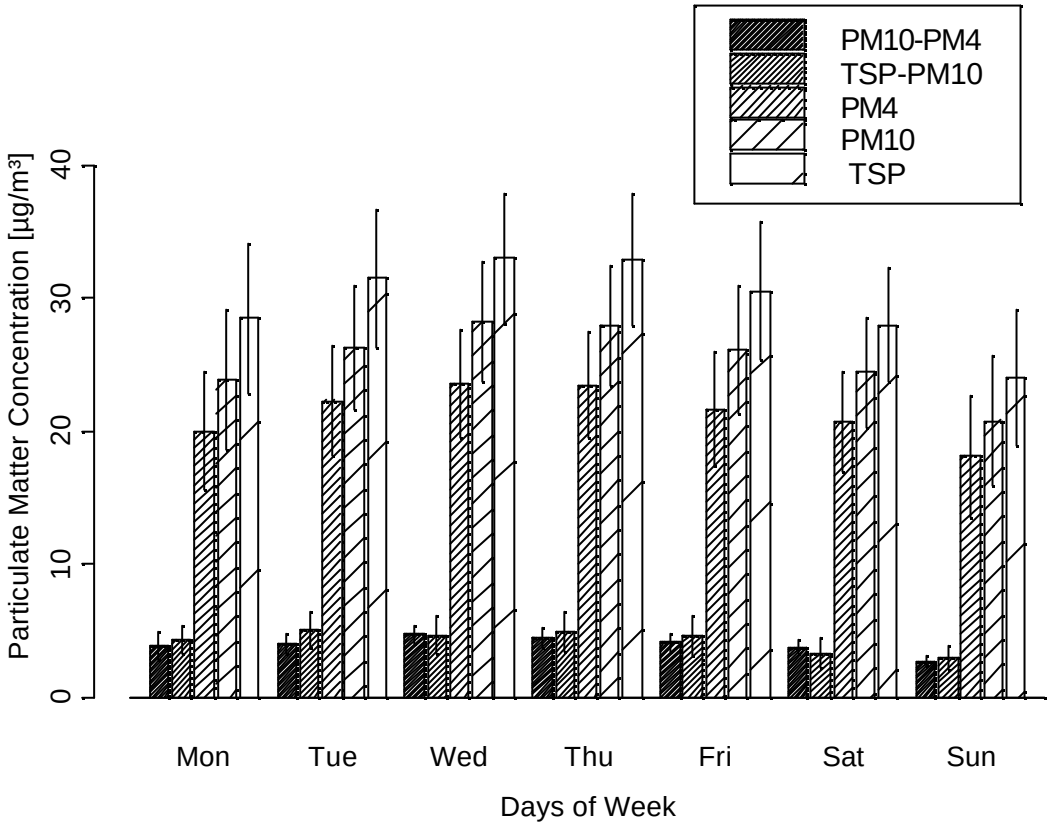

Figure 6-2: Mean concentrations (± confidence interval) of PM_4, PM_{10}, TSP, and coarse fractions ($PM_{(10-4)}$, $PM_{(TSP-10)}$) by day of week. Measurements from the fixed monitoring site (NABEL) for the time period of March to December 1997.

Spatial variability of PM_{10} between urban and rural sites

To assess differences in PM levels between the rural and urban sites, the PM_{10} measurements monitored from April 1, 1998, to April 30, 1999, were used. During this period, the PM_{10} levels were generally lower than in 1997 due to different meteorological conditions. In 1997, the annual mean PM_{10} concentration at urban site C was 27.9 µg/m³, whereas the average PM_{10} level between April 1998 and March 1999 was only 22.4 µg/m³. Figure 6-3 gives the boxplots for the measurements at the four sites from April 1998 to March 1999. The highest PM_{10} concentrations were measured at the urban site D, situated in a street canyon with high traffic density. The annual mean value at this site was more than one-third higher than the PM_{10} concentration at urban site C, which represented urban background exposure. The annual PM_{10} levels at the rural sites A and B were 48% and 31% below the levels of site C, respectively.

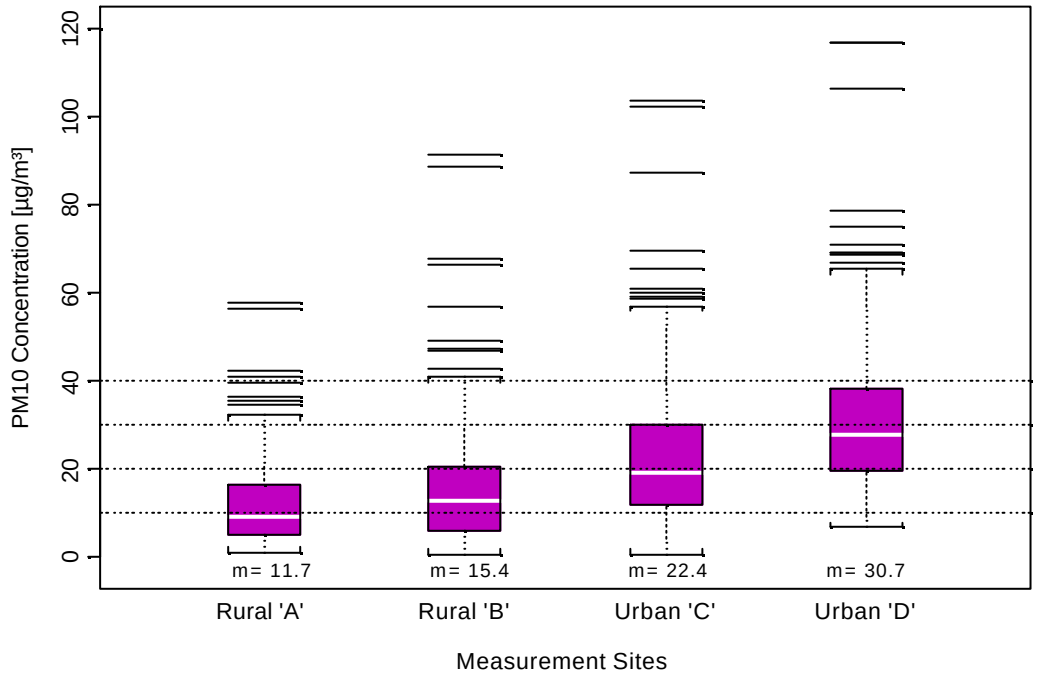

Figure 6-3: Boxplots of PM_{10} measurements at two urban and two rural sites from April 1, 1998, to March 31, 1999. The box contains 50% of all measurements, the white line represents the median value and *m* equals the arithmetic mean value.

Figure 6-4 shows the monthly PM_{10} concentrations at the four monitoring sites. During the summer months (April to October) the variability of the monthly PM_{10} levels was comparable at all four stations. From November to March, however, the monthly mean PM_{10} concentration at the two urban sites increased more than at the rural stations.

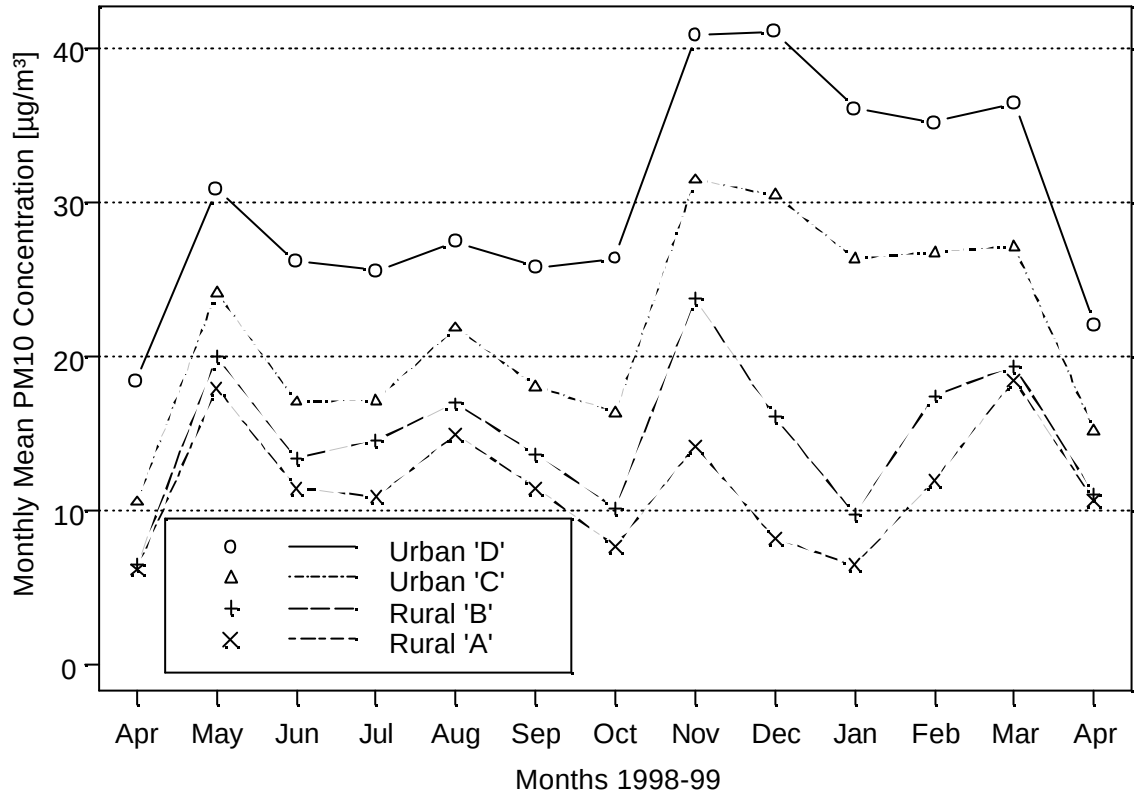

Figure 6-4: Monthly mean values of PM_{10} at two urban and two rural sites.

An analysis of the meteorological conditions revealed days with positive temperature gradients from December to February between the more elevated rural site A (900 m) and the urban NABEL site (250 m). This is characteristic for the presence of an inversion layer and was positively associated with increased levels of PM_{10} at the urban site (see Figure 6-5). During the warm months, on the other hand, vertical temperature gradients were rarely positive.

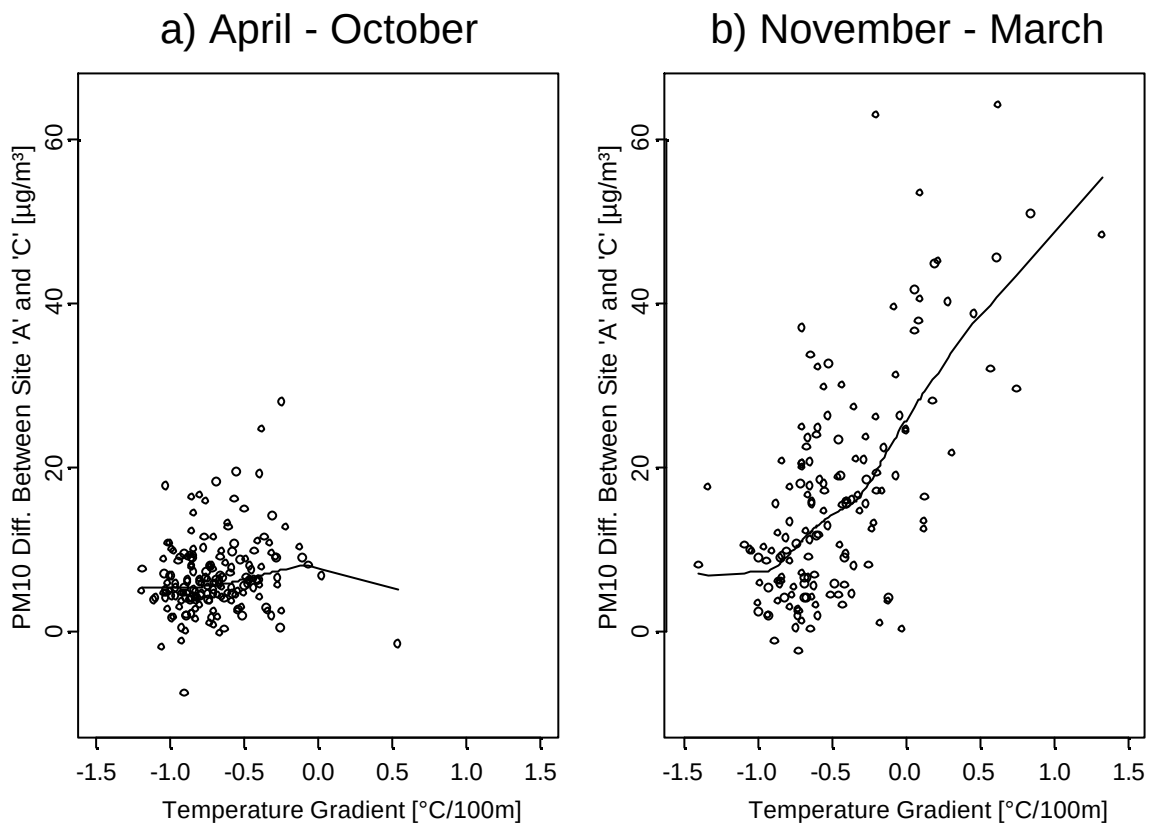

Figure 6-5: Smooth plot of the association of vertical temperature gradient and daily PM_{10} difference between urban site C and rural site A for the period of April-October 1997 (a) and November-March (b).

Discussion

In general, ambient levels of PM_4, PM_{10} and TSP were distributed rather homogeneously within the urban area of Basel. A higher spatial variability was found for the coarse fractions ($PM_{(TSP-10)}$, $PM_{(TSP-4)}$, $PM_{(10-4)}$). These findings are in line with previous findings (Burton et al., 1996, Chow et al., 1992, Harrison, 1997, Wilson and Suh, 1997, Oglesby et al., 2000) and did not depend on the method used to analyze the spatial variability. However, using multiple regression models to calculate mean values yielded slightly higher R^2 values as compared to the crude analyses. Comparing only ratios without taking meteorological conditions into account may bias the result, because the ratios systematically varied with varying pollution levels, which in turn are a function of the meteorological conditions.

The slightly higher spatial variability of PM_4 as compared to PM_{10} was an unexpected finding. It is possible that this result occurred by chance. Alternatively, one might speculate that the size distribution between PM_4 and PM_{10} at the two residential sites as compared to the

more traffic exposed sites was slightly changed on average due to known processes (Horvath et al., 1996, Clarke et al., 1999a, Vignati et al., 1999) such as sedimentation, coagulation and condensation of water vapor on the particle surface. These processes are effective in the ultrafine particles (<50 nm) on a small spatial scale (Shi et al., 1999, Junker et al., 2000). Whether they influence the size distribution of PM_4 and PM_{10} in our study area cannot be determined with certainty with our data.

Only a weak and mostly insignificant association was observed between the volume of road traffic at the six sites and the concentration of PM. The models are, however, limited due to the relatively small range of observed traffic and the fact that only six sites were analyzed. Moreover, other factors, such as building density and distance to the road plane were not absolutely identical among the six sites. The particulate matter concentration at site 3 was higher than expected from traffic counts alone, suggesting factors other than traffic influence the long-term concentration of PM. On the other hand, PM_{10} measurements from site D demonstrate that, at some locations, considerable small-scale spatial gradients in the long-term levels of PM may occur within Basel due to a specific traffic situation. Site D was located in a street canyon close to a traffic light.

The differences in the mean PM concentrations at different days of the week, however, suggest that human activities that are reduced on weekends, such as road traffic, construction work, and industrial processes, can markedly influence the levels of PM in Basel, even if they, generally were not found to have a particular effect on the small-scale spatial variability.

The differences between urban and rural sites were determined by the existence of an inversion layer. A strong impact of persistent surface inversion layers on the levels of PM has also been found in the Swiss alpine region (Gälli Purghart B.C., 1990). In Basel, the accumulation of pollutants in the air on days with persistent surface inversion due to reduced dilution can be demonstrated by the mean PM_{10} levels on the 21 days that had a positive temperature gradient between rural site A and Basel (November-March). The mean PM_{10} concentrations were 56 (D), 48 (C), 27 (B), and 12 µg/m³ (A). The average urban value (C) for this period was more than twice as high as the annual mean PM_{10} level at this site, whereas the PM_{10} concentration at site A was equal to its annual mean value. It is known that, in general, the altitude of the upper boundary of an inversion layer is lowest during December and January, and of the 21 days with a positive temperature gradient, 15 occurred during December 1998 and January 1999. In November and March, persistent surface inversions are not unusual, but the boundary is often higher than 900 m, resulting in increased monthly PM_{10}

levels at the elevated rural sites as well. With our data, it was not possible to identify unequivocal days with inversion above 900 m, due to missing information on the vertical temperature gradient. In summer, persistent surface inversions rarely occur. Thus, from April to October only two days with a positive temperature gradient between site A and C were observed, and they may also have been caused by factors other than inversions.

Therefore, it can be concluded that differences between PM concentrations at urban and rural sites may be more likely caused by varying altitude than by the distance from the city center.

The remarkable spatial homogeneity in long-term mean PM levels clearly reduces the error of assigning data from one fixed monitoring site to all study subjects living in Basel, as was done in the former Swiss health study SAPALDIA. In fact, all participants lived in urban Basel, rendering PM_4, PM_{10}, and even TSP a useful city-wide surrogates for long-term exposure to outdoor air pollution.

Implications

It has become popular in recent years to use the concentration of particulate matter (PM) as an indicator of air pollution exposure in epidemiologic studies. Since many of these studies assess the exposure of subjects based on one measurement per city, the accuracy of this technique will markedly affect the result of cross-sectional studies. High spatial variability of PM could result in a large non-differential misclassification of exposure that would lead to a smaller recognized health effect of air pollution.

The remarkable spatial homogeneity of long-term mean PM levels clearly reduces the error of assigning data from one fixed monitoring site to all study subjects living in Basel, as was done in recent cross-sectional health studies in Switzerland (SAPALDIA, Swiss Study on air pollution and Lung Diseases in Adults; SCARPOL, Swiss Study on Childhood Allergy and Respiratory Systems). In fact, all participants lived in urban Basel, rendering PM_4, PM_{10} and even TSP useful city-wide surrogates for long-term exposure to outdoor air pollution.

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Chapter 7

Airborne Particle Number Profiles, Particle Mass Distributions and Particle Bound PAH Concentrations Within the City Environment of Basel: an Assessment as Part of the Briska Project

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Abstract

A project aimed at assessing a variety of particulate indicators was carried out at three locations in the city of Basel within the scope of the Basel Air Quality Risk Assessment Project (BRISKA). Size specific particle number concentrations (0.018-0.421 µm), particle mass distributions (0.04-49 µm), and particle bound polycyclic aromatic hydrocarbon (pPAH) concentrations were measured. Locations differed in housing and traffic characteristics. For particle number and pPAH concentrations, one separate continuous 27-28-h measurement was performed at each location. Simultaneously, non continuous measurements assessing the particle mass distribution were performed. Distinct day profiles corresponding to the diurnal traffic profile were observed for absolute particle numbers and pPAH concentrations. The number of ultrafine particles as well as pPAH concentrations were more closely correlated to the number of heavy duty vehicles (r = 0.67 - 0.86 and 0.85 - 0.90) respectively than to the number of light duty vehicles (r = 0.43 - 0.59 and 0.64) respectively. The highest particle number concentrations were found in the ultrafine particle mode (< 0.1 µm) averaging between 82 - 87 % of the total particle numbers < 0.421 µm while the accumulation mode (0.1-2.8 µm) made up for most of the particle mass (mean > 82 %). For size specific absolute mass concentrations, the highest concentrations were generally found in the early morning and evening hours. The mass distribution within the accumulation mode differed from rush hour episodes, in between rush hour episodes and a lawn mowing episode.

Introduction

During the last few years much research regarding the health effects of particulate pollution has been carried out. Various studies concluded that PM_{10} (particulate matter < 10 µm as a mass concentration) is a strong indicator for a variety of health effects, both in Switzerland and abroad (Dockery et al., 1989, Pope and D.W. Dockery (1992) Acute health effects of PM10 pollution on symptomatic and asymptomatic children. Am Rev Respir Dis 145/5, 1992, Schwartz, 1994b, Ackermann-Liebrich et al., 1997, Braun-Fahrländer et al., 1997). Consequences due to PM_{10} exposure range from increased mortality and morbidity to decreasing lung functions as well as a number of pulmonary effects, both acute and chronic. However, it is not clear which property of PM_{10} pollution is responsible for a certain health effect. Properties such as particle size, mass, surface and chemical composition of the particles as

well as particle numbers are being discussed in regard to the assessed health impact, and until now the data available emphasizes the complexity of the matter.

With respect to acute symptoms, one study indicates that certain lung function parameters of asthmatics are more strongly associated to the number of ultrafine particles < 0.1 µm (Peters and Wichmann, 1996), while another research group found no such associations (Pekkanen et al., 1997). Additionally, short term exposure (1 h) to diesel exhaust fumes have been attributed to creating acute inflammation (Blomberg and Salvi, 1998). However, laboratory studies on rats showed no pathological consequences to acute expositions of chemically inert ultrafine particles (Ziesenis et al., 1998). For the coarse fraction of PM_{10} on the other hand, cytotoxic effects as well as pro-inflammatory cytokine induction were observed (Monn and Becker, 1999) during in vitro experiments on human monocytes.

In respect to chronic symptoms, some studies resulted in an increased number of lung tumor incidents in rats upon exposition to particles (diesel soot, titanium dioxide) in the ultrafine mode (Heinrich et al., 1994b, Heinrich et al., 1994a, Heinrich et al., 1995). Furthermore, particle-bound polycyclic aromatic hydrocarbons (pPAH: e.g. benzo-a-pyrene) emanating from incomplete combustion processes have been observed to create DNA adducts in bronchial epithelial cells that implicate the development of lung cancer (Denissenko et al., 1996). In regard to studies indicating retarding lung clearance rates due to excessive volumetric particle loading in the alveolar region of the lungs (Morrow, 1988, Oberdörster et al., 1992) a recent study suggests that the particle surface area additionally influences the lung clearance rate (Tran et al., 1998).

Ambient aerosols have a specific size distribution based on the type of source present. The particle number distribution of an urban influenced ambient aerosol has its peak between 0.01 µm and 0.1 µm (ultrafine mode). The ultrafine particle mode of a diesel aerosol is made up of carbonaceous soot particles (black carbon) that coexist with a mixture of organic compounds. Some of these compounds, e.g. polycyclic aromatic hydrocarbons (PAH) grow during the combustion process on the surface of these particles (Kasper and Siegmann, 1998). As particle diameters become larger particle number concentrations decrease (Whitby and Sverdrup, 1980). Ultrafine particles that originate via direct emissions from gasoline and diesel combustion coagulate quickly through diffusion (Hinds, 1982). The rate of coagulation is dependent on the square of the particle number concentration. The particle mass distribution of an urban aerosol has a bimodal distribution with a maximum in the accumulation range (0.1-1 or 2 µm) and another in the coarse particle range (> 1 or 2 µm). Particles in the

accumulation mode have a longer residence time than ultrafine particles because removal by diffusion is negligible. These particles grow slowly by coagulation until they exceed 2 µm where sedimentation and impaction become significant (Hinds, 1982). In summary, most particles are found in the ultrafine range below 0.1 µm, whereas the large part of the mass is concentrated in the accumulation range > 0.1 µm to 1-2 µm and coarse range (> 1 or 2 µm) (Whitby and Sverdrup, 1980).

The purpose of this study is to assess airborne particle number concentrations, particle-bound polycyclic aromatic hydrocarbon concentrations (pPAH measured with the Photoelectric Aerosol Sensor), as well as the mass distribution of particulate matter on specific days at various locations differing in traffic intensity and housing characteristics within the city of Basel.

Methods

Field sampling sites

The Basel Air Quality Risk Assessment Project (BRISKA: Basler Risikostudie Aussenluft) is carried out within the framework of the Human-Society-Environment program (MGU: Mensch-Gesellschaft-Umwelt) of the University of Basel. The measuring program lasted throughout 1997 and was supervised by the Basel Air Quality Management Agency (Lufthygieneamt beider Basel LHA). The BRISKA Project continuously measured at six consecutive locations in the urban and industrial (chemical, pharmaceutical) area of Basel City. These locations differed in the degrees of air pollution. The instrumentation was located in a measuring wagon that was transported from one location to another. The locations were chosen based on traffic and building density as well as the population density. Each location was visited for two successive weeks so that a full assessment of the six locations lasted 12 weeks. This 12 week rotation was repeated four times.

