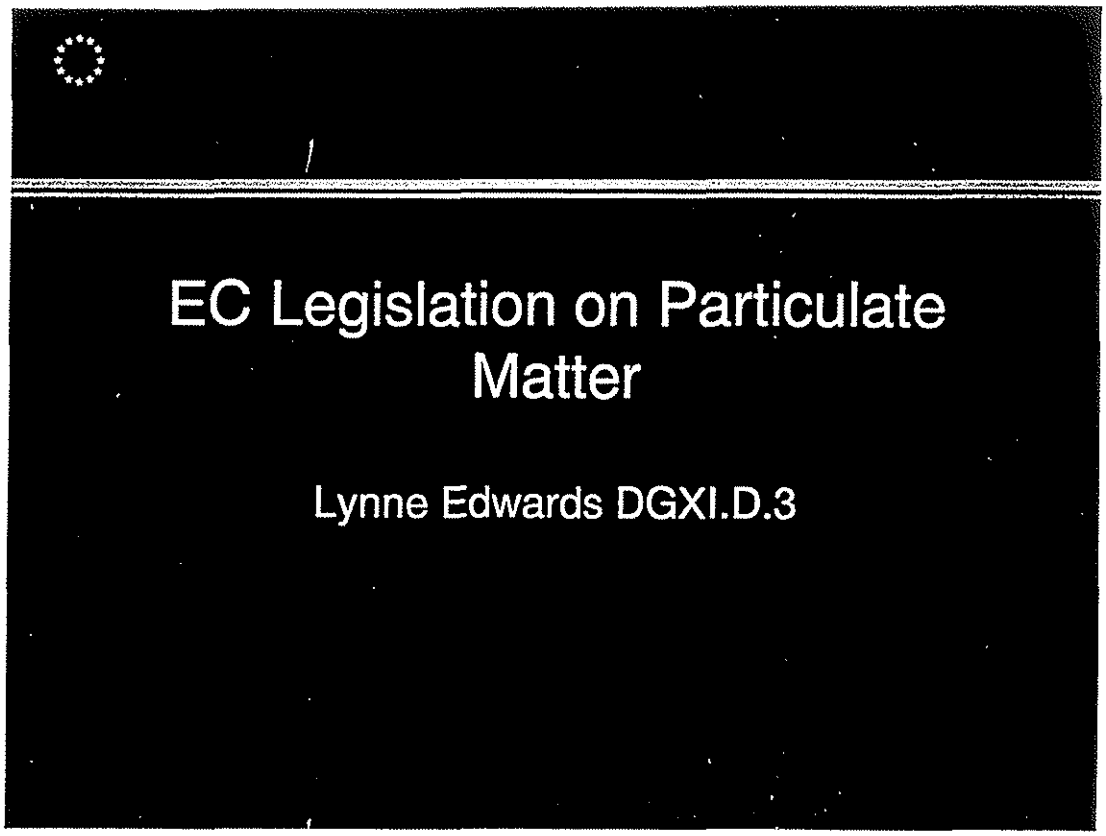
II. MEETING PRESENTATIONS

Opening Session

Chair: *Prudencio Perera*, European Commission, DG XI

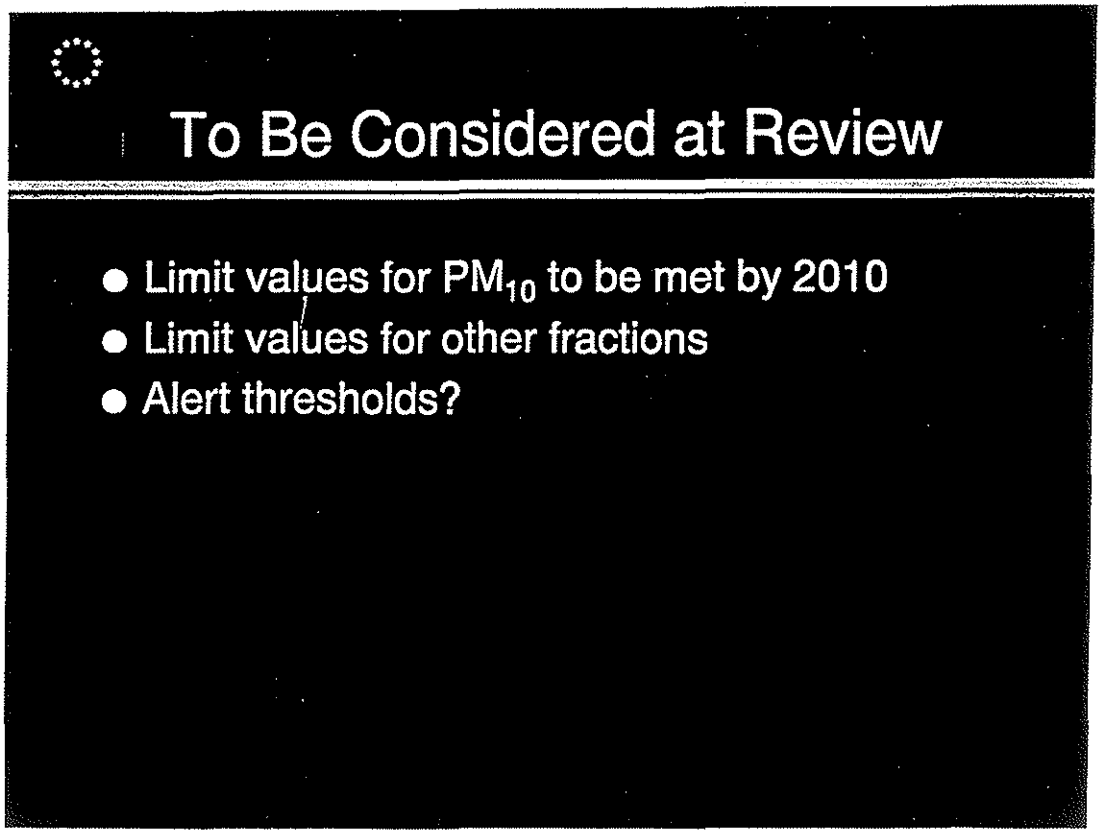