Within the BRISKA project the Laboratory for Solid State Physics (LFKP) and the Institute for Hygiene and Applied Physiology (IHA) both from the Swiss Federal Institute of Technology conducted size specific particle number and particle mass measurements on three specific days between 7 August and 1 October 1997. Because of instrument availability and technical considerations, the measurements resulted in three 27- to 28-hour particle number concentration profiles ranging from 0.018 - 0.421 µm and a number of discrete daytime particle mass concentration measurements in the approximate range 0.04-49 µm. This was achieved at three

of the six locations. In addition, particle-bound polycyclic aromatic hydrocarbon (pPAH) and elemental carbon (EC) profiles carried out by the LHA will be shown.

Table 7-1 describes the three locations based on their overall environment and pollution levels throughout 1997 as well as the meteorological conditions during the days the particle number and size specific particle mass measurements were carried out. The annual pollution averages were assessed during the BRISKA project.

Table 7-1: Overall housing, average traffic and pollution characteristics throughout 1997 at three locations within the city of Basel. The pollution data is listed as an average and standard deviation of the measuring period. For the actual measurement days the meteorological conditions are described.

	Luzernerring	**Wasgenring**	**Sevogelstr.**
Housing and traffic			
Housing characteristics	dense	dense	suburban
Heavy duty traffic (per day)[a]	4'920	3'408	264
Light duty traffic (per day)[a]	29'520	16'344	5'040
Pollution[b]	average (SD)	average (SD)	average (SD)
NO [$\mu g\ m^{-3}$]	47.8 (36.0)	35.0 (26.3)	18.9 (22.4)
NO_2 [$\mu g\ m^{-3}$]	45.3 (12.6)	40.3 (18.4)	33.4 (10.2)
CO [$mg\ m^{-3}$]	0.9 (0.4)	0.9 (0.4)	0.7 (0.2)
Elemental Carbon [$\mu g\ m^{-3}$]	2.7 (1.4)	4.0 (3.0)	2.7 (1.4)
PAH [$ng\ m^{-3}$][c]	73.6 (34.4)	32.8 (13.7)	28.5 (14.4)
PM_4 [$\mu g\ m^{-3}$]	28.8 (19.9)	29.9 (28.7)	17.8 (11.6)
PM_{10} [$\mu g\ m^{-3}$]	34.8 (23.7)	30.6 (22.3)	22.4 (12.8)
Date of profile measurements	6. - 7.8.97	30.9. - 1.10.97	25. - 26.8.97
Duration of measurement period	27 hours	28 hours	28 hours
Meteorology	no inversions, no rain, well aired	inversion during night until late morning hours, small wind speeds, sunny after-noon	no inversions, no rain, well aired

[a] Baudepartement and Basel-Stadt, 1994

[b] 52 24-hour averages throughout 1997; the locations were not assessed simultaneously

[c] PAH was averaged out of 26 24-hour averages from 6.7.97 - 31.12.97

Instrumentation

Scanning mobility particle sizer

Particle number versus size distributions from ambient aerosol were measured using a Scanning Mobility Particle Sizer (SMPS version 2.3, TSI Inc., St. Paul MN, USA). This was accomplished in the size range between 0.018 and 0.421 µm mobility diameter at a resolution of 11 channels on a logarithmic diameter axis. The recording time per spectrum was set to 300 seconds assuming constant size distribution in the aerosol during this period. All spectra were measured in sequence, i.e. without delay between two spectra, resulting in a time resolution of 5 min.

In this system, the aerosol is first guided through a ^{85}Kr diffusion charger to establish a well-defined charge distribution on the particles (Fuchs, 1963, Adachi et al., 1985). Subsequently the aerosol enters a differential mobility analyzer (DMA) for mobility classification (Knutson and Whitby, 1975a, Knutson and Whitby, 1975b). A condensation nucleus counter (CNC) is used to determine the number of aerosol particles per unit volume by means of optical counting. The whole system is computer controlled by the SMPS software. This program evaluates the particle size spectrum by relating particle concentrations read from the CNC to mobility diameter values as selected by the DMA.

Quartz Crystal Microbalance Cascade Impactor System

Particle mass versus size distributions of the ambient aerosol were measured using a 10 stage Quartz Crystal Microbalance cascade impactor system (Model PC-2, California Measurements Inc., USA). This impactor system consists of a series of 10 aerodynamic inertial impactors capturing particles in the size range 0.04-49 µm and that are arranged in a cascade with jets segregating the larger aerosol particles on top (Table 7-2). Each stage utilizes „active“ piezoelectric crystal sensors for sample collection within seconds, providing real-time signals proportional to the mass of the aerosol samples. By means of a pump, the aerosol stream is accelerated through the nozzle and impinges on a quartz crystal plate coated with a thin layer of grease. As impacting occurs the oscillation frequency of the crystal decreases. The change of the frequency is a measure for the impacted particulate mass in the specific stage.

Table 7-2: Median cutoff diameters (aerodynamic) of the ten stage Quartz Crystal Microbalance Cascade Impactor (calculated based on the operating instructions)

Stage	1	2	3	4	5	6	7	8	9	10
Median cutoff in µm	0.07	0.14	0.28	0.57	1.13	2.25	4.5	9.0	17.5	35

Photoelectric Aerosol Sensor

A Photoelectric Aerosol Sensor (PAS, Type: LQ1-TV, Matter Engineering Inc., Switzerland) was used to determine the concentration of particle-bound PAH (pPAH) with the method of photoelectric charging of particles. The sensor consists of an electrostatic pre-filter, a UV-irradiation unit and an aerosol electrometer. Air passes through the pre-filter, removing charged particles and ions. In the subsequent quartz tube the aerosol is irradiated by an excimer lamp (0.208 µm); thus, electrons are photo-emitted from PAH coated particles. The photoelectric activity is related to the product of the number concentration, surface area and the coverage of photo-emitting substances. The electrons are removed from the gas by a small electric field whilst the positively charged aerosol particles are precipitated onto a filter. The electric current from the filter is amplified and yields a signal which is proportional to the mass of pPAH (Burtscher and Siegmann, 1994, Niessner and Walendzik, 1989).

Additional Instrumentation

In addition to the provided instrumentation mentioned above, a number of other pollutants were continually assessed throughout 1997 by the LHA. For particulate components, gravimetric analyses of TSP, PM_{10}, PM_4 (high volume sampling), BC (Aethalometer) as well as elemental analyses of PM_{10} (X-ray and coulometric analyses) were carried out. The gaseous pollutants comprise CO, NO_x, SO_2, Benzene, Toluene and Xylene (BTX) and a number of other volatile organic compounds.

Results

Day profiles of particle numbers concentrations

Day profiles of absolute particle number versus size (0.018-0.421 µm) distributions were obtained using SMPS. Figure 7-1 illustrates the concentration profile measured at Luzernerring, an area with a large amount of transit traffic (see Table 7-1) passing by the measuring

unit at a distance of approximately 10 meters. The average total particle number concentration during the measurement period was $2.4 \cdot 10^4$ particles cm^{-3}. The highest one hour concentrations were measured during the evening and morning rush hours (up to $4.8 \cdot 10^4$ particles cm^{-3}), with a minimum during the night ($9.6 \cdot 10^3$ particles cm^{-3}) and a second minimum during the early afternoon ($1.5 \cdot 10^4$ particles cm^{-3}). The majority of the particles (87%) was found in the ultrafine particle range (< 0.1 µm).

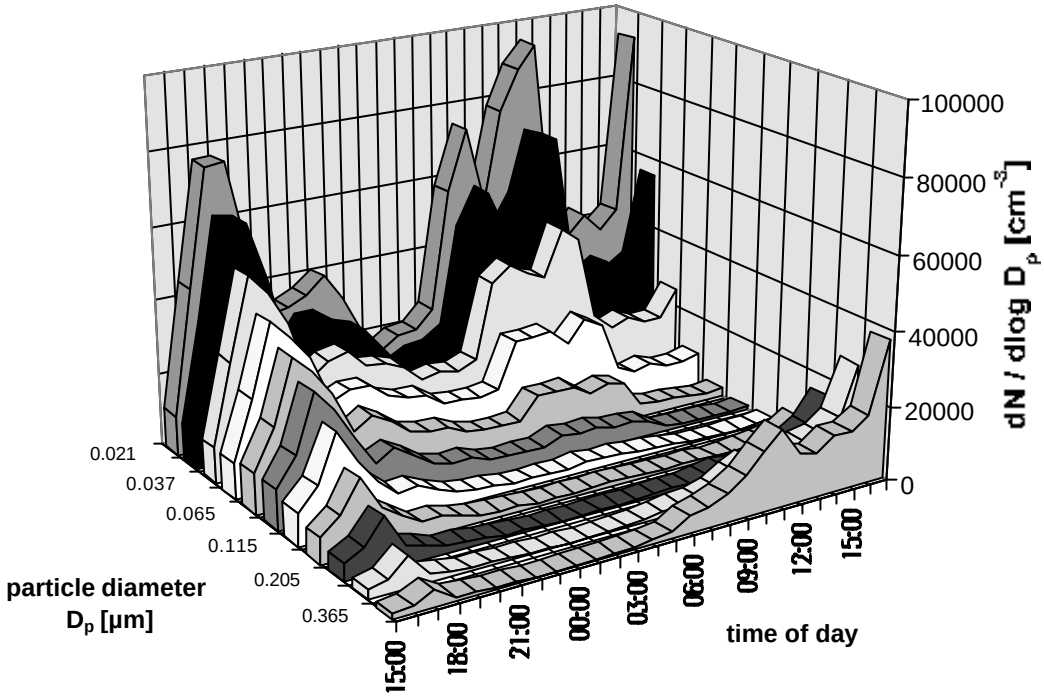

Figure 7-1: Daily profile of absolute particle numbers versus size at Luzernerring measured with SMPS on the 6.-7. August 1997. Note that the diameter axis is on a logarithmic scale.

A similar observation was made one and a half months later at Wasgenring (see Table 7-1), a site with a high degree of truck traffic with an average particle number concentration of $1.4 \cdot 10^4$ particles cm^{-3} (Figure 7-2). The particle number concentrations remained elevated from early morning until noon, with a maximum one hour concentration reaching $2.8 \cdot 10^4$ particles cm^{-3}, a second one hour peak in the evening $1.4 \cdot 10^4$ particles cm^{-3} and a minimum during the night and afternoon respectively of $5.9 \cdot 10^3$ particles cm^{-3}. Again, the majority of the particles lay in the ultrafine range and amounted to 82% of the total particle numbers < 0.421 µm.

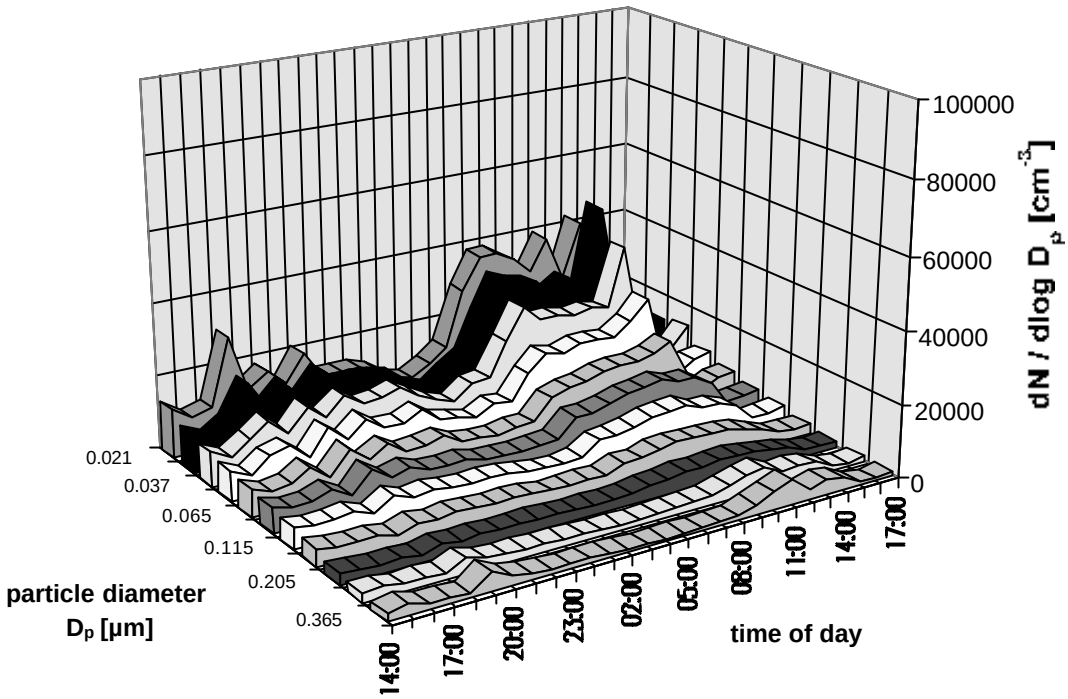

Figure 7-2: Daily profile of absolute particle numbers versus size at Wasgenring measured with SMPS on the 30 September - 1 October 1997. Note that the diameter axis is on a logarithmic scale.

At the residential location at Sevogelstrasse (see Table 7-1) the 28-hour average particle number concentration was low compared to the other sites, amounting to $6.8 \cdot 10^3$ particles cm^{-3}, the highest hourly particle counts were measured at 11 AM ($1.3 \cdot 10^4$ particles cm^{-3}), the lowest ($3.4 \cdot 10^3$ particles cm^{-3}) at midnight. The ultrafine fraction averaged 86 %. The concentrations stay elevated throughout the morning hours, decrease in the afternoon and increase once again in the evening (Figure 7-3).

In general, the profiles of the three locations were similar in quality. Low particle number concentrations were observed throughout the night, while in the early morning hours the concentrations began to rise as the rush hour commenced. These concentrations remained elevated throughout the morning, fell in the afternoon and increased again during the evening rush hour. The highest one-hour average particle number concentrations measured at the three locations were observed in the ultrafine particle range contributing between 62% to 97% of the total particle numbers (< 0.421 µm).

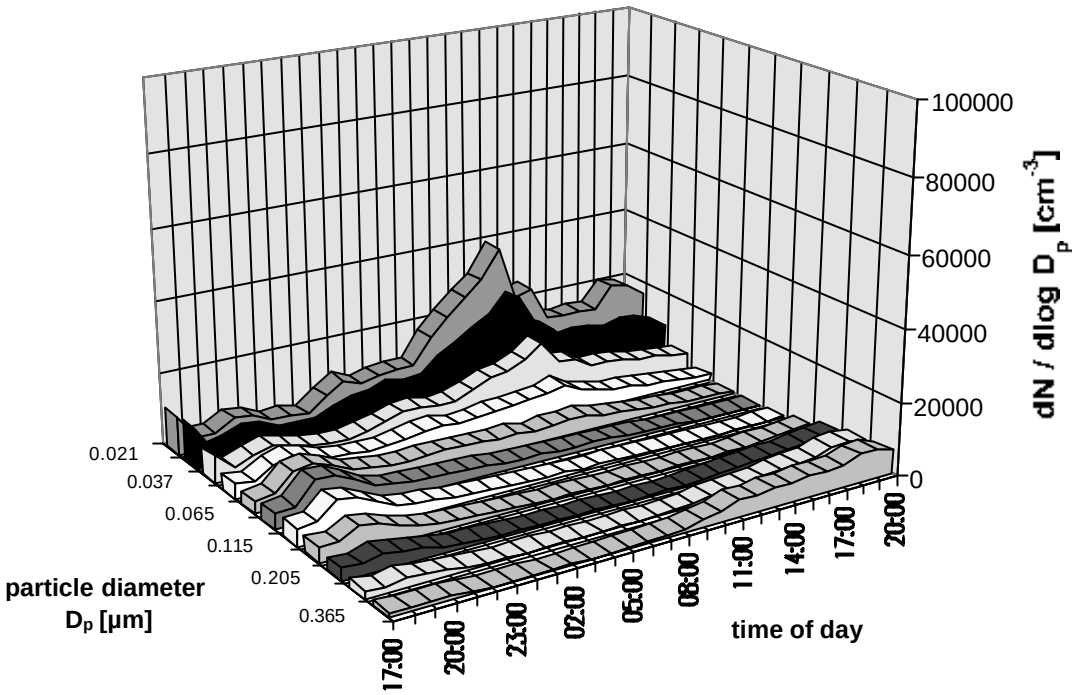

Figure 7-3: Daily profile of absolute particle numbers versus size at Sevogelstrasse measured with SMPS on the 25.-26. August 1997. Note that the diameter axis is on a logarithmic scale.

Day profiles of particle bound polycyclic aromatic hydrocarbons (pPAH), soot (black carbon) and ultrafine particle numbers

Figure 7-4 shows the day profile of three particulate indicators at Luzernerring: pPAH, soot (black carbon (BC)) and ultrafine particle numbers. As can be seen, these pollutants maintain a similar day profile. This applies for Wasgenring and Sevogelstrasse as well, yet absolute concentrations between the sites vary substantially.

Including the data of the particulate indicators at Wasgenring and Sevogelstrasse (hourly concentrations) the profiles for pPAH and soot note a temporal correlation coefficient *r* of at least 0.97 ($p < 0.001$), for pPAH and ultrafine particle numbers an *r* of at least 0.90 ($p < 0.001$) and for soot and ultrafine particle numbers *r* amounts to at least 0.89 ($p < 0.001$). The temporal correlation coefficient for the primary gaseous pollutants NO and CO amount to $r > 0.88$ ($p < 0.001$) and $r > 0.86$ ($p < 0.001$), respectively. The quantitative differences between the three sites are most likely due to different traffic and meteorological conditions (Table 7-3).

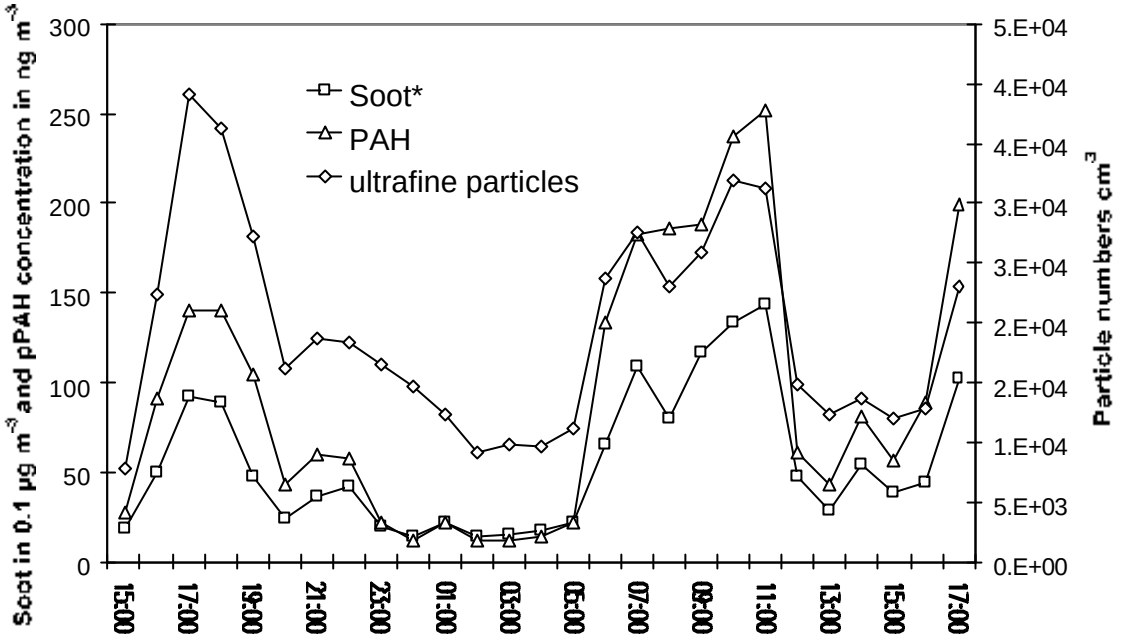

Figure 7-4: **Particulate pollution profile measured at Luzernerring on 6 and 7 August 97. Ultrafine particle number concentrations were measured with SMPS, particle-bound PAH with the PAS Sensor and soot with an Aethalometer.**

*** For better data representation the soot concentration (BC) has been amplified by a factor of 10.**

Table 7-3: **Average pollutant concentrations over a period of 27 - 28 hours measured at three sites in the city of Basel. Standard deviations are indicated in brackets. Measurement at Luzernerring (6 and 7 August 97), Wasgenring (30 September and 1 October 97), and Sevogelstrasse (25 and 26 August 97).**

	Luzernerring	Wasgenring	Sevogelstrasse
NO [µg m^{-3}]	45.4 (44.2)	37.6 (41.5)	19.9 (15.7)
NO_2 [µg m^{-3}]	48.5 (22.1)	52.3 (16.4)	26.8 (15.6)
CO [mg m^{-3}]	0.81 (0.33)	0.84 (0.32)	0.65 (0.18)
EC [µg m^{-3}]	5.5 (3.9)	3.4 (1.7)	1.6 (1.0)
PAH [ng m^{-3}]	92.1 (73.3)	45.5 (31.8)	19.1 (19.8)
ultrafine particles [cm^{-3}]	19’300 (8’580)	10’750 (4’930)	5’610 (2’200)

The highest 1-h pPAH concentrations were measured at Luzernerring in the morning and reached 252 ng m^{-3}. The highest soot concentration was also observed at Luzernerring and noted 14.3 µg m^{-3}.

Traffic versus pollution

Traffic data could not be assessed on the same days the particulate measurements were carried out. However, available traffic data for Sevogelstrasse throughout a two week period from 1-14 September 1997 and for Wasgenring from 1-14 June 1997 (Baudepartement and Basel-Stadt, 1994) show that consistent vehicle numbers exist throughout the week (Figures 7-5 and 6-6).

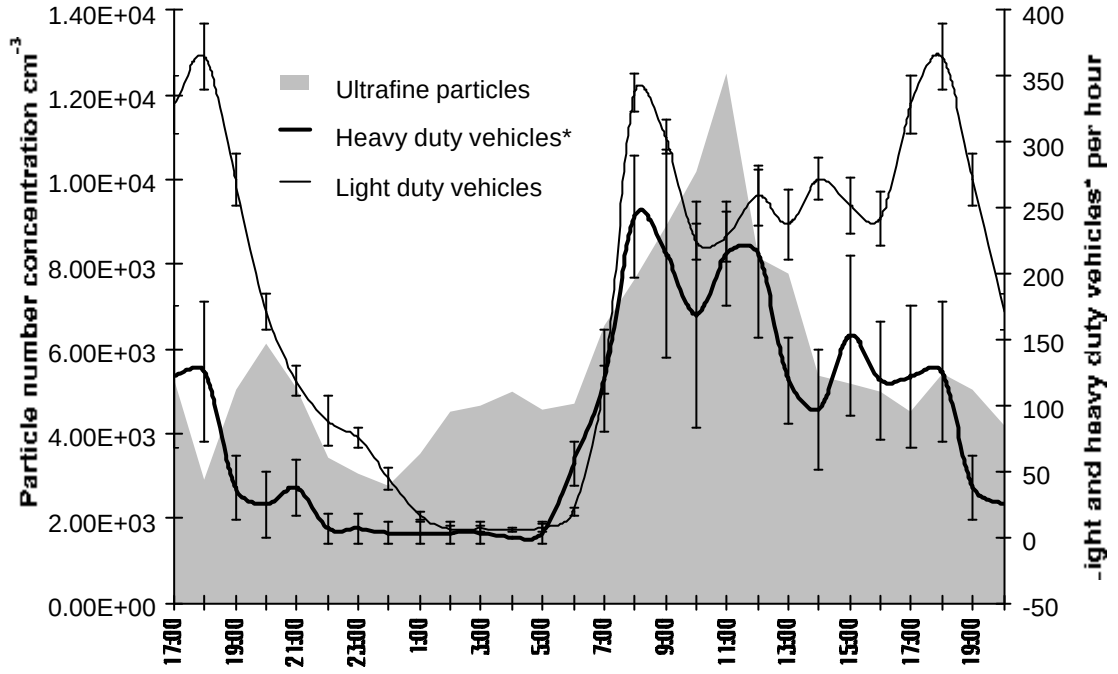

Figure 7-5: Ultrafine particle number concentrations measured at Sevogelstrasse on 25 and 26 August 97 and average light and heavy duty vehicle numbers (± standard deviation) assessed by the Department of Construction Kanton Basel-City out of eight weekdays (twice Monday to Thursday) between 1-14 September 97.

*** For better data representation the heavy duty vehicle numbers were multiplied by 25.**

Furthermore, ultrafine particle number concentrations in relation to the average diurnal traffic density at both locations are shown. It is assumed that on the days the pollution profiles were measured the traffic intensities lay within the range counted in the two week episode. The traffic density is averaged out of 10 weekdays (twice Monday to Friday) for Wasgenring and eight weekdays (twice Monday to Thursday) for the residential site at Sevogelstrasse. The

pollution measurements were carried out from Monday to Tuesday at Sevogelstrasse and from Tuesday to Wednesday at Wasgenring. The traffic data is subdivided into light duty (LD) and heavy duty (HD) vehicles. For better representation of the data, the HD vehicle numbers were multiplied by 25 for Sevogelstrasse and 5 for Wasgenring. For Luzernerring, no traffic data was available. The majority of the vehicles are LD vehicles (98% at Sevogelstrasse, 89% at Wasgenring).

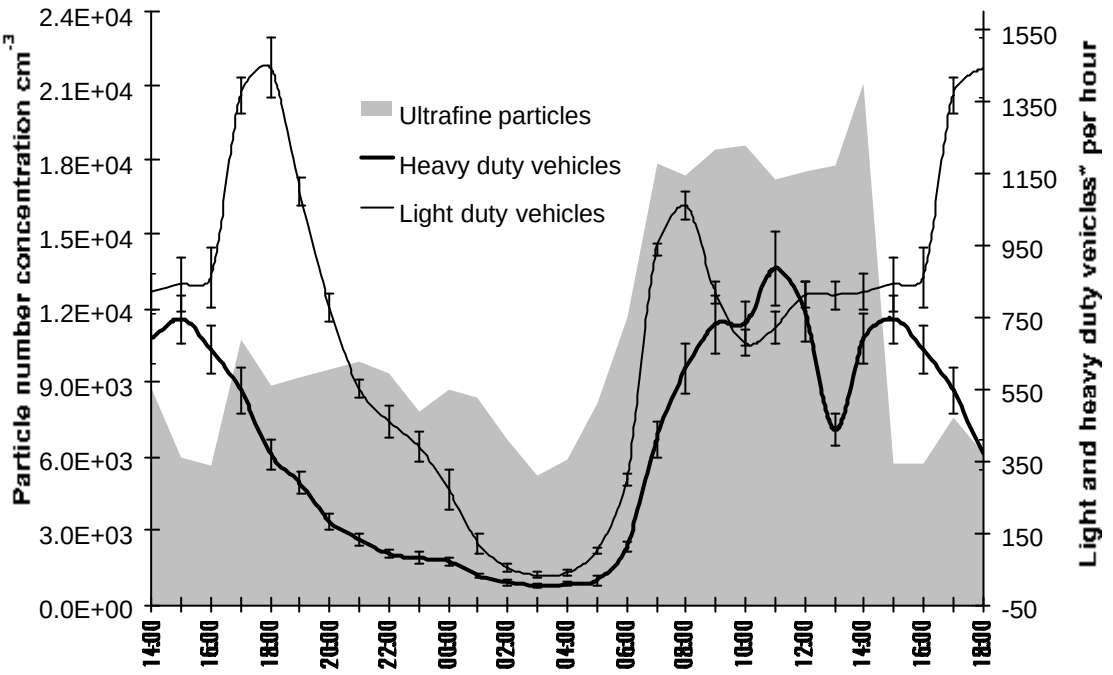

Figure 7-6: Ultrafine particle number concentrations measured at Wasgenring on 30 September and 1 October 97 and average light and heavy duty vehicle numbers (± standard deviation) assessed by the Department of Construction Kanton Basel-City out of ten weekdays (twice Monday to Friday) between 1-14 June 97.

*** For better data representation the heavy duty vehicle numbers were multiplied by 5.**

At Wasgenring as well as at Sevogelstrasse ultrafine particle number concentrations don't significantly correlate to LD traffic ($r = 0.43$, $p > 0.05$; $r = 0.59$, $p > 0.05$). For HD traffic, a significant correlation was found especially at Sevogelstrasse ($r = 0.86$; $p < 0.001$) while at Wasgenring the correlation was not as significant ($r = 0.67$; $p < 0.05$). For soot, a significant correlation to HD vehicles exists at both locations (Wasgenring: $r = 0.83$, $p < 0.001$; Sevogelstrasse: $r = 0.90$ $p < 0.001$) while for LD vehicles a less significant correlation exists only at Sevogelstrasse ($r = 0.68$, $p < 0.01$). For pPAH, a significant correlation is observed for HD vehicles while for LD vehicles a less significant correlation is observed at both locations (HD at Wasgenring: $r = 0.85$, $p < 0.001$; HD at Sevogelstrasse: $r = 0.90$, $p < 0.001$; LD at

Wasgenring: $r = 0.64$, $p < 0.05$; LD at Sevogelstrasse: $r = 0.64$, $p < 0.05$). The LD vehicles were significantly correlated ($p < 0.001$) to the HD vehicles at both locations (Sevogelstrasse $r = 0.86$, Wasgenring $r = 0.79$). At all of the assessed locations wind speeds remained under the limit of detection (0.5 m s^{-1}) throughout the evening, night and morning hours. At noon until 3 PM, wind speeds generally increased but remained under 1 m s^{-1}. No data was available concerning mixing heights. However, meteorological data indicated that no inversion zones were present throughout the measurements at Luzernerring and Sevogelstrasse, for Wasgenring on the other hand, a temperature inversion was registered throughout the night until the late morning hours.

Particle mass distributions

Particle mass distribution measurements were carried out parallel to the particle number measurements. However, due to practical reasons day profiles could not be generated.

Figure 7-7 depicts particle mass concentrations based on data derived from rush hour periods and in between rush hours periods. Mean concentrations are given for the ultrafine (0.04-0.1 µm), accumulation (0.1-2.8 µm) and coarse (> 2.8 µm) modes. In general, mean total mass concentrations were higher during the rush hour periods (37.1 µg m^{-3}) than in between rush hours (25.6 µg m^{-3}). The reason for the differences in mass concentration between the two periods is to be found in the accumulation mode (32.5 µg m^{-3} mean during rush hour, 22.4 µg m^{-3} mean in between rush hours). For the ultrafine and coarse modes mean concentrations were observed to being the same for both periods (2.6 µg m^{-3} and 1.9 µg m^{-3}, respectively). Alternatively, during a 30 minute lawn mowing episode in close vicinity (5 - 25 meters) to the samplers at Wasgenring the absolute mean total particle concentration reached 75.3 µg m^{-3} (mean of the coarse mode: 30.2 µg m^{-3}, mean of the accumulation mode: 41.5 µg m^{-3}, mean of the ultrafine mode: 3.6 µg m^{-3}).

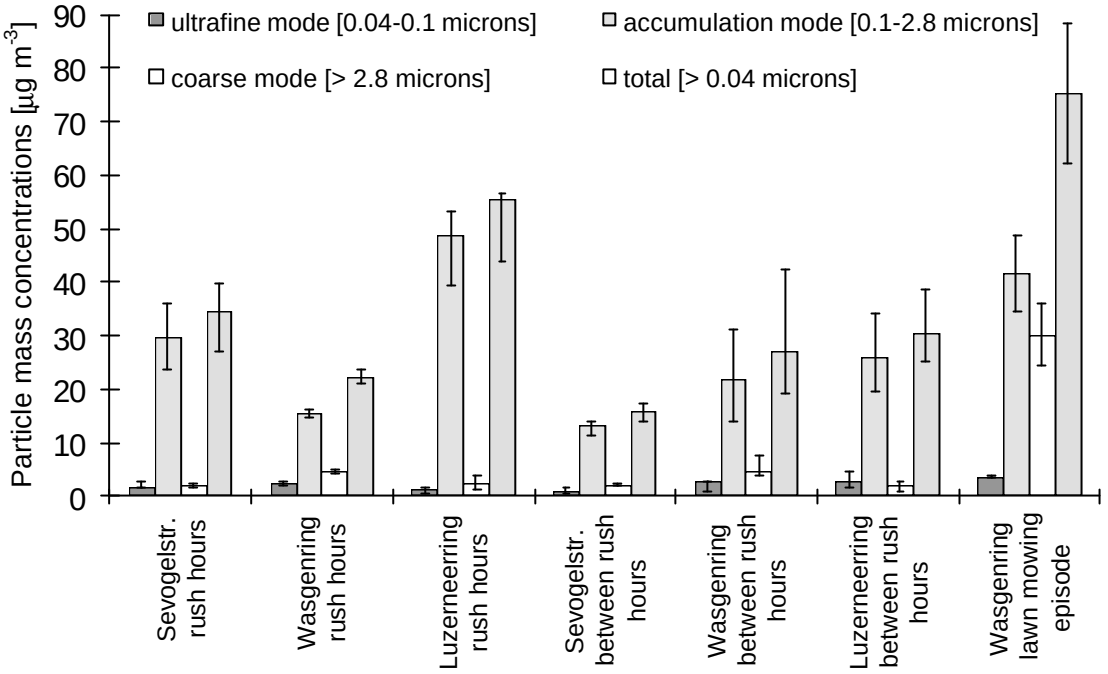

Figure 7-7: Mean particle mass distributions > 0.04 µm measured with the Quartz Crystal Microbalance Cascade Impactor during rush hours, in between rush hours and during a lawn mowing episode. The rush hours for Sevogelstrasse and Wasgenring are based on the diurnal traffic profile under Figures 7-5 and 7-6 (Department for Construction of Basel City). Due to resembling road types, a rush hour profile similar to the one at Wasgenring was assumed for Luzernerring. The size specific stages of the cascade impactor are aggregated to form the following size modes based on an aerodynamic diameter: ultrafine mode (0.04-0.1 µm: impactor stage 1), accumulation mode (0.1-2.8 µm: impactor stage 2-6), coarse mode (> 2.8 µm: : impactor stage 7-10) and the total particle mass mode (> 0.04 µm). The whiskers depicted are 1st and 3rd quartiles. The following data sets have been employed:

- Sevogelstrasse rush hour episode: 9 ten minute values (7:45-9:15) + 4 ten minute values (17:05-17:45) + 8 five minute values (18:05-18:45) on 26 August 1997

- Wasgenring rush hour episode: 5 fifteen minute values (17:20-18:30) on 1 October 1997

- Luzernerring rush hour episode: 10 five minute values (7:15-8:20) on 7 August 1997

- Sevogelstrasse in between rush hours: 14 ten minute values (10:05-12:25) on 26 August 1997

- Wasgenring in between rush hours: 9 fifteen minute values (11:15-13:30) + 9 fifteen minute values (14:30-16:45) on 1 October 1997

-.Luzernerring in between rush hours: 28 five minute values (11:05-13:25) on 7 August 1997

- Wasgenring lawn mowing episode: 2 fifteen minute episodes (14:00-14:15) on 1 October 1997

In respect to the relative particle mass distributions the accumulation mode accounted for most of the particle mass (Table 7-4). Differences between mean rush hour episodes and mean in between rush hours were small. The relative concentration increase in the coarse particle

mass during the lawn mowing episode is to be attributed to visibly whirled up crustal material and debris from lawn mowing activity.

Table 7-4: Mean particle mass distributions during rush hour episodes, in between rush hours and during a lawn mowing episode. 1st and 3rd quartile are depicted. Ultrafine mode: 0.04-0.1 mm, accumulation mode: 0.1-2.8 mm, coarse mode: > 2.8 mm are based on an aerodynamic diameter.

Modes	Rush hour episodes		In between rush hours		Lawn mowing episode	
	Mean (%)	1- 3 quartile (%)	Mean (%)	1-3 quartile (%)	Mean (%)	1-3 quartile (%)
Ultrafine	6.7	5.2-9.9	10.3	6.0-13.3	5.3	4.6-6
Accumulation	88	83.3-90.5	82	74.7-86.6	55.3	55.1-55.4
Coarse	5.2	3.6-7.4	9.2	6.7-12.4	39.5	38.6-40.3

Table 7-5 compares the mass distribution within the accumulation mode between the three episodes. During the lawn mowing episode the majority of the mass is shifted to the greater diameters of the accumulation mode whereas for the rush hour and especially for the in between rush hour episodes the smaller diameters of the accumulation mode account for the main part of the particle mass.

Table 7-5: Mean accumulation mode particle mass distributions during rush hour episodes, in between rush hours and during a lawn mowing episode in percent. The range of the accumulation mode 0.1-2.8 mm is based on an aerodynamic diameter.

	Mass distribution within the accumulation mode in percent		
Accumulation mode sub-range	Rush hour episodes	In between rush hours	Lawn mowing episode
0.1-0.4 µm	49	60	30
0.4-2.8 µm	51	40	70

Discussion

In comparison to an urban influenced ultrafine particle number (< 0.1 µm) background in the United States of about 6'600 particles cm^{-3} (Whitby and Sverdrup, 1980), ultrafine particulate pollution in some locations in the city of Basel (e.g. Luzernerring) is up to 7 times higher than background. The ultrafine particle number concentrations observed within the ambient

environment of the city of Basel is lower than an urban average defined by Whitby and Sverdrup ($1.1 \cdot 10^5$ particles cm^{-3}).

The data for absolute particle numbers in the range 0.018-0.421 µm indicate that distinct day profiles exist at all of the considered locations in this study. Generally, diurnal pollution profiles are altered by variable emissions of the sources, wind speeds and mixing heights. In this study, absolute particle number concentrations vary a great deal from location to location. This is interpreted to most likely be due to differences in traffic intensities between the locations. The generated particle number concentration profiles correspond to the assumed overall daily traffic intensities with peak concentrations in the early morning and late afternoon respectively early evening hours and minimum number concentrations during the night. Wind speeds were measured to be constantly low. It is therefore assumed that wind speed was not a factor responsible for altering the concentration of the pollutants and thus affecting the diurnal profile of the pollution curves. The inversion during the night until the late morning hours at Wasgenring did not indicate to have altered the size specific distribution of the particles throughout the measurement. The majority of the total particle numbers (< 0.421 µm) measured at the three locations lie in the ultrafine mode (< 0.1 µm) and average 82-87%. To what extent differing meteorological conditions contribute to our observations has to be studied in the future. Small wind speeds may elevate particle concentrations through insufficient mixing of air well throughout the day, while inversion zones may reduce the number of ultrafine particles due to coagulation and therefore particle growth into larger size ranges.

In respect to the other particulate pollutant profiles, there was great coincidence between particle number concentrations and continuous pPAH as well as soot concentration measurements. As to the sources responsible the data reveals that heavy duty vehicles are better and more significantly correlated to the particulate pollutants than light duty vehicles. In Switzerland, the vast majority of diesel vehicles are trucks, while hardly any diesel powered automobiles exist. Based on emission factors for particle numbers measured in driving cycle tests at 50 km h^{-1} light duty diesel vehicles emit 200 to 2000 times as many particles as gasoline driven light duty vehicles do (Hall et al., 1998). Although no particle number emission data was found for heavy duty diesel driven vehicles it is assumed that emission factors for HD diesel vehicles would exceed emission factors for LD diesel vehicles. Taking into account the that the number of LD vehicles on average exceeds the number of HD vehicles measured at the locations (109 for Sevogelstrasse, 16 for Wasgenring) the majority of emitted particle

numbers would stem from heavy duty vehicular emissions. For particulate matter mass diesel driven vehicles emit of up to two orders of magnitudes more particulate matter than gasoline driven vehicles (Weingartner et al., 1997). About one third of this particulate matter is attributed to elemental carbon. In California, emission factors for elemental carbon are between one and two orders of magnitude higher for heavy duty diesel trucks than for light duty vehicles while for particle-bound PAHs, emission factors are found to be over an order of magnitude higher for heavy duty diesel vehicles than for light-duty vehicles (Miguel et al., 1998). In general, differences in emission factors for particulate mass and pPAH concentrations are not as great as for particle numbers. The highest one hour pPAH peak concentration observed amounted to 252 ng m^{-3}.

Size specific absolute mass concentrations over time generally revealed the highest concentrations in the earlier morning and evening hours indicating a traffic based relationship. For the relative mass distribution it is shown that under normal circumstances most of the mass (> 82 %) is found in the accumulation mode (0.1 -2.8 µm). However, special activities can alter the mass distribution substantially (e.g. lawn mowing). Percentile differences of the accumulation mode of the particulate mass between rush hour and in between rush hour episodes were insignificant within the 1^{st} and 3^{rd} quartile. However, differences in the mass distribution within the accumulation mode were observed. During the in between rush hour episode most of the mass was observed in the smallest particle diameters of the accumulation mode (0.1 - 0.4 µm), while during the rush hour episode the mass was shifted to greater particle diameters (0.4 - 0.8 µm). This may be due to the coagulation of ultrafine particles forming larger particles. Since the rate of coagulation is dependent on the square of the particle numbers present, coagulation is more efficient during rush hour episodes than during the in between rush hour periods.

From the health effects point of view, there is evidence that lung function values are more strongly associated to truck traffic density than to automobile traffic density (Brunekreef et al., 1997). In a further study, truck traffic intensities and the concentration of black smoke were found to be significantly associated to chronic respiratory symptoms such as cough, wheeze, runny nose and asthma (van Vliet et al., 1997). Furthermore, heavy vehicular traffic was associated to an increasing occurrence of lower respiratory tract infections early in a child's life while at school age wheezing and bronchitic symptoms became more evident (Ciccone et al., 1998). In addition to a potentially elevated long term risk, truck intensive

morning and early afternoon hours may increase the short term health risk for lung inflammation (Blomberg and Salvi, 1998).

Additional high resolution long-term monitoring of submicron particle numbers and particle mass distribution measurements as well as high resolution traffic measurements should be carried out in order to statistically validate the underlying findings. Furthermore, new findings regarding the long term health effect of the particle surface area on alveolar particle clearance as well as the long and short term influence of diesel exhaust fumes should be taken into consideration for future epidemiological studies.

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Chapter 8

Temporal and Spatial Variation of the Chemical Composition of PM_{10} at Urban and Rural Sites in the Basel Area, Switzerland

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Abstract

Particulate matter measurements of different size fractions (PM_4, PM_{10}, TSP) were performed in the Basel area (Switzerland) at seven urban sites throughout 1997 and at two urban and two rural sites during the following year (April 1998 to May 1999). Based on a sample of filters which was chemically analyzed, we investigated the chemical composition of PM_{10} both within the city of Basel and among urban and rural sites. The temporal and spatial variability of the chemical composition of PM_{10} was evaluated taking into account additional data from meteorology and further air pollutants.

The chemical analyses of PM_{10} showed that carbonaceous substances (elemental carbon, organic matter) and inorganic substances of secondary origin such as sulfate, nitrate and ammonium were the most abundant component of PM_{10} in the Basel area (approximately 60-70%).

Difference in the PM_{10} concentration between urban and rural sites was larger during the cold season than during the warm season. This was mainly due to the presence of an inversion layer between the city and the more elevated rural sites resulting in higher concentrations of nitrate, ammonium and organic matter in the city during the cold season. The higher PM_{10} concentration on workdays compared to weekends was mostly a result of the temporal variation of the concentration of Ca, elemental carbon, Ti, Mn, and Fe, indicating that these compounds are for the most part caused by regional human activities.

Although total PM_{10} mass concentration was found to be in general uniformly distributed within the city of Basel, the chemical composition was more variable due to specific sources like road traffic and other anthropogenic emissions.

Introduction

In recent years, epidemiological studies have shown associations between ambient particulate matter concentration and health. Exposure to increased levels of particulate matter concentrations is related to increased mortality and a number of pulmonary effects, both acute and chronic (Ackermann-Liebrich et al., 1997, Dockery et al., 1993). Several analyses have revealed more consistent correlations for the concentration of the fine ($PM_{2.5}$) and inhalable (PM_{10}) particles with health effects than for any other air pollutant. (Schwartz, 1994a, Schwartz et al., 1996).

Ambient airborne particulate matter can be either of primary or secondary origin. Primary particles are directly released into the atmosphere. Secondary particles are formed within the atmosphere by gas-to-particle conversion from gaseous precursor substances such as sulfur dioxide (SO_2), nitrogen dioxide (NO_2), ammonia (NH_3) and volatile organic carbon compounds (VOC). Secondary particles are mainly small particles. Aerosol particles originate from both natural and human sources. Typical examples of particles from natural sources are sea salt particles or wind blown dust. Combustion aerosols and particles from industrial processes are expected to be large contributors of human sources (Baltensperger and Nyeki, 1998).

The chemical composition of particulate matter may vary within a broad range according to the sources of the particles and the conditions of their dispersion as was shown in the aerosol measurements from the IMPROVE Network at 36 stations throughout the United States (Malm et al., 1994, Eldred et al., 1997) and other studies (e.g. Chow et al., 1994, Brook et al., 1997, Müller, 1999). The purpose of this study was to analyze the chemical composition of PM_{10} in the region of Basel at urban and rural sites. Moreover, we have evaluated influences from meteorology and from emission sources on the temporal and spatial variability of the chemical composition of PM_{10}.

Methods

Air pollutant measurements

In the present analyses two sets of data were used. The first set includes the measurements which were performed within the BRISKA study (Basel Risk Assessment Study of Ambient Air Pollutants) from 22 January 1997 to 5 January 1998. Twenty-four-hour mean concentrations of PM_4 (measured only from March, 1997, onwards), PM_{10}, and TSP were measured at six sites within the city of Basel by a mobile monitoring station rotating every 2 weeks. This resulted in one 13-day sampling period for each temporary site per season (52 measurements in total per site). The six temporary sites differed with respect to traffic density and were assumed to be representative of the traffic exposure of the Basel population (Table 8-1). The maximum distance between these six sites was 3.3 km. In addition, PM_{10} was permanently measured at one fixed monitoring station of the Air Quality Management Agency of Basel (site 'C' in Table 8-1), representing urban background.

The second set of data was recorded from 1 April 1998, to 31 May 1999, at two rural and two urban sites. In addition to site 'C' (urban background), a monitoring station was located in a street canyon with high traffic density near a traffic light at a crossroad (site 'D'). Rural sites 'A' and 'B' were situated 25 and 20 km southeast of the city. Their altitude was 900 m and 600 m above sea level, respectively (Basel: 250 m). Concurrent measurements of 24-h mean concentrations of PM_{10} were performed at all four stations.

Table 8-1: Overview of the database and the characteristics of the measurement sites. Traffic counts originate from the traffic inventory of the city of Basel (Baudepartement and Basel-Stadt, 1994)

Date	PM	Sampling period	Type	Site	Passenger cars/24h	Heavy duty vehicles/24h	Comments
1/22/97 - 1/5/98	PM_4[b], PM_{10}, TSP	Permanent	urban	**C**	11.500	950	Urban background[a]
		Temporary monitoring station, 13 days per season at each site		**1**	5.000	260	Residential
				2	5.200	370	Residential
				3	6.200	500	Center of Basel
				4	16.300	3.410	High truck quota
				5	27.100	1.130	Sloped lane
				6	29.500	4.920	Cross-way
4/1/98 -5/31/99	PM_{10}	permanent	urban	**D**	17.000	890	Street canyon, traffic light
				C	11.500	950	Urban background[a]
			rural	**B**	no traffic		Altitude: 600 m, 20 km SE from Basel
				A			Altitude: 900 m, 25 km SE from Basel

[a] Monitoring site of the Air Quality Management Agency
[b] Measured only from 1st March, 1997, onwards

Twenty-four-hour mean particulate matter concentrations were measured with high volume samplers (HVS). The samples were collected on quartz filters (QF 20 Schleicher & Schuell), and changed automatically at 0.00 a.m. (GMT+1h, wintertime). The details of the sampling procedure is described elsewhere (Röösli et al., 2000a). Data quality control of the measurements revealed that actually PM_4 instead of $PM_{2.5}$ had been recorded. This was due to errors in the calculation of the geometry of the device's head which had led to a larger cut-off. To assess how comparable the PM_4 measurements were with the more common measurement of $PM_{2.5}$, an EPA-WINS Impactor used for the EXPOLIS-Basel study (Oglesby et al., 2000) was

repeatedly installed at a fixed site monitoring station. The measurements indicated that PM_4 levels are highly correlated with $PM_{2.5}$ levels though on average 11% higher than the latter (25.2 vs. 22.3 µg m^{-3}).

The coarse fractions were determined by calculating the differences between the three size fractions ($PM_{(10-4)}$, $PM_{(TSP-10)}$, $PM_{(TSP-4)}$).

Chemical analyses

Due to limited financial resources the chemical analyses of the particulate matter filters had to be reduced to a subset of the filters (Table 8-2): In 1997, mean concentrations of NO_3^-, SO_4^{2-}, NH_4^+, As, Cl, Pb, Zn, and Cd were determined by analyzing the 13 quartz filters of each period during which the monitoring car was located at one temporary site as a mixed sample. In 1998/99, mixed samples of every fourth PM_{10} filter were used to determine monthly mean values of these compounds. For the mixed samples the selected filters were shredded to small pieces (1-2 cm^2), macerated with nitric acid (65%), and centrifuged. The ammonium analysis was done by standard spectrophotometry using a LAMBDA 2, Perkin-Elmer spectrometer. Pb, Cd, and Zn were quantified by inductively coupled plasma atomic emission spectroscopy (ICP-AES, Philips PV 8060). Arsenic (As) was measured by atom absorption spectroscopy using a Perkin Elmer 110b instrument. Sulfate, nitrate, and chloride were analyzed by means of ion chromatography (DIONEX 2000i SP). All calibrations were made with standard solutions from Merck. These analyses were performed by the Cantonal Environmental Laboratory in Liestal, Switzerland.

The chemical analyses of elemental (EC) and organic carbon (OC) concentrations of the 1997 data were performed in a laboratory at the Technical University of Berlin, Germany, and the 1998/99 data were analyzed by UMEG (Gesellschaft für Umweltmessung und Umwelterhebung GmbH) in Karlsruhe, Germany. In 1997 EC and OC concentrations were determined as daily mean values from filters collected during 1 week out of a 13-day measurement period. In 1998/99 monthly mean values were analyzed based on a sample consisting of every fourth filter. EC and OC were quantified with a thermographic method. The filters (d=30 mm) were burned in an oxygen atmosphere. The released carbon compounds were measured after catalytic oxidation as CO_2 using IR spectroscopy. In a first step the sample was heated to 620°C and the released carbon compounds were determined as OC. Further heating to 700°C resulted in the quantification of EC. Organic matter concentration (OM) was obtained by

multiplying OC concentration with a factor of 1.4, commonly reported in the literature (e.g. Eldred et al., 1997, Pryor et al., 1997, Brook and Dann, 1999).

Throughout 1997 black carbon (BC) was measured every day at the six temporary sites with an optical method (Aethalometer AE-10i2M). With these BC values, EC concentrations were calculated for all missing days based on a regression model of black carbon and EC.

Energy-dispersive X-ray fluorescence spectrometry (ED-XFR) was used for analyzing elemental concentrations from 80 PM_{10} filters from the permanent urban site 'C' as well as from 21 PM_4, PM_{10} and TSP filters of each temporary monitoring site (3 complete weeks per site) resulting in a total of 458 analyzed filters. However, the first 10 TSP filters analyzed from the temporary monitoring sites as well as the first 21 PM_4 filter could not be used due to technical problems. These analyses were carried out at the Institute for Mineralogy and Petrography of the University of Basel with a 'Spectrace 5000' instrument (Spectrace Instruments, 1993). Detection limits and sensitivity can be found in Kneifel, 1998. The concentrations of S, Cl, K, Ca, Ti, Cr, Fe, Pb, Cu, Zn, Ba, Sb, Ni, Mn, Br, and Mo were determined. The standards used were A1 and B1 (BCR-CRM128-Standard, Commission of the European Communities and NIST3087a (US Department of Commerce). From the 458 analyses for each element, the following numbers of measurements were found to be below the detection limit: 1 (Cl), 1 (K), 1 (Cu), 29 (Pb), 55 (Mn), 56 (Mo), 63 (Br), 111 (Cr), 235 (Ni), 252 (Sb) and 273 (Ba). Values below the detection limit were replaced by half the value of the detection limit.

In order to assess the unidentified proportion of $PM_{10,}$ elements were assumed to be present as chemical compounds, in oxidized form. An exception being calcium which was assumed to be present as carbonate because $CaCO_3$ is the main component of the Earth's crust in the Basel area. The following substances were supposed to be present: NaCl, K_2O, $CaCO_3$, TiO_2, NiO, Cr_2O_3, MnO_2, Fe_2O_3, Br^-, PbO, Cu_2O, ZnO, MoO_2, Sb_4O_6, and $BaCO_3$. This assumption is based on published literature (e.g. Malm et al., 1994, Horvath et al., 1996, Brook et al., 1997).

Various polycyclic aromatic compounds (PAH) in the PM_{10} fraction were analyzed from the 1997 data (Table 8-2). For each measurement period (13 days) mean values of the PAH compounds were calculated by analyzing a sample with gas chromatography and mass spectroscopy (GC/MS) using a Carlo Erba 5300/Finnigan ITD 800 instrument.

Table 8-2 summarizes for each substance the number of filters analyzed and the resulting number of mean values.

Table 8-2. Description of the chemical analyses and the number of available measurements for each compound.

Analyzed Species	Measuring device	Number of resulting mean values (n)	
		Dataset I: 22 Jan. 1997 - 5 Jan. 1998 temporary sites '1'-'6' and site 'C'	Dataset II: 1 Apr. 1998 - 1 May 1999 sites 'A' to 'D'
SO_4^{2-}, NO_3^-, Cl^-	ion chromatography (DIONEX 2000i SP)	n = 4 period mean values per temporary site (one per season).[a]	n = 14 monthly mean values per site.[b]
Pb, Zn, Cd	ICP-AES (Philips PV 8060)		
NH_4^+	Spektralphotometry (LAMBDA 2)		
As	Perkin Elmer 1100B		
EC (elemental carbon) OC (organic carbon)	calorimetric method	n=28 daily mean values per temporary site.[c]	n = 14 monthly mean values per site.[b]
BC (black smoke)	optical method (Aethalometer AE-10i2M)	n=52 daily mean values per temporary site.	not analyzed
various PAH[d]	GC/MS (Carlo Erba 5300/Finnigan ITD 800)	n = 4 period mean values (=13 days) per temporary site (one per season).[a]	not analyzed
S, Cl, K, Ca, Ti, Cr, Fe, Pb, Cu, Zn, Ba, Sb, Ni, Mn, Br, and Mo	ED-XFR (Spectrace5000)	n=21 daily mean values per temporary site n=80 daily means for site 'C'.[e]	not analyzed

[a] Mean concentration based on a mixed sample of all 13 daily PM_{10} filters from one measurement period at one temporary monitoring site.

[b] Monthly mean concentration analyzing a mixed sample of 7 PM_{10} filters consisting of every fourth filter.

[c] Analyzing daily mean concentrations on the PM_{10} filters from every other week.

[d] Cyclopenta[c,d]pyrene, benzo[b]naphto[2,1-d]thiophene, benzo[a]anthroacene, benzo[k,b]fluoranthene, benzo[a]pyrene, benzo[e]pyrene, perylene, benzo[ghi]perylene, dibenz[a,h]anthracene, indeno[1,2,3-cd]pyrene, anthranthene and coronene.

[e] Analyzing daily mean concentrations on the PM_4, PM_{10} and TSP filters of the temporary sites from every other week (excluding period from 30. April to 21. July) as well as analyzing 80 PM_{10} filters from site 'C'.

Data analysis

(i) We compared the elemental composition in three size fractions to evaluate the particle size range in which the elements were predominately located. Due to missing PM_4 values in January and February 1997, ratios of the elemental concentrations in the three size fractions were calculated from the data recorded at the temporary sites from March onwards. In order to avoid ratios in the calculation which represented the ratio of the detection limits in the three size fractions, only days on which the elemental concentrations in all three size fractions were above the detection limit were taken into account. As the distribution of ratios is asymmetrical, mean values were obtained by taking the logarithm of the ratios and then exponentiating that value.

(ii) Concentrations of selected compounds on PM_{10} filters during workdays and weekends were compared in order to evaluate the short-term influence of regional human activities such as road traffic, construction work, and industrial processes. For this calculation, only measurements available as daily mean values could be used (EC, OM, elements). For these substances, data from 18 weeks distributed between January 1997 and January 1998 were analyzed.

(iii) Pearson correlation coefficients were calculated between concurrently performed measurements as an indicator for small scale spatial variability within the city of Basel. Variability was assessed based on 41 daily mean values of the elemental concentrations from the fixed urban monitoring site 'C' and from one of the temporary monitoring sites (from the same day). Larger scale spatial variability between urban and rural sites was quantified with the coefficient of variation. The coefficients of variation were obtained by dividing the standard deviation of the mean concentrations from the four sites by the mean value across the four sites.

Results and discussion

The chemical composition of PM_{10} within Basel

Table 8-3 summarizes the seasonal and average chemical composition of PM_{10} in Basel in 1997 and the relative proportion of each substance. Each seasonal mean value represents an average of the six 13-days value of each temporary monitoring site. OM was found to be the main constituent of PM_{10}. Another substantial part of PM_{10} was composed of sulfate, nitrate, and ammonium, the components of secondary origin, as well as primarily emitted EC. Most

prominent elements were Fe, Ca, K and Cl. Assuming that elements were present as chemical compounds (generally in oxidized form), approximately 11% of the PM_{10} mass concentration could not be identified.

Table 8-3. Chemical composition of PM_{10} within the urban environment of Basel between 22 January 1997 and 5 January 1998. Each seasonal mean value (first four columns) is a mean value from the six temporary monitoring sites. The fifth column shows a mean value for the three seasons with available data for all compounds, followed by a minimum and maximum measurement period value. Percentage proportions are shown in brackets.

	Winter mean 22 Jan.-28 Apr.		Spring mean 30 Apr.-21 Jul.		Summer mean 23 Jul.-13 Oct.		Autumn mean 15 Oct.-5 Jan.		Mean value of col. 1, 3, 4*		Min. period mean	Max. period mean
	[µg m^{-3}]	([%])	[µg m^{-3}]	([%])	[µg m^{-3}]	([%])	[µg m^{-3}]	([%])	[µg m^{-3}]	([%])	[µg m^{-3}]	[µg m^{-3}]
PM_{10}	31.31	(100)	16.53	(100)	22.40	(100)	31.14	(100)	28.28	(100)	13.48	56.10
OM	7.21	(23.0)	4.91	(29.7)	6.20	(27.7)	7.62	(24.5)	7.01	(24.8)	3.76	12.04
SO_4	4.30	(13.7)	3.03	(18.3)	4.10	(18.3)	4.16	(13.4)	4.19	(14.8)	1.67	6.48
NO_3	5.50	(17.6)	0.65	(3.9)	1.08	(4.8)	5.49	(17.6)	4.02	(14.2)	0.23	12.01
EC	3.47	(11.1)	2.20	(13.3)	2.86	(12.8)	4.55	(14.6)	3.63	(12.8)	1.30	9.05
NH_4	2.35	(7.5)	0.86	(5.2)	1.18	(5.3)	2.25	(7.2)	1.93	(6.8)	0.59	4.32
Fe	0.74	(2.4)	NA		0.76	(3.4)	0.65	(2.1)	0.72	(2.5)	0.37	1.18
Ca	0.67	(2.1)	NA		0.72	(3.2)	0.50	(1.6)	0.63	(2.2)	0.24	1.29
K	0.48	(1.5)	NA		0.63	(2.8)	0.40	(1.3)	0.51	(1.8)	0.26	1.74
Cl	0.26	(0.83)	0.03	(0.17)	0.041	(0.19)	0.47	(1.5)	0.26	(0.90)	0.01	0.73
Ba	0.13	(0.41)	NA		0.11	(0.49)	0.10	(0.34)	0.11	(0.41)	0.054	0.223
Zn	0.088	(0.280)	NA		0.073	(0.327)	0.090	(0.290)	0.084	(0.296)	0.049	0.135
Cu	0.075	(0.239)	NA		0.075	(0.335)	0.069	(0.221)	0.073	(0.258)	0.048	0.109
Mo	0.080	(0.257)	NA		0.057	(0.254)	0.058	(0.186)	0.065	(0.230)	0.042	0.163
Pb	0.059	(0.190)	NA		0.051	(0.229)	0.053	(0.170)	0.055	(0.193)	0.023	0.109
Ti	0.033	(0.106)	NA		0.038	(0.171)	0.024	(0.077)	0.032	(0.112)	0.023	0.109
Sb	0.021	(0.068)	NA		0.029	(0.129)	0.023	(0.073)	0.024	(0.086)	0.015	0.071
Br	0.012	(0.038)	NA		0.011	(0.048)	0.030	(0.095)	0.017	(0.062)	0.005	0.086
Mn	0.018	(0.058)	NA		0.016	(0.073)	0.013	(0.041)	0.016	(0.056)	0.008	0.028
Ni	0.007	(0.024)	NA		0.008	(0.036)	0.010	(0.032)	0.009	(0.030)	0.004	0.014
Cr	0.008	(0.024)	NA		0.008	(0.034)	0.009	(0.030)	0.008	(0.029)	0.003	0.015
PAH	0.007	(0.022)	0.001	(0.006)	0.002	(0.009)	0.013	(0.041)	0.007	(0.026)	0.001	0.019
As	0.001	(0.002)	0.000	(0.002)	0.001	(0.003)	0.001	(0.004)	0.001	(0.003)	0.000	0.003
Cd	0.001	(0.002)	0.000	(0.002)	0.000	(0.002)	0.001	(0.002)	0.001	(0.002)	0.001	0.019
O†	1.86	(5.9)	NA		1.85	(8.3)	1.64	(5.3)	1.78	(6.3)	1.06	3.00
unident.	3.93	(12.6)	4.85	(29.4)	2.50	(11.1)	2.90	(9.3)	3.11	(11.0)	-1.18	9.38

* No ED-XFR performed between 30 Apr. and 21 Jun.

† Total mass of all oxygens assuming elements were present in oxidized form as well as CO_3 from $CaCO_3$.

PM_{10} was clearly higher during winter (October-March) than during the warm months. This seasonal pattern was highly distinctive for chloride, nitrate, ammonium and PAH. For K, Ti, Ca, Sb, and Fe, higher relative proportions were recorded during summer (July to September) than during the cold months.

Considerably elevated concentrations of nitrate, ammonium, chloride and PAH during the cold season have also been reported by other studies (e.g. Flessel et al., 1991, Puxbaum et al., 1993, Müller, 1999). This finding can be explained with the temperature dependent sampling efficiency of nitrate, ammonium and PAH (Wang and John, 1988, Chang et al., 2000). The substantially increased chloride values during winter may be an effect of the resuspension of road salt (Gälli Purghart B.C., 1990, Harrison, 1997, Clarke et al., 1999b) or due to a larger proportion of chloride in the particles originating from sea during winter, as volatilization of chloride from sea salt particles is much lower in winter than in summer (Pio and Lopes, 1998).

The distribution of the elements in the three size fractions

The analysis of 105 daily samples of each particulate matter size fraction (PM_4, PM_{10} and TSP) from March 1997 to January 1998, showed that from all particles in the TSP fraction, 60% were smaller than 4 µm and 77% were smaller than 10 µm. Some compounds such as S, Cu, Br, Pb, and Zn mainly occurred in small particles (Figure 8-1). Ca, Fe, Ti, and Cl were predominately found in the coarse fraction (> 4 µm). During summer the proportion of the small particles was smaller than during winter. From March to October 1997 the proportion of PM_4 in TSP was only 57% and the proportion of PM_{10} in TSP 74% compared to 68% (PM_4 in TSP) and 84% (PM_{10} in TSP) from November1997 to January 1998. This seasonal pattern was observed for all analyzed elements. It was especially distinct for Cl and K (Figure 8-1).

The high proportion of Ca, Fe and Ti in the coarse fraction leads to the conclusion that they were mostly mechanically generated. They are known mineralogical elements (Fergusson, 1992, Chan et al., 1997), whereby other sources than the Earth crust may also be important, such as brake lining dust for Fe (U.S.EPA, 1990) or cement plants for Ca (Lee and Pacyna, 1999). In contrast, elements found in the small fraction (S, Cu, Br, Pb) may be secondary produced or originating from combustion processes. These types of emission sources seems to be larger during the cold season in Basel, as the proportion of small particle was increased. The observation that K was mostly found in the small fraction during winter but in the coarse fraction during the warm season may suggest two different sources for this element depending

on the season. As K originates probably mostly from biological material (U.S.EPA, 1990) it may be concluded that during winter K originates mainly from wood combustion (heating), whereas during summer direct emission of biological material such as pollen are more important. From the substantial higher proportion of Cl in the small fraction during the cold months compared to summer it might be speculated that during the cold season Cl deriving from HCl may contribute to the particulate chloride additional to Cl coming from road and sea salt. This speculation is supported by a recent study of Kaneyasu et al., 1999, identifying events with extremely high chloride concentrations which were caused by high HCl concentrations originating from refuse incineration or coal combustion in Japan.

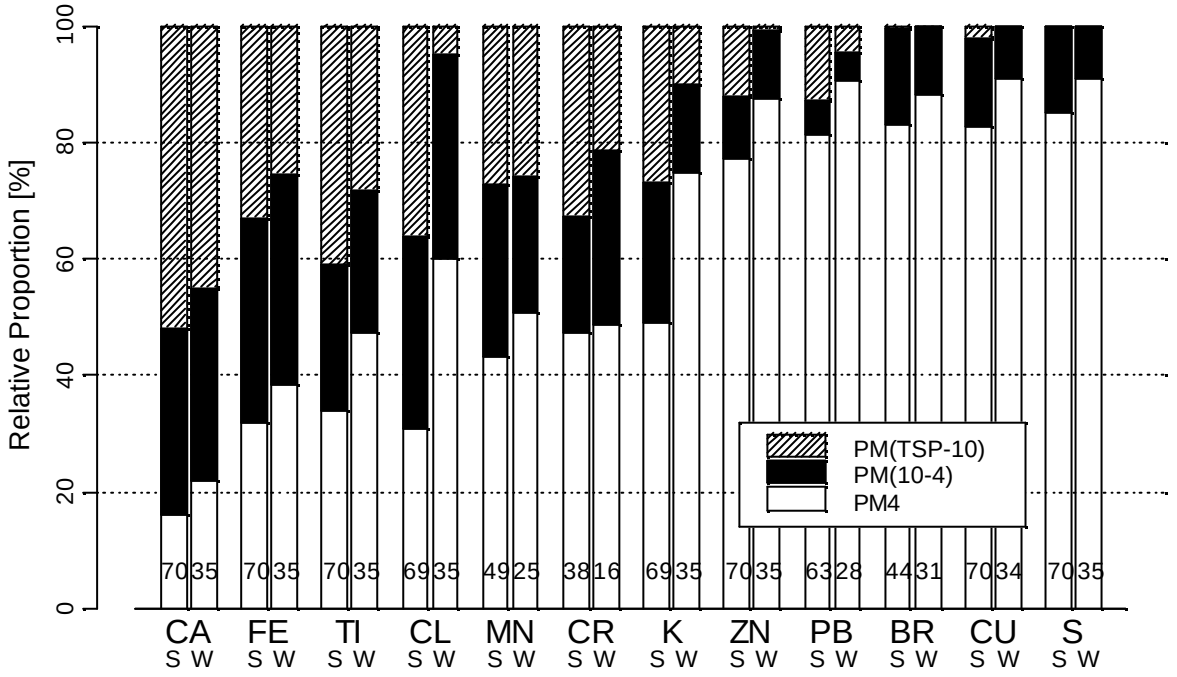

Figure 8-1: Mean proportion of elemental concentrations in the PM_4 and PM_{10} fractions compared to elemental concentration in the TSP fraction during March to October 1997 (S) and during November 1997 to January 1998 (W). The bottom numbers denote the number of samples available for analyses (above detection limit).

Comparison of PM_{10} concentrations during workdays and weekends

The ratio between mean concentration during workdays and mean concentration on weekends was above 1 for all compounds (Figure 8-2) reflecting the influence of regional human activities such as road traffic, construction work, and industrial processes which is generally

higher during workdays. Highest workday/weekend ratios were found for mineralogical elements (Ca, Ti, Mn, Fe) and EC, lowest ratios for OM, Pb, Cl, Cu. Generally, compounds mainly found in small particles (Figure 8-1) showed a smaller workday/weekend ratio than total PM_{10} concentration, and compounds which mainly occurred in the coarse fraction showed bigger ratios. This might be explained by the longer atmospheric residence time of small particles compared to larger particles. However, the ratio of EC, which is expected to be mainly present in small particles (EC was only analyzed on PM_{10} filters), was quite high.

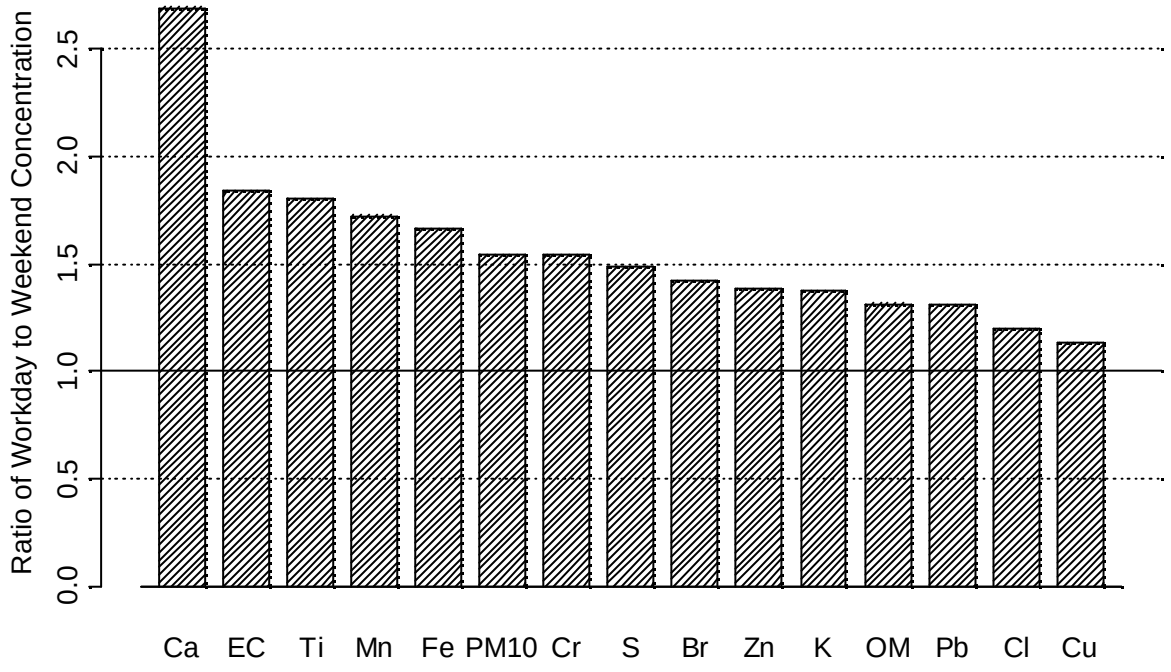

Figure 8-2: Ratio of mean concentration during workdays to mean concentration on weekends for certain substances and PM_{10} concentration. Analyzed samples consisted of daily measurements from 18 separate weeks between January 1997 and January 1998 in Basel.

As passenger cars with diesel engines are not common in Switzerland, trucks and off-road vehicles are considered as the main sources of EC (Weingartner et al., 1997). The large difference in the concentration between workdays and weekends is therefore probably a result of strong restrictions for heavy duty vehicles on weekends in Switzerland. The pronounced difference between the concentrations of Ca, Fe, Ti and Mn during workdays compared to weekends suggests that they may predominantly originate from man-made mechanical processes such as resuspension from road traffic or construction work which are more

prevalent during workdays. This conclusion is consistent with the findings of Chan, 1999, who reported that road side dusts can contribute to more than half of the crustal matter. In Basel wind erosion may be a minor source of these mineralogical elements as the concentration of these elements in all three size fractions was negatively correlated with daily mean and maximum wind velocity.

This elemental analysis of the workday/weekend ratios was restricted to the subset of filters with available elemental analyses. In this subset the PM_{10} concentration was 54% higher on workdays than on weekends (Figure 8-2). However, taking into account all PM_{10} measurements from 1997 the mean PM_{10} concentration would be only 17% higher during workdays than the mean concentration on weekends. This suggests that the workday/weekend ratio of our sample is not only influenced by human activities but also by meteorology. Nevertheless, the order of the ratios of the different substances may be caused mostly by human activities as meteorology is not expected to act strongly distinct for different substances.

Comparison of the chemical composition of PM_{10} at the six temporary sites in the city of Basel

Figure 8-3 represents plots of the 41 measurements which were simultaneously performed at the urban site 'C' and at one of the temporary monitoring sites, situated within the city of Basel. For PM_{10}, S, Cl, Ti, and K the correlation coefficients were very high (> 0.95), indicating uniform distribution throughout Basel. For Br, Cr, Fe, and Pb the correlation coefficients were lower (< 0.81), indicating more spatial variability. The correlation coefficient of Cl is highly influenced by one data point. Without this influencing measurement correlation would be 0.91.

Systematic differences between the six temporary monitoring sites (represented by different symbols in Figure 8-3) were only observed for Pb. At the temporary sites '5' and '6' with high passenger car traffic the Pb concentration was always above the 1:1 line, indicating higher values than the urban background. This systematic pattern was not observed at the other four sites. For other elements the samples were too small and too strongly influenced by different meteorological conditions to detect systematic differences across the six sites.

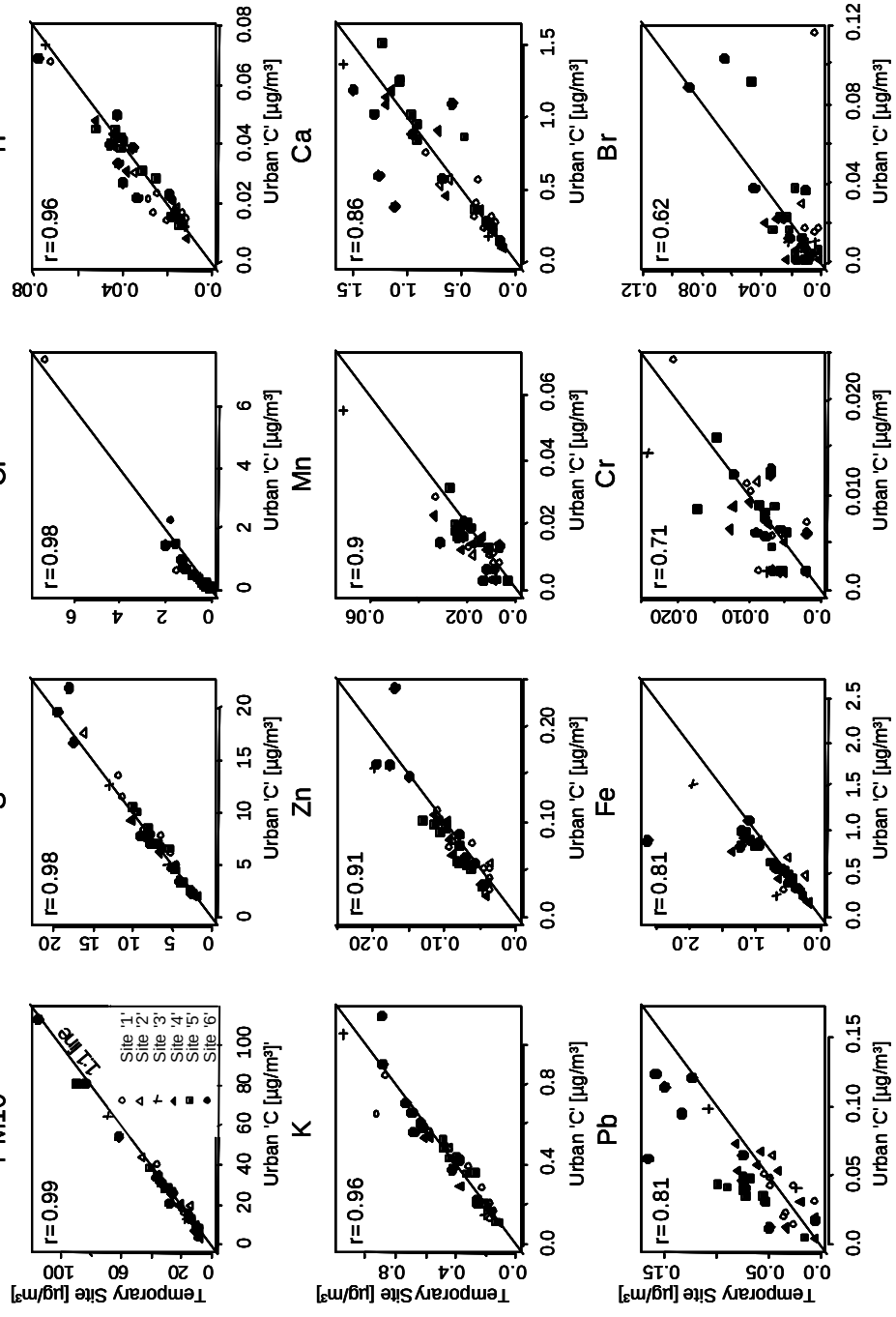

Figure 8-3. Pearson correlation of elemental concentration measured simultaneously at the fixed monitoring site 'C' and one of the six temporary monitoring stations (41 data points). The line represents the 1:1 line.

The higher Pb concentration at site '5' and '6' is likely to be caused by the high passenger car traffic at these sites. Leaded gasoline was allowed until 1 January 2000, in Europe. Although lead emissions from passenger cars have decreased in the last years due to an increased use of unleaded gasoline, passenger cars were still the most important emission source of particulate lead in urban environments in Switzerland in 1997 (Chiaradia and Cupelin, 2000). Moreover, the correlation between the concurrent Pb and Br measurements was substantial higher ($r>0.60$) at the sites '4' to '6' with high passenger car frequencies compared to the other three sites ($r<0.46$). Like Pb, Br is considered to be a tracer for passenger car emissions (Williams et al., 1998).

Chemical composition of PM_{10} at two urban and two rural sites

Chemical composition of PM_{10} at urban and rural sites was investigated based on data measured from April 1998 to May 1999. During this period PM_{10} measurements in Basel were generally lower than in 1997 due to different meteorological conditions. Throughout 1997, the annual mean mass concentration at the urban site 'C' was 27.9 µg/m^3, whereas the annual mean value from April 1998 to March 1999 was only 22.4 µg/m^3.

Annual mean concentrations of all compounds analyzed on the PM_{10} filters were always highest at the urban site 'D' and lowest at the rural site 'A' (Table 8-4). The largest coefficients of variation (cv) between the four sites were found for the traffic-related substances lead, OM, EC, and chloride (road salt). Lead was found to be almost 9 times higher at the urban site 'D' than at rural site 'A'. Chloride was 6 times higher, and OM and EC approximately 4 times higher. The relative proportions of these compounds were therefore higher at the urban sites than at the rural sites, whereas the relative proportions of sulfate, ammonium and cadmium were higher at the rural sites than in the city. The proportion of nitrate was around 11% at all four sites.

The most homogenous distribution of sulfate confirms the hypothesis that in Switzerland this substance originates mainly from long-range transport (Eliassen, 1978), as the levels of SO_2 in Switzerland are rather low (mean SO_2 concentrations from April 1998 to March 1999 at sites 'A', 'B', and 'C' were 4.0, 6.0, and 7.4 µg m^{-3}, respectively). Long-range transport may also be responsible for the uniform distribution of cadmium since the Cd levels are low and important emission sources in the Basel area are not known (Gälli Purghart B.C., 1990). The uniform distribution of Ammonium is probably not a result of long-range transport but is due to the fact that ammonium is a secondary pollutant produced from ammonia which originates

from agriculture as well as from vehicles with catalysts and industrial sources (Sutton et al., 2000). Therefore, the levels of the precursor gas ammonia may not differ widely between urban and rural sites.

Table 8-4: Annual mean chemical composition of PM_{10} at two rural and two urban sites between 1 April 1998 and 31 March 1999 as well as coefficient of variation (cv=standard deviation/mean value) and ratios between the concentrations at sites D and A. Percentage proportions are shown in brackets.

		Rural 'A'	Rural 'B'	Urban 'C'	Urban 'D'	cv	Ratio D/A
PM_{10}	[µg m^{-3}]	10.8	14.4	21.1	29.6	0.44	2.7
OM	[µg m^{-3}] ([%])	1.8 (17.0)	2.5 (17.6)	5.0 (23.6)	7.7 (26.1)	0.63	4.2
EC	[µg m^{-3}] ([%])	1.4 (12.9)	1.6 (10.8)	3.0 (14.1)	5.4 (18.3)	0.66	3.9
SO_4^{2-}	[µg m^{-3}] ([%])	2.2 (20.0)	2.4 (16.7)	3.0 (14.4)	3.2 (10.9)	0.19	1.5
NO_3^-	[µg m^{-3}] ([%])	1.1 (10.6)	1.6 (11.0)	2.5 (11.6)	3.1 (10.4)	0.42	2.7
NH_4^+	[µg m^{-3}] ([%])	0.8 (7.4)	0.9 (6.2)	1.2 (5.9)	1.3 (4.5)	0.24	1.7
Cl^-	[ng m^{-3}] ([%])	32 (0.30)	45 (0.31)	68 (0.32)	194 (0.65)	0.88	6.0
Zn	[ng m^{-3}] ([%])	20 (0.19)	24 (0.17)	45 (0.22)	67 (0.23)	0.55	3.3
Pb	[ng m^{-3}] ([%])	7 (0.07)	9 (0.06)	25 (0.12)	61 (0.21)	0.98	8.5
Cd	[ng m^{-3}] ([%])	0.3 (0.003)	0.3 (0.002)	0.4 (0.002)	0.5 (0.002)	0.26	1.6

Nitrate showed the highest spatial variability of the inorganic compounds of secondary origin. This is most likely a result of the marked differences in the concentration of the precursor substances NO_2 between the four sites. The mean NO_2 concentrations between April 1998 to March 1999 were 8.1, 10.7, 34.3, 57.6 µg m^{-3} at the sites 'A', 'B', 'C', and 'D', respectively. The large difference of the NO_2 concentration between urban site 'C' (background) and site 'D' (street canyon) results in a relatively large difference in the nitrate concentration (+ 26%), though the two sites were only 1 km apart. It may therefore be concluded that small-scale spatial differences in the nitrate concentration may occur in an urban environment if the NO_2 gradient is large.

Mean OM concentration was considerably higher at urban site 'D' than at urban site 'C' (+54%). This is due to the influence of road traffic. OM is of primary as well as of secondary

origin. Therefore, the OM levels may be the result of primary particle emissions or secondary particle formation from precursor compounds. During 1997, the mean volatile organic compounds (VOC) concentration was more than three times higher at site 'D' (92 µg m^{-3}) than site 'C' (28 µg m^{-3}). However, Castro, 1999 and Schauer, 1996 have shown that in urban environments OM is mostly of primary origin.

The large difference in the mean PM_{10} concentration between the urban sites 'C' and 'D' was mainly caused by traffic-related compounds. However, it must be emphasized that such a large difference was rather unusual compared to the other measurements within the city of Basel. This difference must be seen as a result of the special situation of the site 'D' which was located in a street canyon near to a traffic light. Analyses of the spatial variability of different fractions of particulate matter at the six temporary monitoring sites have shown that PM_{10} is in general uniformly distributed within Basel (± 10%) despite a broad range of traffic density at these six sites (Röösli et al., 2000a).

Seasonal differences between urban and rural sites

From November to March, monthly mean PM_{10} concentrations were consistently more elevated at the urban sites than at the rural sites (Figure 8-4). This was most clearly noticeable for nitrate and ammonium. The values of OM were also elevated in the city during the cold months. For sulfate, the difference between urban and rural sites was found to be slightly higher during the cold months. No seasonal pattern was observed for EC and lead. Chloride concentration was markedly elevated at urban site 'D' during December and January.

Previous analyses (Röösli et al., 2000a) have shown that large differences in the PM_{10} concentration between the urban and the more elevated rural sites in the Basel area occurred mainly during days with persistent surface inversions. The presence of an inversion layer reduces the air exchange. In addition it is associated with lower temperatures in the city than at the rural sites influencing the sampling efficiency of volatile substances such as nitrate and ammonium.

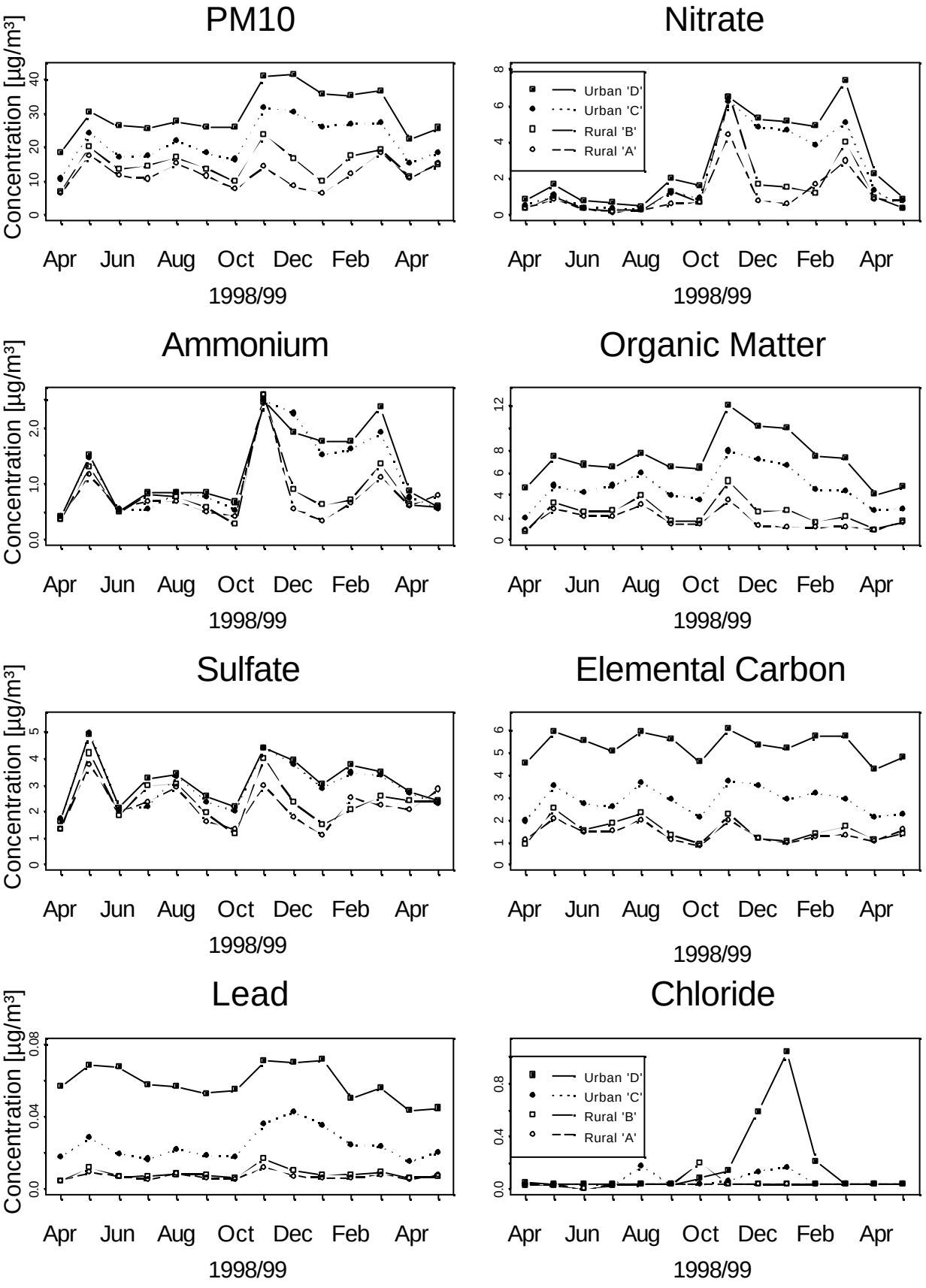

Figure 8-4: Monthly mean values of certain substances at two urban and two rural sites between April 1998 and May 1999.

Bonfire and national holiday

Figure 8-5 shows the effect on PM_{10} of a large bonfire in the night from 31 July to 1 August as well as additional privately burning of firework during the national holiday (1 August). The concentration of bonfire-related substances such as Ti (Figure 8-5), S, K, and Cl was considerably elevated, however, the increase in total PM_{10} concentration was relatively small. This figure illustrates that the total PM_{10} mass concentration is only moderately influenced by one emission source due to the background PM_{10}. An analogous situation may be found at busy streets: only a relative small increase of the total average PM_{10} concentration can be observed, but some specific traffic-related compounds such as EC, OM and Pb may increase much more.

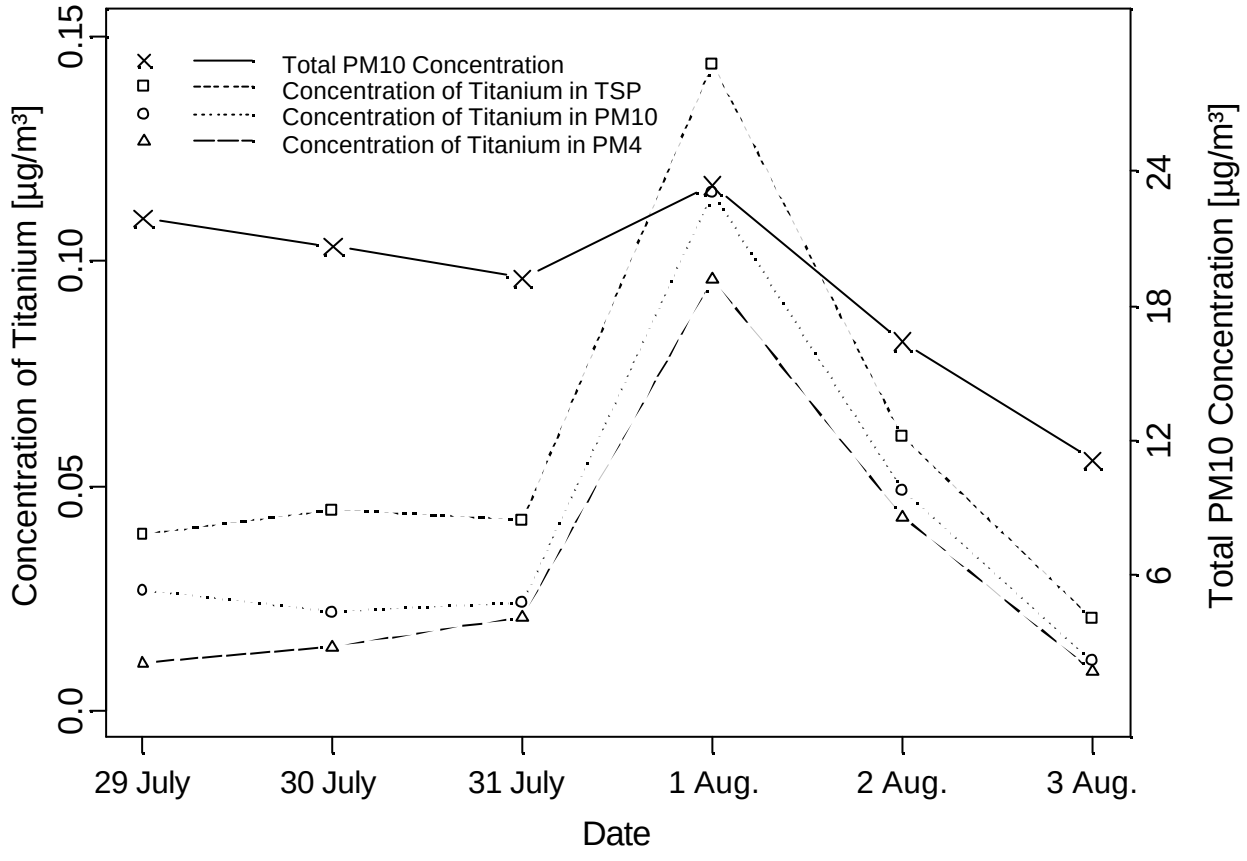

Figure 8-5: Illustration of the influence of a bonfire in the night from 31 July to 1 August on the concentration of titanium in the three size fractions as well as on PM_{10}, measured from 29 July to 3 August 1997 at site '2'.

Summary and conclusions

The chemical analyses of PM_{10} within the city of Basel and at two rural sites have shown that carbonaceous substances (EC, OM) and inorganic substances of secondary origin such as sulfate, nitrate and ammonium are the predominant components of PM_{10} in the Basel area (approximately 60-70%). In the urban environment the proportion of the carbonaceous compounds was higher than at the rural sites, whereas at the rural sites the proportion of sulfate and ammonium was higher than in the city.

Differences in the chemical composition between urban and rural sites were more pronounced during November-March than during the warm months. This may be explained by the fact that on certain days during winter, there was an inversion layer between the city and the more elevated rural sites resulting in reduced air exchange. In addition, the lower temperature in the city increased the sampling efficiency of volatile substances such as nitrate and ammonium.

With the exception of one site in a street canyon next to a traffic light, mean mass PM_{10} concentration was uniformly distributed within Basel (± 10%). However, the chemical composition was more variable due to specific sources like road traffic and other anthropogenic emissions. Due to the large contribution of the background concentration spatial variability of PM_{10} mass concentration was not markedly affected by these elements.

Highest ratios between mean concentration during workdays and weekends were found for mineralogical elements (Ca, Ti, Mn, Fe) and EC. This finding indicates that these compounds were for the most part caused by regional human activities such as resuspension from road traffic or construction work. In Basel wind erosion seems to be of minor importance for the mineralogical elements as the concentration of these elements in all three size fractions was negatively correlated with daily mean and maximum wind velocity.

Largest difference within the urban environment of Basel as well as between urban and rural sites was found for primarily traffic-related compounds. Our analyses indicate that these traffic-related compounds consist not only of exhaust particles (EC, OM, Pb), but also of resuspended compounds (e.g. Ca, Cl). The concentration of nitrate was also clearly associated with traffic density as its precursor substance NO_2 is a tracer for traffic emissions. However, the spatial variation of this compound of secondary origin was lower than the spatial variation of the primarily traffic related compounds.

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PART V RISK ANALYSIS

Chapter 9

Which Effect Measure Should Be Used for Impact Assessment in a New Population Context?

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Abstract

One characteristic feature in impact assessment is the transfer of an exposure-response association into a new population context. The consequences of basing this estimation procedure either on relative effect measures (e.g. relative risks) or on measures on the difference scale (e.g. excess rate) were exemplary illustrated with two air pollution studies which quantified the association between lung cancer and PM_{10} in cohorts with varying lung cancer incidence due to different smoking habits. This example showed that the type of measure chosen may markedly influence the result of an impact assessment, if the frequency of the disease of interest varies across different populations due to other risk factors. Thus, it was concluded that whenever studies from different population contexts are pooled, careful considerations about the appropriate type of measure are required. In environmental health risk assessment with usually small risks and unspecific (i.e. multicausal) health endpoints, an excess rate-based approach may provide a more reliable and stable estimate than a relative risk based approach in many circumstances. Methodological details of an excess rate-based approach are presented in the paper.

Introduction

Quantitative risk assessment plays a major role in the setting of environmental standards or exposure limits. Environmental epidemiology often provides important input for such assessments, particularly in the low range of general population exposure to complex mixtures (Wahrendorf, 1994, Samet, 2000, Smith, 1988, Hertz-Picciotto, 1995b). The availability of an exposure-response function is crucial to conduct an impact assessment. Most often an exposure-response function has to be generalized or transferred to the population of interest. This may be either a spatial or temporal generalization or both.

Generally, in quantitative risk assessment studies relying on epidemiologic evidence the exposure-response association is based on relative measures such as odds ratios or relative risks - often used as synonyms in the low range of risks (Aunan et al., 1998, Levy et al., 2000, Künzli et al., 2000, Zmirou et al., 1999, Ostro, 1993, Schwartz, 1994a, Lipfert and Wyzga, 1995, Lipsett M., 1999, Bhatia R., 1997). Relative measures are most commonly used to express effect estimates. In particular relative odds have some interesting mathematical properties, and are therefore suited to measure associations in case-control studies. In many

instances it may be appropriate to transfer relative measures of risk to a new population context (Engels et al., 2000). However, relative measures of effect depend on the magnitude of the baseline occurrence (Dupont and Plummer, 1996, Rothman, 1986). Thus, different relative effects in two populations can correspond to the same number of cases, if the baseline occurrence of the health endpoint in question varies due to differences in the profiles of other risk factors than the one considered (Peacock, 1971).

For several air pollution related health endpoints, smoking is a primary risk factor. Therefore, the incidence of a health endpoint such as, e.g., lung cancer varies widely between populations depending on the respective smoking habits. Thus, assuming no interaction between smoking and air pollution the number of lung cancer cases attributable to air pollution might be the same in two populations with largely different smoking habits but equal levels of air pollution. Yet, the observed air pollution related relative risks may be much larger in a non-smoking population. Conversely, the same relative risk may imply largely different numbers of attributable cases if the smoking habits differ.

A popular measure to quantify the impact of an exposure is the calculation of attributable cases. The calculation of the number of attributable cases is generally based on a relative risk estimate, derived from epidemiological studies, and the incidence/prevalence in the target population (Krzyzanowski, 1997). Thus, the estimated attributable number of cases is sensitive to the disease frequency in the target population.

In this paper, an alternative approach to estimate the number of attributable cases is presented, which is not affected by the disease frequency in the target population. This is achieved by directly calculating excess rates from the study specific incidences. The performance of this alternative approach was evaluated in an assessment of the impact of air pollution on lung cancer based on the Six-Cities study (Dockery et al., 1993) and the Seventh Day Adventists study (Abbey et al., 1999) which quantified the association in two populations with different smoking habits. These methodological considerations were part of a quantitative health risk assessment for the population living in the Basel area (Röösli et al., submitted (a)).

Methods

Both, the Six-Cities study and the Seventh Day Adventists study (AHSMOG) reported the association between lung cancer mortality and long-term mean PM_{10} concentration. These cohort studies provided relative risk estimates (Table 9-1). We used the results of the two

studies do derive the number of air pollution attributed cases in two different ways. The first approach applies a relative risk estimate derived from the original studies to the incidence in the target population. The second approach generalizes the excess rates derived from the original studies to the target population. This approach is independent of the incidence in the target population. In order to illustrate the consequence of using different types of measure in impact assessments, both approaches were applied to an external population, namely the population living in the Basel area, to estimate the number of lung cancer cases attributable to air pollution.

Table 9-1: Characteristics of the three American cohort studies which quantified the association between particulate matter concentration and lung cancer incidence.

	Lung cancer incidence (per 100,000 person years)	Proportion of current (former) smokers	RR[2] (95%-CI)	Air pollution range[3]
Six-cities	106	36% (24%)	1.37 (0.81; 2.31)	PM_{10}: 28.3 µg/m³
Adventists[1]	42	0% (25%)	2.35 (1.09; 5.08)	PM_{10}: 24.1 µg/m³

[1] Abbey et al. performed two separate analyses for males and females which were then averaged.

[2] Relative risk originally reported in the study.

[3] Reference item for the original RR.

For both methods, the impact assessment is based on the following assumptions:

i.) In the Basel area, 70% of the population are older than 30 years (Bundesamt für Statistik, 1997). (The exposure response functions of the two epidemiological studies corresponds only to adults older than 30 years.)

ii.) The population weighted average PM_{10} exposure is 25 µg/m³ (Röösli et al., submitted (a)). As in other impact assessment studies (Künzli et al., 2000) a PM_{10} reference concentration of 7.5 µg/m³ was used. Health effects below this level were not quantified.

iii.) The baseline lung cancer incidence (I_0) is 50 cases per 100,000 person years. This was the observed lung cancer incidence in the Basel area between 1989 and 1997 (Torhorst, 1999).

The first step in the relative risk based approach is the calculation of the relative risk (RR_{10}) per 10 µg/m³ PM_{10} in each study. This was done by using formula 9-1.

$$RR_{10} = \exp\left(\frac{\ln(RR_{tot})}{\Delta C} \cdot 10 mg/m^3\right) \quad \textit{(9-1)}$$

where RR_{tot} is the relative risk corresponding to a PM_{10} difference (ΔC) in the considered study.

Thus, the number of lung cancer cases per year (N_{RR}) was calculated by using equation 9-2

$$N_{RR} = \frac{RR_{10} - 1}{10 mg/m^3} \cdot I_{tar} \cdot P \cdot C_{av} \quad \textit{(9-2)}$$

where, I_{tar} is the observed lung cancer incidence in the target population (50 cases per year), P is the proportion of persons older than 30 years (70%) and C_{av} is the PM_{10} concentration above background in the target area (25 µg/m³-7.5 µg/m³ =17.5 µg/m³). The denominator (10 µg/m³) indicates that RR_{10} refers to a 10 µg/m³ increment in PM_{10}.

Alternatively, the first step in the second method to estimate the number of air pollution attributed cases consists in calculating the excess rate per 10 µg/m³ PM_{10}. In the Six Cities and the AHSMOG-study, as in most other studies, the data are not analyzed on an additive scale. Thus, the excess rate has to be estimated from the given data in the studies.

Generally the excess rate (ER_{10}) can be calculated from the relative risk (RR_{10}) and the baseline incidence in the study population ($I_{0(stud)}$) (Rothman, 1986):

$$ER_{10} = I_{0(stud)} \cdot (RR_{10} - 1) \quad \textit{(9-3)}$$

However, the baseline incidence of lung cancer in the study population, given zero exposure to air pollution, is not known. Therefore, one may use the observed incidence of lung cancer, leading, however, to an overestimation of the excess rate. To prevent unwarranted overestimation, we used a more generalizable method to estimate ER_{10} (equation 9-4). This method is based on the actually observed lung cancer incidence in each study ($I_{av(stud)}$) and the relative risk. The derivation is presented in the appendix.

$$ER_{10} = I_{av(stud)} \cdot \ln(RR_{10}). \quad \textit{(9-4)}$$

In our example equation 9-3 and 9-4 provide similar results, as small risks imply: $I_{av} \tilde{} I_0$ and ln(RR) ˜RR-1. However, simulations showed that whenever the relationship between

incidence and exposure is linear and data are fitted by a Poisson regression model, the true excess rate ER is more accurately estimated by equation 9-4 for small relative risks.

By means of the second approach, based on excess rates, the number of lung cancer cases per year (N_{ER}) was calculated by using formula 9-5

$$N_{ER} = \frac{ER_{10}}{10 mg/m^3} \cdot P \cdot C_{av} \qquad \textit{(9-5)}$$

where ER_{10} is the excess rate per 10 µg/m³ PM_{10} and 100,000 person years, P is the proportion of person older than 30 years and C_{av} is the PM_{10} concentration in the Basel area. Unlike the first approach (equation 9-2), the second approach does not involve the lung cancer incidence of the target population (I_{tar}).

Results

The estimated relative risks and excess rates per 10 µg/m³ increment in PM_{10} for the Six-Cities study and the Adventists study as well as the estimated number lung cancer cases attributable to air pollution in the Basel area are summarized in Table 9-2. The estimated number of lung cancer cases due to air pollution in Basel varied to a large extent according to the type of measure used and the reference study selected.

Table 9-2: Estimated relative risks (RR) and excess rates (ER) per 100,000 person years and per 10 µg/m³ increase in the average level of PM_{10} as well as estimated number of lung cancer cases due to air pollution in the Basel area.

	Six-Cities Study	Adventists Study
Relative Risk per 10 µg/m³ in PM_{10} (95%-CI)	1.12 (95%-CI: 0.93; 1.34)	1.42 (95%-CI: 1.03; 1.96)
Excess Risk per 10 µg/m³ in PM_{10} (95%-CI)	11.8 (95%-CI: -7.9; 31.3)	16.7 (95%-CI: 3.4; 30.2)
N_{RR}: Number of annual cases (derived from the relative risk)	7.4 (95%-CI: -4.3; 20.8)	25.7 (95%-CI: 1.8; 58.8)
N_{ER}: Number of annual cases (derived from the excess risk)	14.5 (95%-CI: -9.7; 38.3)	20.5 (95%-CI: 4.2; 37.0)

When applying the relative risk based approach, the lowest estimate was obtained from the Six-Cities study (7.4 cases per 100,000 person years). A 3.5 times higher estimate was derived from the results of the Adventist study (25.7 cases). In contrast, the excess rate based estimates from both studies were rather similar. They differed only by a factor of 1.4 (14.5 and 20.5 cases per year, respectively). The choice of measure also strongly influenced the confidence intervals. When considering relative risk based estimates, the confidence interval of the Adventists study was broader than the one of the Six-Cities study. However, the reverse picture was found when estimates were derived from excess rates.

Discussion

The example clearly showed that the type of measure used to derive risk attribution can considerably influence the results of the impact assessment. The lung cancer incidence in the non-smoker cohort of the Adventist study was considerably lower than in the cohort of the Six-Cities study, including both, smokers and non-smokers. Thus, the relative risks were quite different, although the absolute number of lung cancer cases due to air pollution, (i.e. the excess rate), was fairly similar in the two study.

The reason for the observed inconsistent estimates relies on the fact that relative risks depend on the incidence of a disease. Thus, applying a relative risk estimate for lung cancer derived from a non-smoker cohort to a population containing smokers results in an improper estimate of the number of cases attributed to air pollution. In general, environmental health risk assessment deals with small risks and unspecific (i.e. multicausal) health endpoints making estimates susceptible for the kind of bias demonstrated in this example. For instance, it was observed among 29 European cities that the estimated percent increase in daily mortality associated with a 10 $\mu g/m^3$ increment in PM_{10} was correlated with the age standardized mortality rates of the respective cities (Katsouyanni et al., 2001). In cities with high mortality rates the relative increase was generally lower. This supports the hypothesis that risk measures on the difference scale (e.g. excess rates) are more homogeneous than relative measures among populations with different risk profiles. Thus, to transfer relative risks from U.S. and European studies to developing countries may be even more problematic, since the incidence of specific diseases may be considerably different in such countries due to differences in the distribution of other influential risk factors.

It can be concluded that whenever the baseline occurrence of the health endpoint in question differs across study populations, the result of pooling relative risks from the respective studies

may be misleading. If so, in many circumstances pooling excess rates may be more reliable than pooling relative risks.

However, in other cases, the use of absolute instead of relative measures in meta-analysis can be counterintuitive. For instance, applying an absolute measure to a different age group than the one considered in the original study may lead to unrealistic results. Thus, estimating the lung cancer risk from air pollution for a population younger than 30 years based on the above described excess rate among adults older than 30 would result in more lung cancer cases attributed to air pollution than actually occurred in the younger age group.

To make best use of epidemiologic results in impact assessment studies, it would be desirable to have epidemiologic studies that provide both, absolute and relative risk estimates. The decision on whether a multiplicative or an additive model is more appropriate depends on the nature of the association (Lund, 1995, Jarvholm, 1997, Wilson, 2000, Stronegger et al., 1998). The results are usually not much different for small risk factors, but additive models of risk might provide more accurate estimates of excess rates in many circumstances.

We have demonstrated the large influence of strong co-risk factors if epidemiologic risk estimates have to be transferred to other populations with different distributions of these co-factors. We do not claim that the higher consistency of the results obtained by the excess rate based method is fully explained by the differences in baseline incidences across the studies considered. Other sources of bias inherent to the original studies may have contributed to the discrepancies in the relative risks. However, our example emphasizes the problem of generalizing concentration-response functions across populations with large differences in other influential risk factors such as smoking. For health outcomes with similar incidences across different populations the issue may be of minor relevance. The same is true when transferring estimates of associations between very strong risk factors and rather specific outcomes, such as for smoking and lung cancer.

In conclusion it is not possible to define one single correct approach to transfer effect measures into a new population context. In each application, one must carefully consider the choice of effect measure and be aware of the inherent model. As the scientific base for such methodological considerations is often very small, plausibility considerations and sensitivity analyses may clarify the appropriateness of the approach used. Whenever the occurrence of a disease varies across different populations due to other factors than the ones primarily considered, the excess rate-based approach might provide a more reliable and stable estimate of attributable risks than a relative risk based approach.

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Appendix

In the following the derivation of formula 9-4 is presented. Using the excess rate (ER), the cancer incidence in the exposure group *i* (I_i) can be expressed as follows:

$$I_i = I_{av} + ER \cdot (C_i - C_{av}), \qquad \textit{(9-6)}$$

where I_{av} = overall cancer incidence, C_{av} = PM_{10} exposure at which the incidence would equal I_{av}, and C_i = exposure in category i.

On the other hand, using the relative risk, the cancer incidence in category *i* (I_i) is obtained according to equation 9-7:

$$I_i = I_{av} \cdot e^{(C_i - C_{av}) \cdot \ln(RR)}, \qquad \textit{(9-7)}$$

where RR is the relative risk associated with an increase in average PM_{10}-exposure. Dividing formula 9-6 and 9-7 by I_{av} and taking logarithms leads to the following equations:

$$\ln\left(\frac{I_i}{I_{av}}\right) = \ln\left(1 + ER \cdot \frac{C_i - C_{av}}{I_{av}}\right) \qquad \textit{(9-8)}$$

$$\ln\left(\frac{I_i}{I_{av}}\right) = (C_i - C_{av}) \cdot \ln(RR) \qquad \textit{(9-9)}$$

If the expression $ER \cdot (C_i - C_{av})/I_{av}$ is small, then $\ln(I_i/I_{av})$ in equation 9-8 may be approximated as follows (first order approximation):

$$\ln\left(\frac{I_i}{I_{av}}\right) \approx \frac{ER \cdot (C_i - C_{av})}{I_{av}} \qquad \textit{(9-10)}$$

Combining equation 9-9 and 9-10, one obtains for small relative risks

$$\frac{ER \cdot (C_i - C_{av})}{I_{av}} \approx (C_i - C_{av}) \cdot \ln(RR) \qquad \textbf{(9-11)}$$

Hence, the excess rate (ER) can be estimated in each study from the relative risk (RR) and the observed average lung cancer incidence (I_{av}) as follows:

$$ER \approx I_{av} \cdot \ln(RR). \qquad \textbf{(9-12)}$$

Chapter 10

Assessment of Cancer Risk Attributable to Air Pollution Using an Epidemiology Based Method

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Summary

Background: The extent of cancer risk attributable to ambient air pollution generally has been assessed based on unit risk factors. The quantitative evaluation of unit risk factors has put great emphasis on data from animal experiments. However, the extrapolation of these data to the general human population is a known source of large uncertainty. Moreover, the unit risk-based approach does not take into account synergistic effects of various carcinogens in the mixture of ambient air pollution.

Methods: The authors compared an unit risk-based cancer risk assessment with an epidemiology-based method, using PM_{10} as a surrogate of the total air pollution. The excess rate for lung cancer cases attributable to an increase of 10 µg/m³ in average PM_{10}-exposure was estimated from epidemiological cohort studies with a meta-analytic approach.

Results: Applying the epidemiology-based risk method to the Basel (Switzerland) population resulted in 6.7 (95%-CI: -0.8 to 14.2) expected cancer cases per 100,000 person years. This estimate was considerably higher than an unit risk-based estimate yielding 0.8 (range: 0.21-11.4) cancer cases per 100,000 person years.

Conclusions: The different results of the both approaches might be due to either an overestimation of the epidemiology-based method, or an underestimation of the unit risk-based approach, or simply by chance. Explanations supporting either of these hypotheses are discussed in the paper.

Introduction

Although outdoor air has been shown to be contaminated by carcinogens, the extent of cancer attributable to ambient air pollution has been subject to persistent controversy in the last decades (Cohen, 2000). As the general population is exposed to ambient air pollution, the contribution of outdoor air pollution to cancer, especially lung cancer, is of public health concern. Therefore, risk assessment is needed for guiding regulatory decisions.

Cancer risk attributable to air pollution can be assessed by combining the distribution of exposure in the population of interest with a potency factor that describes the increase in risk per unit increase in exposure (Hertz-Picciotto, 1995a). In the last years the use of 'unit risks' as potency factors has been established in cancer risk assessment studies (Kappos and Schmitt, 1993, Törnqvist and Ehrenberg, 1994, Hemminki and Pershagen, 1994, French et al., 1997,

Yetergil, 1998, Woodruff et al., 1998, Morello-Frosch et al., 2000). The unit risk factor describes the cancer risk associated with lifelong exposure to 1 µg/m³ of the substance of interest, assuming a linear dose-response function without a threshold value (WHO, 1987). Unit risk factors for a variety of substances were evaluated and published by the IARC (International Agency for Research on Cancer), the U.S.EPA (United States Environmental Protection Agency), the OEEHA (Office of Environmental Health Hazard Assessment, California) and the LAI (Länderausschuss für Immissionsschutz, Nordrhein-Westfalen, Germany) (OEEHA, 1999, U.S.EPA, 1999b, WHO, 2001, LAI, 1992). The quantitative evaluation of the unit risk factors has put strong emphasis on animal experimental data, even though the extrapolation of animal bioassay data to the general population is known to be afflicted with large uncertainties. Risk estimates from varying extrapolation models can differ by five orders of magnitude in the low exposure range for the same data (Maynard et al., 1997). The magnitude of uncertainty in risk assessment is therefore likely to be greater when using animal data as compared to epidemiologic study results (Hertz-Picciotto, 1995b, Smith, 1988, Smith, 1995).

A further problem of unit risk-based risk assessment is that it quantifies cancer attributable to air pollution by the sum of the risks of each single carcinogen. However, it cannot be verified whether all relevant carcinogens are included in a study; moreover, potential interactions between pollutants in the outdoor air mixture are not taken into account. There is substantial evidence suggesting that environmental carcinogens interact synergistically in causing cancer. Synergistic effects are well documented for smoking combined with asbestos Vainio and Boffetta, 1994, Erren et al., 1999, Albin et al., 1999), radon (Hornung et al., 1995, Hornung et al., 1998, Thomas et al., 1994), and alcohol (Rothman and Keller, 1972, Choi and Kahyo, 1991). However, different compounds may also interact antagonistically.

It is widely recognized that knowing the carcinogenic potency of one single compound is more of an academic than a practical importance (Möller et al., 1994); yet combined effects in complex mixtures of ambient air pollution have hardly been investigated. An assessment of potential health risks, associated with exposure to complex mixtures, requires more than an understanding and quantification of the effects of individual compounds contained in the mixture (Berenbaum, 1984, Wahrendorf, 1994). IARC describes this problem as follows: "Estimating the human cancer risks of exposure to complex mixtures presents formidable methodological problems. However, such exposures are thought to account for a large

proportion of cancers, in particular because of widespread exposure to such mixtures within populations" (Vainio et al., 1990).

Using a proxy measure of total carcinogenic ambient air pollution instead of summing up single carcinogenic agent may be a way of implicitly including combined effects in the risk quantification. Some studies have recently quantified the health risk of air pollution taking PM_{10} as an surrogate for all ambient air pollutants (Künzli et al., 2000, Aunan et al., 1998, Zmirou et al., 1999). However, none of these studies focused on cancer risk from air pollution.

The excess of lung cancer incidence attributable to a 10 µg/m³ increase in the average PM_{10}-level was estimated from published epidemiologic cohort studies. Based on these estimates, the annual number of lung cancer cases in the Cantons of Basel-Stadt and Basel-Landschaft (Switzerland) attributable to air pollution were calculated, taking into account the PM_{10}-exposure distribution in this population. We then compared the results of this epidemiology-based method with estimates for the same population based on unit risk factors.

METHODS

Epidemiology-based risk assessment method

Exposure-response association. All so far published cohort studies, having quantified the association between lung cancer mortality and particulate matter, were taken into account to derive an exposure-response estimate. These were the American Cancer Society Study (Pope et al., 1995, the Six Cities Study (Dockery et al., 1993), and the Adventists Studies (Abbey et al., 1993) (Table 10-1).

For each study, we calculated the excess number of lung cancer cases per 100,000 person years (excess rate) per 10 µg/m³ increase of the PM_{10} concentration from the reported relative risk and the cancer incidence. The lung cancer incidence was assumed to be equal to the lung cancer mortality. A pooled value for the excess rate of all three studies was obtained using an inverse variance weighting (Petitti, 1994a, DerSimonian and Laird, 1986). The three study results were tested for homogeneity using Cochran's Q-statistic (DerSimonian and Laird, 1986). As the between-study variance was not significant ($p=0.11$), we did not adjust for potential random study effects. The detailed calculation procedure is described elsewhere (Röösli et al., submitted (b)).

Table 10-1: Characteristics of the three American cohort studies which assessed the association between PM and lung cancer.

	Lung cancer incidence (per 100,000 person years)	Age range of study population	Prop. of current smokers [%]	Prop. of former smokers [%]	RR[‡] per 10 µg/m³ PM_{10}	95%CI[§]
Six-cities	106	25-74	36%	24%	1.12	0.93 to 1.34
Cancer Society	106[†]	>30	22%	29%	1.01	0.94 to 1.09
Adventists, males[*]	61	27-95	0%	36%	1.65	1.21 to 2.27
Adventists, females	22	27-95	0%	14%	1.13	0.81 to 1.57

[*] Abbey et al. performed two separate analyses for males and females.
[†] Information about cancer incidence in this study population is not available. Thus the lung cancer incidence from the six cities-study was applied.
[‡] Relative risk
[§] Confidence interval

Exposure assessment. PM_{10} exposure of the 450,900 persons1997 living in the Basel area (Cantons of Basel-Stadt and Basel-Landschaft) was assessed by using a dispersion model (Heldstab et al., 1999), which had been developed for a tri-national study (Künzli et al., 2000). The model was based on emission data taking into account primary and secondary particulates and had been validated against measured PM_{10} concentrations. It calculates mean PM_{10}-concentration per kilometer squared. It is described in detail elsewhere (Filliger et al., 1999). The frequency distribution of the Basel population across various PM_{10}-exposure classes is shown in Figure 10-1. The population weighted average of PM_{10} was 24.9 µg/m³.

Calculation of the attributable number of cases. To quantify the number of lung cancer cases attributable to air pollution in the Basel area, we used a PM_{10} reference concentration of 7.5 µg/m³, thus, health effects below this level were not quantified. The same conservative assumption was used in the tri-national study (Künzli et al., 2000). This assumption was applied because PM_{10} concentrations below this reference level were neither measured in the epidemiologic studies from which the exposure-response association was derived, nor in the Basel study area. This level also includes the natural background PM_{10} concentration. Furthermore, this assumption allowed us to compare the result of this cancer risk assessment with the results of the health risk assessment of the tri-national study.

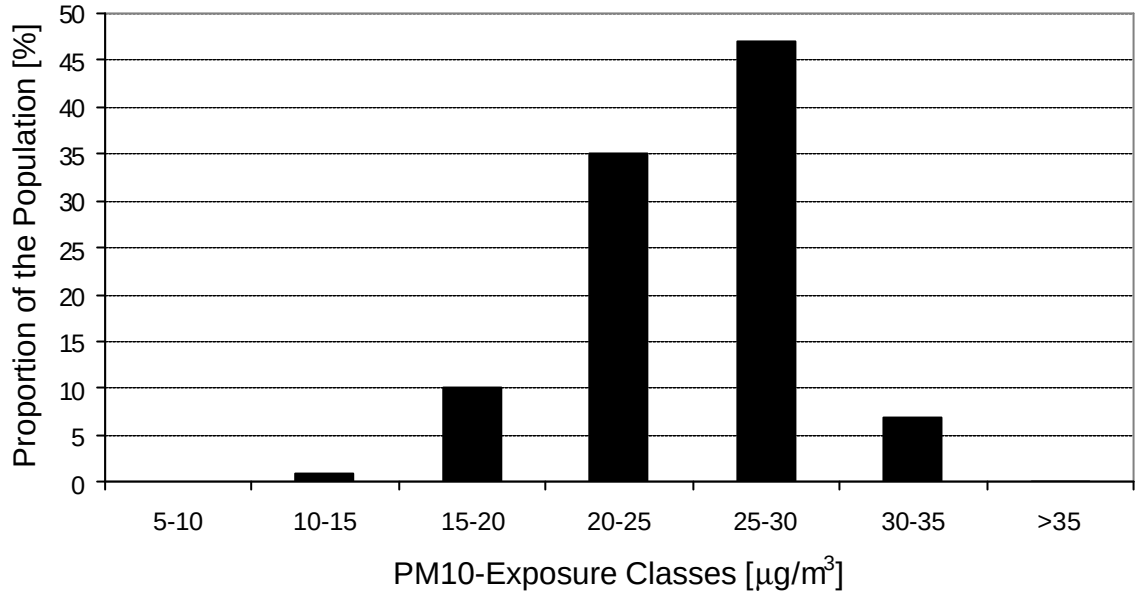

Figure 10-1: Distribution of average exposure to PM_{10} in the residential population of the Basel area.

The number of lung cancer cases attributable to air pollution (N_{ER}) derived by the epidemiology-based method was calculated with the following formula:

$$N_{ER} = 0.7 \cdot P \cdot ER \cdot \frac{(C_{av} - C_{ref})}{10} \qquad \textit{(10-1)}$$

where P is the population size, ER is the excess rate per 10 µg/m³ PM_{10} increase and (C_{av}-C_{ref}) is the average exposure of the population minus the reference concentration. The factor of 0.7 refers to the fact that the excess rate (ER) is applying to a population of adults older than 30 years analogous to the age distribution in the cohorts from which ER is derived. 70% of people living in the Basel area are older than 30 years.

Risk assessment method based on unit risk factors

Exposure-response association. Summary estimates of unit risks were obtained by calculating the geometrical mean of the published unit risks from IARC, U.S.EPA, OEEHA, and LAI (Table 10-2). In order to estimate minimum and maximum cancer risks, additional calculations were performed using the lowest and highest published unit risk factors in Table 10-2 for each substance.

Table 10-2: Unit risk estimates per 100,000 persons for 1 µg/m³ lifelong (70 y.) exposure to the corresponding pollutants.

	LAI (1993)	IARC (1996)	U.S. EPA (1998)	OEHHA (1997)	**geom. mean value**
arsene	400	150	430	330	**300**
benzene	0.9	0.57*	0.41*	2.9	**0.89**
1,3-butadiene	-	-	28	17	**22**
cadmium	1200	-	180	420	**450**
chromium (VI)	-	4000	1200	15000	**4200**
1,4-dichlorobenzene	-	-	-	1.1	**1.1**
diesel exhaust	7	3.4	-	30	**9**
lead	-	-	-	1.2	**1.2**
nickel	-	38	48	26	**36**
PAH (per µg/m³ BaP)†	7000	8700	-	110‡	**7800**
trichloromethane	-	-	2.3	0.53	**1.1**
trichloroethene	-	0.043	-	0.2	**0.093**
tetrachloroethene	-	-	-	0.59	**0.59**

* This value represents a geometric mean value of the advised range.

† Polycyclic hydrocarbons (unit risk of PAH is expressed per 1 µg/m³ concentration of benzo[a]pyrene)

‡ This value allows only for benzo[a]pyrene and is not taken into account for the mean value.

Exposure assessment. By means of the PM_{10} dispersion model, our study population was classified into categories of low, average and high exposure. Persons living in areas with an average PM_{10} concentration of less than 20 µg/m³ were assumed to be exposed to a generally low amount of pollution (50,000 of 451,000 inhabitants). Persons living in areas with a PM_{10} level between 20 and 25 µg/m³ were defined as exposed to an average amount of pollution (160,000 inhabitants), and persons living in areas with a PM_{10} concentration above this range were defined as generally highly exposed (240,000 inhabitants). Between 1997 and 1999 a series of carcinogenic substances were measured repeatedly at 11 different sites in the Basel area, representing the three exposure categories. Concentrations of nickel, lead, arsene and

cadmium were chemically determined from PM_{10} filters using energy-dispersive X-ray fluorescence spectrometry (Ni, Pb), inductively coupled plasma atomic emission spectroscopy (Cd), and atom absorption spectroscopy (As), respectivly. Various polycyclic aromatic compounds from PM_{10} filters (Cyclopenta[c,d]pyrene, benzo[b]naphto[2,1-d]thiophene, benzo[a]anthroacene, benzo[k,b]fluoranthene, benzo[a]pyrene, benzo[e]pyrene, perylene, benzo[ghi]perylene, dibenz[a,h]anthracene, indeno[1,2,3-cd]pyrene, anthranthene and coronene) as well as benzene, 1,4-dichlorobenzene, trichloromethane, trichloroethene and tetrachloroethene (sampled by passive samplers) were analyzed with gas chromatography and mass spectroscopy (GC/MS). 1,3-butadiene was sampled with a passive canister and analyzed by GC/MS. Diesel exhaust exposure was obtained by multiplying the elemental carbon concentration, quantified from PM_{10} filters using a thermographic method, with a factor of 1.8, reflecting the proportion of EC in the diesel exhaust (Pierson et al., 1983).

The details of the sampling procedure is described elsewhere (Röösli et al., 2001, Röösli et al., 1999). Based on these data, the average concentrations in the three exposure categories were estimated and the population weighted average of each carcinogen calculated (Table 10-3).

Calculation of the attributable number of cases. The unit risk-based method assumes a linear cumulative association between exposure to carcinogenic substances and the health outcome WHO, 1987, similar to the epidemiology-based risk assessment. With the unit risk-based method, the annual number of cases attributable to air pollution (N_{UR}) was obtained by multiplying the population size (P) with the unit risk factor (UR_i) and the average exposure level ($C_{av,i}$) of each pollutant of interest *i* (Equation 10-2). The total risk attributable to air pollution was then calculated by summing up the risks of all carcinogenic pollutants (see Table 10-3). In contrast to the epidemiology-based method, reference concentrations were assumed to be zero. This is the usual procedure for an unit risk-based risk assessmentWHO, 1987, because natural sources of these pollutants are negligible. This summary risk estimate includes all types of cancer.

$$N_{UR} = P \cdot \sum_{i=1}^{n} UR_i / 70 \cdot C_{av,i} \qquad \textbf{(10-2)}$$

Division by 70 yields the annual unit risk factor.

Table 10-3: Average concentrations of pollutants in the areas of low, average and high pollution.

	units	low	average pollution	high	**population weighted average**
PM_{10}	*µg/m³*	*<20*	*20-25*	*>25*	***24.9***
arsene	ng/m³	0.3	0.5	0.7	**0.6**
benzo[a]pyrene	ng/m³	0.04	0.3	0.5	**0.4**
benzene	µg/m³	1.5	2.1	2.7	**2.3**
1,3-butadiene	µg/m³	0.13	0.38	0.6	**0.5**
cadmium	ng/m³	0.3	0.4	0.5	**0.4**
chromium (VI)	ng/m³	0.6	0.9	1.3	**1.1**
1,4-dichlorobenzene	µg/m³	0.05	0.08	0.11	**0.09**
diesel exhaust	µg/m³	2.5	3.3	4.3	**3.8**
lead	ng/m³	8.8	23.5	38.3	**30**
nickel	ng/m³	4.2	6.6	8.9	**8**
PAH	ng/m³	0.7	3.2	5.7	**4.2**
trichloromethane	µg/m³	0.05	0.08	0.10	**0.09**
trichloroethene	µg/m³	0.2	0.28	0.39	**0.3**
tetrachloroethene	µg/m³	0.1	0.25	0.39	**0.3**

RESULTS

Epidemiology-based estimate

The weighted average of the study specific estimates of lung cancer excess rates across the three epidemiologic studies yielded 5.8 cases per 100,000 person years and per 10 µg/m³ increase in PM10 (95% confidence interval: -0.7, 12.3) (Table 10-4). As this pooled estimate is based on the same mathematical model as unit risk factors, this cumulative excess rate for an exposure to 1 µg/m³ PM_{10} from age 30 to age 70 may be interpreted as a lifelong unit risk

factor. The corresponding unit risk factor for PM_{10}, as a proxy of the lung cancer effects of the air pollution mixture, is $2.32 \cdot 10^{-4}$ (95% confidence interval: $-2.70 \cdot 10^{-5}$, $4.90 \cdot 10^{-4}$).

Table 10-4: Excess rates (ER) per 100,000 person years and per 10 µg/m³ increase in PM10 and 95 percent confidence interval (95%-CI).

	ER per 1 µg/m³ PM_{10}	95%-CI
Six-Cities	1.18	-0.79 to 3.13
Cancer Society	0.09	-0.70 to 0.89
Adventists	1.67	0.34 to 3.02
Pooled value	**0.58**	**-0.07 to 1.23**

Using the pooled excess rate and multiplying it with the PM_{10} exposure of the Basel population (Equation 10-1) resulted in 6.7 annual lung cancer cases per 100,000 inhabitants of the Basel area. The 95% confidence interval was -0.78 to 14.2 cases (Table 10-5). A minimum estimate, based on the American Cancer Society study, yielded 1.1 annual lung cancer cases per 100,000 persons and the maximum estimate, based on the Adventists study, resulted in 19.5 cases.

Unit risk-based estimate

With the unit risk-based method, the summary risk of all carcinogens yielded 0.79 annual cancer cases per 100,000 persons attributable to air pollution. Confidence intervals cannot be estimated using unit risk factors. Minimum and maximum estimates were 0.21 and 11.4 cases, respectively. The highest cancer risk was found for diesel exhaust, followed by 1,3-butadiene, chromium (VI), PAH, and benzene (Table 10-5). These five compounds accounted for 98% of the total cancer risk attributable to air pollution according to the unit risk-based approach. The minimum and maximum estimates reflect the range of unit risk factors published by different agencies. The wide estimate range of the unit risk-based cancer risk assessment is mainly determined by the large range of published unit risk factors for diesel exhaust.

Table 10-5. Result of the epidemiologic and the unit risk-based cancer risk assessments (Confidence intervals can only be calculated for the epidemiologic based approach).

unit risk-based single pollutants	Point estimate (95%-CI) in cases per 100,000 person years	Minimum estimate in cases per 100,000 person years	Maximum estimate in cases per 100,000 person years	Proportion of cancer risk [%]
diesel exhaust	0.48	0.054	10.8	61.5
1,3-butadiene	0.15	0.117	0.19	19.1
chromium (VI)	0.063	0.018	0.23	8.0
PAH	0.044	0.007	0.049	5.5
benzene	0.030	0.014	0.097	3.8
nickel	0.004	0.003	0.005	0.50
arsene	0.003	0.001	0.004	0.33
cadmium	0.003	0.001	0.007	0.33
tetrachloroethylene	0.003	0.003	0.003	0.33
1,4-dichlorobenzene	0.001	0.001	0.001	0.18
trichloromethane	0.001	0.001	0.003	0.17
lead	0.001	0.001	0.001	0.07
trichloroethylene	0.000	0.000	0.001	0.05
Total of the unit risk-based estimate	**0.79**	**0.21**	**11.4**	**100**
Epidemiology-based estimate	**6.7 (-0.78; 14.2)**	**1.1**	**19.5**	**100**

DISCUSSION

By means of the epidemiology-based cancer risk assessment, we obtained a point estimate of 6.7 annual lung cancer cases attributable to air pollution per 100,000 persons living in the Basel area. The unit risk-based method yielded a point estimate of 0.79 for all types of cancer, a value almost nine times lower than that of the epidemiology-based method. However, the minimum and maximum estimates of the two applied approaches were overlapping in a broad

range (1.1-19.5 and 0.21-11.4 for the epidemiologic and unit risk-based approach, respectively).

The uncertainty range of the unit risk-based assessment was mainly determined by the uncertainty regarding the risk of diesel exhaust, which was due to systematic differences between toxicological and epidemiological studies. The upper bound of the unit risk-based estimate was determined by an unit risk factor for diesel exhaust derived from an occupational diesel exhaust study (Steenland et al., 1998, Stayner et al., 1998).

Several assessments of cancer risk from air pollution based on unit risk factors are in line with our unit risk-based result: a study in Switzerland (Yetergil, 1998) yielded 0.85 (0.21-4.1) annual cancer cases per 100,000 persons. Diesel exhaust was estimated to cause 60% of the total risk attributable to air pollution. An unit risk-based assessment in Germany (Kappos and Schmitt, 1993) found a cancer risk of 1.14 excess cases per 100,000 person years in urban environments and 0.21/100,000 in rural areas, mostly caused by diesel exhaust. Furthermore, in Sweden particulate organic material (POM) was estimated to cause 0.7 cancer cases per 100,000 person years, and exposure to 1,3-butadiene was associated with 0.31 cases per 100,000 person years (Törnqvist and Ehrenberg, 1994). A recent unit risk-based assessment in CaliforniaMorello-Frosch et al., 2000 yielded an average of 0.43 cancer cases per 100,000 person years attributable to air pollution, with POM and 1,3-butadiene providing the largest contribution.

However, other approaches to assess cancer risk from air pollution resulted in higher estimates and are in line with our epidemiology-based risk assessment. For instance, ecologic studies, evaluating smoking-adjusted lung cancer risks between urban and rural sites, generally found a difference of 50% (Katsouyanni and Pershagen, 1997, Hemminki and Pershagen, 1994). This would correspond to 15 excess lung cancer cases per 100,000 person years in urban areas (Hemminki and Pershagen, 1994). However, it is not clear whether this number can be fully attributed to air pollution, as control of confounding is problematic in such studies.

Recently, a Swedish case-control study concluded that the proportion of lung cancer cases attributable to traffic-related air pollution in Stockholm can be as high as 10% (Nyberg et al., 2000). Compared to the actual observed lung cancer incidence of 50 annual cases per 100,000 persons in the Basel area (Torhorst, 1999), this finding corresponds to our epidemiology-based estimate (6.7 annual cases per 100,000 persons).

An estimate of the total annual mortality cases (all causes) attributable to air pollution in the Basel area, based on the method used in the tri-national study (Künzli et al., 2000), yielded 59 cases of death per 100,000 persons (Röösli et al., 2000b). In this light, an estimate up to 10 cancer cases does not seem unrealistic.

It is possible that the observed difference between the epidemiology-based and the unit risk-based point estimate may be caused by pure chance, considering the broad overlapping range of the two approaches. However, it is striking that the point estimate of the epidemiology-based assessment, using human epidemiologic data, provides a substantially higher point estimate than the unit risk assessment mainly based on toxicological data. This difference may reflect the conceptual differences between toxicology and epidemiology. In fact, the unit risk-based estimate would be very similar to the epidemiology-based estimate, if only human studies had been considered to derive the unit risk factors, in particular for diesel exhaust.

In a risk assessment study of cancer mortality from smoking a similar difference was observed between an estimate based on toxicological data as compared to an estimate based on human data, using animal and human data, respectively (Hertz-Picciotto et al., 2000). Thus, it is conceivable that the observed difference may be systematical, either due to an underestimation of the toxicology-based unit risk assessment or due to an overestimation of the epidemiology based assessment.

One reason which could explain an underestimation of the toxicology-based unit risk assessment is the possibility that an important carcinogen was not included in the unit risk-based assessment. However, this seems unlikely as none of the previous studies has identified such a strong additional carcinogenic air pollutant.

A further explanation could be that a large part of the carcinogenic potency from air pollution is not caused by single pollutants but by interaction processes of various components in the ambient air pollution mixture. Therefore, the total effect of all components influencing different organ systems in humans might be larger than the sum of the individual effects of each component. This would imply that it is not possible to quantify the cancerogenity of air pollution by simply adding up the carcinogenic potencies of each pollutant as done in the unit risk method. Thus, only studies on human populations under real exposure conditions could adequately assess the cancer risk of the general population attributable to ambient air pollution. In such studies (e.g. Adventists-, the Six-Cities- and the American Cancer Society Study as well as the occupational diesel exhaust studies) PM_{10} or diesel exhaust may then be interpreted as a proxy of the total cancerogenity of ambient air pollution.

However, other arguments may suggest an overestimation of the epidemiology based assessment. The key assumption of the epidemiology-based method (i.e. that PM_{10} can be used as a proxy for the total carcinogenic effect of ambient air pollution) cannot be adequately proven at present. Generally, site to site correlation between various pollutants is relatively high. However, the American Cancer Society Study (Pope et al., 1995), which found an association between lung cancer and sulfate, but almost no relation to $PM_{2.5}$, indicates the limits of the proxy-concept. It might be especially problematic to extrapolate results of long term effect studies from the US to European populations, as there may be systematic differences in the composition of the ambient air pollution mixture between the U.S. and Europe. On the other hand, according to recent time series studies of air pollution and health there is no evidence of relevant discrepancies between European and U.S. studies (Samet et al., 2000a, Katsouyanni et al., 1997).

An overestimation of the epidemiology-based assessment could also be the result of residual confounding in the cohort studies. Though the important confounding factors were taken into account it cannot be excluded that the reported relative risks are overestimating the true exposure-response association due to residual confounding. Even slightly overestimated relative risks would appreciably affect the result of the risk assessment.

Furthermore, one may argue that cohort studies overestimate the exposure-response function, if exposure assignment is based on current rather than historic pollution levels. For pollutants with steadily decreasing concentrations this may play a role. Further studies on exposure trends, latency periods and effects are needed, particularly in Europe.

Every approach to assess cancer risk in a given population has its inherent limitations. The presented epidemiology-based method may be an integrative way to deal with combined effects of an air pollutant mixture, which is often considered a key issue in risk assessment. However, more experience is needed to judge the validity of the assumption that one single pollutant (in this case PM_{10}) can be used as a proxy of the cancerogenity of ambient air pollution.

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PART VI
GENERAL DISCUSSION AND CONCLUSIONS

11 Discussion and Conclusions

11.1 Initial situation

Spatial and temporal variability of PM determines exposure assessment

In environmental epidemiology exposure assessment is a central topic. On the one hand exposure assessment is part of each environmental study, on the other hand it is a step in risk assessment. The quality of the exposure assessment determines in large part the validity of an environmental epidemiology study as well as the result of a risk quantification. Unfortunately, it is a basic characteristic of nature that a perfect exposure assessment can never be developed. Thus, every exposure assessment is based on assumption, which should be as reliable as possible. Knowledge of spatial and temporal variability of air pollutants is the base for accurate assumptions in air pollution health studies.

For that reason we investigated the temporal and spatial variability of air pollutants in the Basel area, Switzerland. Based on these findings we performed a risk assessment to quantify the carcinogenic and non-carcinogenic health risk, which is associated with the occurring pollution levels.

11.2 Main findings

Comparing PM_4, PM_{10}, and TSP

In Basel, the ratio of the mean PM_4 to mean PM_{10} concentrations was 81% and the ratio was 75% for the mean PM_{10} versus TSP concentrations. Comparison measurements showed that PM_4 was approximately 11% higher than $PM_{2.5}$. From the elemental analysis it can be concluded that Zn, Pb, Br, Cu and S were mainly present in small particles, the mineralogical elements and chloride primarily existed in the coarse fraction.

Seasonal variation of PM

In the city of Basel, during the cold season particulate matter concentrations were considerably higher than during the warm season. In contrast, at the more elevated rural sites this distinctive seasonal pattern was not observed. The chemical analyses revealed that seasonal differences were mainly caused by nitrate, ammonium and organic matter whereas elemental carbon and mineralogical elements remained more or less constant during the year at both

urban and rural sites.

Daily variation of PM

The day-to-day levels of particulate matter were fluctuating noticeably. These variations could be explained to a large part by meteorological parameters. The statistical association was particularly strong with temperature, wind direction, wind velocity, origin of air, and vertical temperature gradient. Comparing mean values by day showed a clear trend with highest PM values on Wednesday and Thursday and lowest values on weekend. This daily pattern was strongest for TSP and lowest for PM_4. The workday/weekend difference of the chemical compounds - as an indicator of human activities - was most pronounced for mineralogical elements and elemental carbon.

Spatial variability of PM within Basel

Generally, the spatial variability of PM was rather limited in the city of Basel, i.e. mean PM_{10} concentration at the six temporary sites with different traffic densities was in the range of less than ±10% of the mean urban PM_{10} level. Generally, traffic density had only weak tendency to increase mean PM levels. However, considerably higher values were found at one site near to a traffic light in a street canyon. The spatial variability in the city was similar for PM_4 and PM_{10} but somewhat higher for TSP. Highest small-scale spatial variability was found for the carbonaceous compounds, Ca, Pb, Fe, Cr and Br. Most homogeneous distributed were S, Ti and K.

Spatial variability of PM between urban and rural sites

The difference between urban and rural sites showed a clear seasonal pattern. In summer, differences between urban and rural sites were small, whereas in winter the differences were more pronounced. This was caused mainly by days with persistent surface inversion. It could therefore be concluded that spatial variability of annual mean PM concentration between urban and rural sites was more likely be caused by varying altitude than by distance to the city center. From the analyzed substances the differences between urban and rural sites were most pronounced for carbonaceous compounds, Pb and chloride. Smallest differences were found for sulfate, ammonium and cadmium.

Comparison of PM with other pollutants

Comparing the spatial variability of PM_{10} in the city of Basel with other air pollutants revealed that a likewise homogenous distribution was found for O_3 and CO. All other pollutants, such as SO_2, NO_X, PAH, VOC were more heterogeneously distributed. Highest small-scale gradients were found for NO

and the VOC (Röösli et al., 1999).

Particle numbers

The ultrafine particle mode (<0.1 µm) included approximately 85% of the total particle numbers <0.421 µm while the accumulation mode (0.1-2.8 µm) made up for most of the PM_{10} mass. Day profiles of size specific number profiles resulted in highest concentration during morning and evening rush hours. The profile was more pronounced at traffic-exposed sites compared to a residential site and more distinctive for small particle size than larger particle size. The diurnal profile of particle numbers as well as the concentrations of particle-bound PAH (pPAH) was more closely correlated to the number of heavy duty vehicles than to the number of passenger cars.

Carcinogenic health risk

The quantification of the cancer risk from air pollution yielded 0.8 (range: 0.2-11.4) annual lung cancer cases per 100,000 persons in our study area when the unit risk-based approach was used. By means of the indicator-based approach resulted 6.7 (95%-CI: -0.8 to 14.2) cases. Generally estimates based on human data yielded a higher risk. The range of the unit risk-based estimate is mainly determined by a wide range of published unit risk factors for diesel exhaust according of the source of data, animal or human ones. Estimating the cancer risk from diesel exhaust only based on human studies resulted in 11.4 (95%-CI: 3.1-19.7) lung cancer cases per 100,000 person years in the study area.

Meta-analysis

The indicator-based method showed that in meta-analysis the type of effect measure influences the result by a large extent when the health end point of interest is associated with stronger risk factor than air pollution such as smoking and lung cancer. Absolute risk measures are independent of the incidence in the target population, in contrast to relative measures, which are influenced by it. However, the incidence in the target population can be quite different from the one in the base population due to differences in the profiles of other risk factors than considered (e.g. smoking habits).

Non-carcinogenic health risk

In total 59 (95%-CI: 36-81) premature deaths per 100,000 person years are associated with air pollution in the cantons Basel-Stadt and Basel-Land. Further a significant number of respiratory and cardiovascular hospital admissions, chronic bronchitis, acute bronchitis, restricted activity days, and asthma attacks had been quantified (Röösli et al., 2000).

11.3 Discussion

11.3.1 Air pollution analysis

Traffic and spatial variability

PM_{10} was generally homogeneously distributed in the urban environment of Basel. This result was also found in other cities in Europe and United States with different building characteristics and different topography (Burton et al., 1996, Chow et al., 1992, Harrison, 1997, Wilson and Suh, 1997). In this study we compared mean particulate matter concentration of several sites differing mainly with respect to their road traffic densities. In the first place this concept of measure allows to investigate the effect of road traffic on mean particulate matter measurement. The measurements showed that proximity to road traffic had only a weak tendency to increase mean PM levels, though traffic is considered as the main source of particulate matter in industrialized regions. How can this finding be explained?

Residence time

First of all, particles have a long residence time in the atmosphere. The residence time in the order of several days is longer than for most gaseous pollutants. This allows an efficient mixing of emitted particles in a wide area around the emission source and explains the more homogeneous distribution of particles as compared to most other gaseous pollutants with shorter atmospheric lifetime. The effect of the residence time on the spatial variability can be observed when comparing PM_{10} and TSP; the shorter residence time of large particles (>10µm) resulted in a higher spatial variability of TSP than PM_{10}.

Mean value

Another point is that we investigated 24 h mean values. As was shown in the daily profiles of particle numbers at sites exposed to high traffic volume, the levels were mainly elevated during the morning and evening rush hours but not the rest of the day and night. Thus, short-term difference during rush hours are flattened out in 24 h mean values.

PM_{10} is a mixture of different sources

Another smoothing effect yielding homogeneous distribution is a result of the fact that PM is a mixture of different sources. This was clearly demonstrated by the example of the bonfire from 1st August. Though the concentration of some bonfire related substances such as potassium were up to 100 times higher after the bonfire than before, the total PM mass concentration was only

slightly influenced. An analogous situation may be found at busy streets: only a relative small increase of the total average PM_{10} concentration can be observed, but some specific primary traffic related compounds such as EC, OM and Pb may increase much more.

Secondary particles

Moreover, it is to take into account that in Basel almost half of the particulate matter concentration is consisting of particles, which are secondary produced from gaseous precursor substances. Traffic is as well an influential emission source of these precursor substances, mainly NO_3, partly also NH_4 and VOC. However, the gas-to-particle conversion is a relatively slow process, blurring spatial variability.

Non-traffic influences on the spatial variability

Despite traffic, some indications were found that construction work seems to have a noticeable influence on the spatial variability of PM, mainly on the coarse fractions. For instance, the Ca concentration - usually part of construction materials - showed a more distinctive workday/weekend ratio than would be expected from a substance emitted by traffic resuspension alone. Other emission sources strongly influencing the spatial variability of particulate matter in Basel were not found.

PM_{10} as exposure mass

From the homogeneous distribution of particulate matter mass concentration (e.g. PM_{10}) in an urban environment it can be concluded that its use as exposure mass in cross-sectional studies is not expected to introduce a large amount of exposure error. However, from the point of view of biological plausibility it is often argued that the mass concentration may not be the most plausible exposure measure relating air pollution and human health because it is strongly influenced by large particles, which are often found not acting on human health (Laden et al., 2000). The number of particles or some specific compounds in PM is often declared as more reliable exposure measure (Peters and Wichmann, 1996). Still, there is no concordance concerning this open issue (Pekkanen et al., 1997).

Particle numbers and specific compounds as exposure measure

Both, particle numbers and the concentration of traffic related chemical compounds in PM are considerably stronger associated with traffic density and therefore more heterogeneously distributed within Basel than PM mass concentration.

Investigate causality using epidemiologic studies

Thus, the higher spatial variability increases the likelihood of exposure misclassification in cross-sectional studies. This reduces the precision, when the error is uncorrelated with exposure, or even bias the effect strength, when the error is correlated with the exposure levels or when the site-specific pollutant values are not representing average exposure of the population living in the corresponding site. Thus, the strength of the statistical association between health effects and air pollutants is determined by the spatial distribution of the pollutants as well as by the biological causality. Simply comparing the statistical association in order to conclude on biological causality might be crucial (e.g. Mar et al., 2000, Moolgavkar, 2000, Samet et al., 2000a).

PM_{10} as an indicator of air pollution exposure

It is possible that the homogenous distribution of particulate matter within urban environments explains its successful application as an indicator of air pollution exposure in many circumstances. The smaller likelihood of exposure misclassification as compared to other pollutants may allow a more stable generalization of the exposure situation in different geographic contexts. Moreover, particulate matter is representing most air pollution sources, whereas other pollutants are often typical emissions of specific sources such as NO_X from road traffic or SO_2 from coal and oil burning.

Influence from regional sources

Changing the point of view from an epidemiologist to a policy maker, the homogenous distribution of particulate matter raises another question: what spatial scale of emission sources needs to be focused in order to reduce particulate matter levels at a given place? Does a homogenous distribution mean that long-range transport is most important and regional emissions are negligible? These questions may not be concluding answered without a detailed source appointment. It is undoubtedly appreciated that long scale transport of particles influences the PM levels. However, this study gained some hints that regional sources cannot be neglected.

Workday to weekend ratio

First, a pronounced difference between mean concentration on workday compared to the one on weekend was found. This ratio is mainly influenced by regional human activities, which are reduced over the weekend such as road traffic, construction work and industrial processes. Workday/weekend

patterns from sources more far away are expected to be smoothed due to longer transport time.

Inversions

Second, highest PM concentrations were found in Basel during persistent inversion situations of several days. In those situations the air exchange on a large spatial scale is small leading to an enrichment of locally emitted pollutants below the inversion layer. Thus, highest levels in Basel are probably mainly caused by regional emissions. At the same time at the more elevated rural sites, when uncoupled from the city, considerably lower values were measured.

11.3.2 Risk assessment

Individual vs. collective risk

The estimated magnitude was about 4-30 annual lung cancer cases attributable to air pollution in the cantons Basel-Stadt and -Landschaft. Compared to the total 230 annual lung cancer cases that had actually occurred in the study area it can be concluded from the estimated number that the public-health impact from air pollution is substantial. Moreover, air pollution is related with a number of non-carcinogenic health risks, such as premature mortality (264 annual cases), bronchitis, asthma, etc. In contrast, average individual risk to develop lung cancer attributable to air pollution during lifetime is below 1 percent. Thus, from an individual perspective the lung cancer risk from air pollution is relatively small, especially when compared with smoking. The lung cancer risk of smokers is at least one order of magnitude higher than that of non-smokers (Lewtas, 1993). However, smoking is a self-chosen risk, whereas everybody is exposed to air pollution. Risks are generally judged differently depending whether they can be chosen or not.

Preventable fraction

Attributable cases are commonly interpreted as the preventable fraction. Based on the model of the sufficient cause consisting of several component causes, the preventable fraction is meant to be prevented when one component cause (i.e. air pollution) had been removed. However, it has to be taken into account that for a long-term effect such as lung cancer, the benefit of lower air pollution levels would take years to be fully realized (Künzli et al., 2000). Further, one has to consider the fact that the attributable risk estimate does not take competing risk into account. Thus, it is well known in multi-

causal diseases that the sum of attributable cases across several risk factors may be larger than 100 percent (see Chapter 3.7). It is therefore warranted of the wrong conclusions that the 15 percent which is attributed to air pollution can be added to 100% to estimate that maximal 85% percent of the lung cancer cases can be caused by smoking. Impact measures that take competing risks into account need to be developed, first.

Difference between unit risk- and indicator-based estimate

The most striking finding of the risk assessment was the considerable difference of the estimates of cancer risk according to the method used. The indicator-based estimate was approximately nine times higher than the unit risk-based. Moreover, a unit risk estimate for diesel exhaust only taking human occupational studies into account yielded a 14 times higher value than the point estimate of the unit risk-based approach, which is based mainly on animal studies.

As every risk assessment is based on the current state of knowledge, which can never be perfect, there will always remain some extent of uncertainties. Possible uncertainties are discussed in the following and it is judged the likelihood that they explaining the systematic difference between the two approaches.

Limitations in the exposure assessment of PM_{10}

The accuracy of the exposure assessment determines the result of a risk assessment immediately. The exposure assessment of PM_{10} was based on a dispersion model with a spatial resolution of 1 km^2. Comparisons of modeled and measured PM_{10} values in our study area showed a good concordance. Thus, the exposure assessment of PM_{10} may be quite accurate in particular when regarding that mean PM_{10} levels generally do not show strong small-scale gradients.

Exposure assessment of carcinogens in Basel

The exposure assessment of the single carcinogens might be less accurate. Some substances were found to be quite heterogeneously distributed within Basel. Nevertheless, the study area was classified only in three exposure levels, as the data quality was considered to be too poor to define a more detailed differentiation. The measurements performed throughout one year at six sites representing different types of living places in the city of Basel are expected to provide a reliable mean value for the whole city. Thus, it is

expected that inevitable over- and underestimations of pollution levels for some quarters in Basel are roughly compensating each other, resulting in a reliable mean exposure assessment.

Carcinogens at rural sites

For some substances lowest levels occurring at rural sites may be quite inaccurate, since they were solely based on assumptions but not on measurements. However, it has to be considered that these low levels in low densely populated areas are not strongly affecting the result of a risk assessment, even when the true levels are quite different.

Assignment of the geographical distribution

The geographical distribution of the three pollution levels of the single carcinogens was assigned based on the spatial variability of PM_{10}. None of the single carcinogens is optimally correlated with PM_{10} concerning the spatial distribution. Most carcinogens showed a considerably higher spatial variability than PM_{10}. Thus, errors in the reallocation of the areas to the three pollution levels are inevitable and possibly biasing the result of the risk assessment. However, it is expected that in the average exposure assessment a large amount of these errors are compensating each other due to over- and underestimation.

Moving people

A similar compensating effect is expected from the fact that most people stay not all the time at their living place. Thus, the exposure of people living in high polluted areas are generally overestimated because they stay sometimes in lower polluted areas, whereas the exposure of people living in low polluted areas is generally underestimated due to their stay in more polluted areas at times.

Conclusions concerning exposure assessment

Finally, it can be concluded that the PM_{10} exposure assessment is more accurate than the exposure assessment of the single carcinogens. The latter may be afflicted with a large amount of uncertainty at one specific point in the study area. However, most of these exposure misclassifications are expected to compensate each other by calculating mean values. It is therefore not expected that the large difference between indicator- and unit risk-based assessment could be entirely explained with errors in the exposure assessment.

Indicator key assumption

The key assumption of the indicator-based assessment is that PM_{10} can appropriately be used as an indicator of the carcinogenic potency of air pollution. PM_{10} contains some carcinogenic compounds such as diesel exhaust, chromium VI, nickel, PAH, etc. However, large parts of PM_{10} are not expected to be carcinogenic. On the contrary a group of gaseous air pollutants is well known to be carcinogenic such as benzene and 1,3-butadiene.

Transfer of the exposure-response association from USA to Europe

Thus, it is possible that PM_{10} is not an ideal measure of cancerogenicity, especially when transferring an exposure-response association of PM_{10} and cancer from the United States to the situation in Basel. In various geographical contexts PM_{10} may represent different emission sources emitting pollutants with different cancerogenicity. Generally, there is little evidence that the composition of PM_{10} is substantial different between Europe and the United States comparing corresponding climatic zones. Only in the arid zones of the U.S. the proportion of mineralogical elements in PM_{10} is much higher than in Central Europe. Assuming that mineralogical elements are not associated with cancer (Laden et al., 2000), this fact would result in underestimation of the true association between PM_{10} and cancer if transferred to Europe. Moreover, in regard to short-term health effects of PM_{10} there is no evidence of relevant discrepancies in the exposure response relation between European and U.S. studies (Katsouyanni et al., 1997).

Indicators in the Cancer Society study

However, the Cancer Society study (Pope et al., 1995) shows the weakness of the indicator assumption. It found an association between lung cancer and sulfate but hardly a relation to $PM_{2.5}$. This finding is still not explained. There exist some hints that an appropriate indicator of air pollutant has mainly to represent combustion emission sources (Pope, 2000, Mar et al., 2000, Wyler et al., 2000, Laden et al., 2000). Thus, It is possible that $PM_{2.5}$ does not represent exposure to these sources when concurrently applied in arid and non-arid zones as done in the Cancer Society study.

Absolute vs. relative measure

The assumption, which has to be made to combine air pollution effects in smokers and non-smokers, is another uncertainty of the indicator-based estimate. In the Adventist's non-smoker cohort the relative risk for lung

cancer attributable to 10 µg/m³ increase in average PM_{10} level was considerably higher than in the other two cohorts consisting of both, smokers and non-smokers. However, due to a substantially lower cancer incidence in the Adventist's cohort, this heterogeneity among the studies could be considerably reduced when absolute risk measures (excess rates) were compared instead of relative measures of risk. Regarding absolute risk measures instead of relative risk measures implies that one assumes implicitly that the both mixture tobacco smoke and air pollution are causal acting on an additive scale independently of each other. Though this assumption cannot be proven from the perspective of biological plausibility, it is justified based on the theoretical consideration that both factor are acting on the same biological mechanism, which is presumed for two additively acting factors.

Additivity and synergy

To avoid misapprehension when considering the additively acting of two mixtures, it is emphasized that this can nevertheless mean that synergistic effects can be occurring among the various pollutants within each mixture. It will be discussed later that this situation is very likely.

Latency time

A further uncertainty associated with the indicator-based method relates to the fact that the exposure-response relation may be overestimated when considering current pollutant levels since levels of air pollution have been decreasing in recent years and lung cancer develop only after a latency period. On the other hand the long latency period increased the likelihood of exposure misclassification in the studies, possibly biasing the true exposure-response association to the null.

Age

The lung cancer incidence of the Six-Cities study population is relatively high (106/100,000 person years) compared to the actually observed lung cancer incidence in Basel (50/100,000 person years). This difference is not caused by the smoking habits, as the smoking rates are similar in both populations. Likewise, the lung cancer incidence in the Adventists study was relatively high (36/100,000 person years) when taking into account their non-smoker status. The higher susceptibility to lung cancer of the study populations as compared to the target population can probably be explained by a different age distribution. It was not possible to prove this hypothesis based on the

available data, but if so, it would be expected to overestimate the risk of the lung cancer attributable to air pollution. On the contrary, persons below 30 years were not considered resulting in a slightly underestimation of lung cancer risk.

Including of all relevant carcinogens?

In the unit risk-based assessment, it is conceivable that an important carcinogen was not included. However, this seems to be relatively unlikely, as none of the previous studies has identified such a strong additional carcinogenic air pollutant.

Geometrical vs. arithmetic mean

One may argue whether it is appropriate to average the unit risk factors published by various agencies using the geometric mean. The arithmetic mean would better conform to the additive model, on which the unit risk concept is based. However, for some substances published unit risk factors differ by an order of magnitude or more. Therefore, given its high sensitivity to extreme values, the arithmetic mean may be of limited validity. In fact, a sensitivity analysis has shown that applying the arithmetic mean value would not substantially increase the result of the unit risk-based method.

Confounding

It is very unlikely that one of the uncertainties stated above can explain completely the observed substantial difference between the unit risk-based and the indicator-based assessment. Only possibly confounding in the cohort studies, from which the association between PM_{10} and cancer has been derived, can be an explanation, which theoretically can completely explain the difference between the unit risk- and the indicator-based risk estimate, since it is a basic characteristic of nature that the extent of confounding cannot be quantified. However, the most important confounding factors 'smoking' and 'occupational exposure' were controlled in each of the three studies. Influence on the study results from residual confounding or factors, which were not considered, cannot be excluded.

Systematic difference between animal and human studies

However, the systematic pattern between the unit risk and the indicator-based method suggests a systematic difference between the use of epidemiologic and experimental data in risk assessment. Hints for such a systematic difference between an estimate based on animal and on human data was also observed in a risk assessment study of cancer mortality from smoking using

either toxicological or human data (Hertz-Picciotto et al., 2000).

Underestimation of cancer risk from diesel exhaust in animal studies

It seems possible that animal studies greatly underestimate the cancer risk of diesel exhaust. Animal studies of cancer risk from diesel exhaust can be varying in a wide range depending on the species. General highest cancerogenicity was found in rats, somewhat lower in mice, and barely effects were detected in cats and monkeys (U.S.EPA, 1999a). Moreover, animal studies showed consistently a lower risk than occupational studies. Thus, unit risk estimates for diesel exhaust published by the IARC and WHO which are poorly based on animal data are considerably lower than an unit risk estimate from the OEHHA considering animal as well as human data. A cancer risk assessment based on an unit risk estimate of diesel exhaust taking into account only occupationally studies would reveal a similar result like the indicator-based method. That means, that in such an unit risk-based assessment almost all cancer cases attributable to air pollution were caused by diesel exhaust. The cancerogenicity of the other components of air pollution would be small compared to diesel exhaust. 1,3-butadiene, the substance with the second highest carcinogenic potency, would cause almost 100 times less cancer cases than diesel exhaust.

Synergistic effects

Another possible systematic reason which can explain the difference between the unit risk-based and the indicator-based assessment may be that a large part of the carcinogenic potency from air pollution is not caused by single pollutants but by interaction processes of various components in the ambient air mixture. Therefore, the total effect of all components influencing different systems in humans might be larger than the sum of the individual effects of each component. For instance, substance A inhibits the DNA repair mechanism, substance B is genotoxic, substance C influence cell growth, and substance D acts on the immune system. It is evident that the summary effect of these four substances is larger than the added single effects. However, these synergistic effects are not quantified when conceiving the cancer risk of air pollution as sum of the potency of the single agents as was done in the unit-risk-based approach.

Comparing PM_{10} studies and diesel exhaust studies

If synergistic effects mainly cause the cancerogenicity of air pollution, then only studies on human populations under real exposure conditions can adequately assess the cancer risk of the general population, such as the Adventists-, the Six-Cities- and the Cancer Society study using PM_{10} as a pollution indicator. However, under this perspective the occupational diesel exhaust studies (Steenland et al., 1990, Steenland et al., 1992, Steenland et al., 1998, Garshick et al., 1987, Garshick et al., 1988) may be belonging in the same category. In these studies exposure to diesel exhaust can be interpreted as surrogate for exposure to the general air pollution, as their study subjects are expected to be exposed to a similar pollutant mixture like the general population. Performing an exposure assessment of PM_{10} or diesel exhaust would very likely produce a similar result; both can be conceived as a proxy.

This viewpoint may be supported by the fact that the epidemiologic PM studies as well as the occupational diesel exhaust studies result in a similar cancer risk attributable to air pollution when applied on the situation in Basel.

Unit risk of diesel exhaust is including synergistic effects

Comparing the cancerogenicity of the substances evaluated by means of the unit risk-based approach, it is striking that diesel exhaust has the highest carcinogenic potency. This may support the hypothesis of synergistic effects, because just diesel exhaust is the sole compound, which was a priori analyzed as a mixture. Thus, synergistic effects among the solely components of diesel exhaust are included in the unit risk estimate, even in animal studies.

Implications for a policy maker

When changing the perspective from an epidemiologist to a policy maker, the hypothesis that cancerogenicity is caused rather by synergistic effects than by single pollutants may set hurdles in regulatory decisions on the first view. It raises the question which substance has to be reduced. It implies a new paradigm in the setting of environmental exposure standards. So far, it was set standards for some single pollutant mainly based on their causal acting on human health. Pursuing this strategy, worldwide a lot of success was achieved in the last years. For instance, in the last decade the sulfur dioxide concentration in ambient air was significantly reduced in Switzerland due to specific sanctions, such as reduction of sulfur in mineral oil or revisions of heating

systems (BUWAL, 1998). However, regarding synergistic effects new strategies may increase the effectiveness of regulatory decisions in the future.

New paradigm in regulatory decision

A new way would be a perspective, which is less focussed on single agents than rather on the entire air pollution. It is interesting to notice that due to the introduction of the PM_{10} standard in 1998, the Swiss regulatory policy has developed in this direction, yet. Considering the catalog of possible sanctions to reduce PM_{10} levels (Kropf, 2000), some particle specific actions were found (e.g. particle filters), however, most suggestions are acting integrative on all air pollutants, such as improvement in regional planning, steering tax, propagating of car sharing and public traffic, etc.

Of course, the implications of an indicator-based strategy on the air pollution control are not trivial. Finally, some pollutant specific standards cannot resign in regulatory decision. Without objective measures an air pollution control strategy is likely to become arbitrary. Thus, the regulatory policy, which was performed so far, has not been changed markedly in practice. However, allowing for synergistic effects, air pollutants standards may rather be set and controlled on the 'indicator-perspective' - as a proxy of specific emission sources - than on their causal acting on human health as single agent.

Abbreviations

BC	Black carbon (black smoke)
BRISKA	Basel Air Quality Risk Assessment Project (Basler Risikostudie Aussenluft)
BUWAL	Swiss Federal Agency of Environment, Forest and Landscape (Bundesamt für Umwelt, Wald und Landschaft)
CO	Carbon monoxide
CO_2	Carbon dioxide
EC	Elemental carbon
HD	Heavy duty vehicles
HVS	High Volume Sampler
IARC	International Agency on Research on Cancer
ISPM	Institute for Social and Preventive Medecine (Institut für Sozial- und Präventivmedizin)
LAI	Länderausschuss für Immissionsschutz des Landes Nordrhein-Westfalen
LD	Light duty vehicles
LHA	Air Quality Management Agency of Basel (Lufthygieneamt beider Basel)
LOAEL	Lowest observed concentration that did produce an adverse effect
MGU	Stiftung Mensch - Gesellschaft - Umwelt der Universität Basel
NOAEL	Highest observed concentration that did not produce any adverse effect
NO	Nitrogen monoxide
NO_2	Nitrogen dioxide
NO_X	Nitrogen oxide
OC	Organic carbon
OEEHA	Office of Environmental Health Hazard Assessment, California
OM	Organic matter
PAH	Polycyclic aromatic hydrocarbons

PAS	Photoelectric Aerosol Sensor
PM_{10}	Particles with an aerodynamic diameter smaller than 10 µm
$PM_{2.5}$, PM_4	Particles with an aerodynamic diameter smaller than 2.5 µm, and 4 µm, respectively
pPAH	Particle-pound polycyclic aromatic hydrocarbons
SMPS	Scanning Mobility Particle Sizer
SO_2	Sulfur dioxide
UR	Unit Risk
U.S.EPA	United States Environmental Protection Agency
TSP	Total suspended particulate

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Curriculum Vitae

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Education and Professional Experience

1974-1980	Primary school in Hitzkirch LU, Switzerland
1980-1983	Gymnasium in Hochdorf LU, Switzerland
1983–1988	Lehrerinnen- und Lehrerseminar in Hitzkirch LU, Switzerland
1988-1991	Primary School teacher in Dagmersellen LU, Switzerland
1991-1992	Various stages and temporary jobs as well as preparation of the entrance examination for the Swiss Federal Institute of Technology in Zürich (ETHZ)
1992–1997	Studies in Environmental Science at the Swiss Federal Institute of Technology in Zürich (ETHZ) with focus on atmospheric physics.
1997-1998	Research assistant at the Institute of Atmospheric Science of the Swiss Federal Institute of Technology Hönggerberg. Data preparation and analysis of UV-B Brewer measurement in collaboration with the Schweizerische Meteorologische Anstalt
1998-2001	Project assistant for the Basel Air Quality Risk Assessment Project (BRISKA) and Ph D student at the Institute of social and preventive medicine, University of Basel, Switzerland under supervision of PD Dr. med. Charlotte Braun-Fahrländer.
1999	Summerschool in Epidemiology at the Epidemiology Research Institute in Boston, MA. (Lecturers: P. Cole, M.A. Mittleman, D.B. Petitti, K. Rothman, D. Trichopoulos)
1999	Master of public health postgraduate course in Environmental Epidemiology (incl. Workshop) in Basel. (Lecturers: U. Ackermann-

	Liebrich, C. Braun-Fahrländer, D. Dockery, N. Künzli, A. Pope, J. Schwartz)
2000-2001	Parts of the postgraduate course in statistic at the Swiss Federal Institute of Technology in Zürich (regression analysis, multivariate data analysis and time series). (Lecturers: Y. Grize, M. Hürzeler, M. Mächler, R. Maronna, A. Papritz, L. Pritscher, A. Ruckstuhl, W. Stahel)
1998-2001	Various colloquiums at the Institute for Social and Preventive Medicine (div. lecturers) and 'Reading Epidemiologic Papers' (Lecturer: N. Künzli).

Publications

Röösli, M., Braun-Fahrländer, C., Künzli, N., Oglesby, L., G., T., Camenzind, M., Mathys, P. and Staehelin, J. (2000): Spatial variability of different fractions of particulate matter within an urban environment and between urban and rural sites. *Journal of the Air & Waste Management Association,* **50,** 1115-1124.

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