LYNNE EDWARDS

European Commission, DG XI



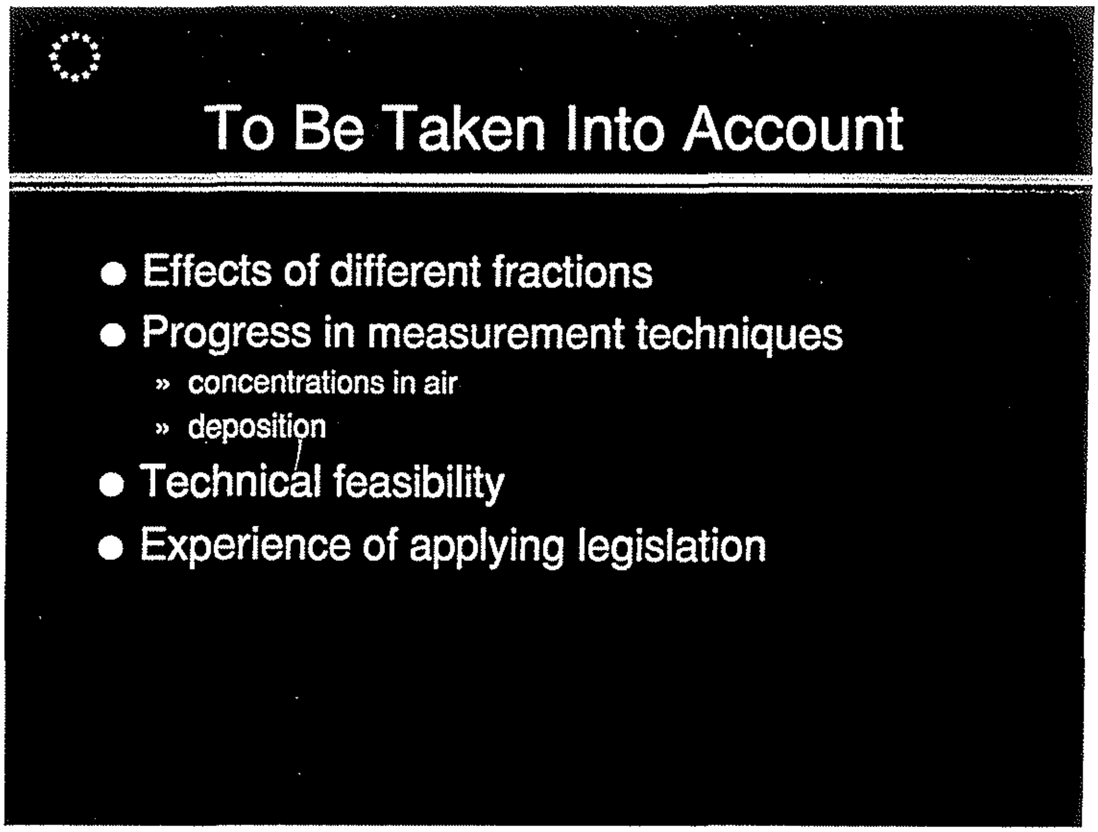
EC Legislation on Particulate Matter

Lynne Edwards DGXI.D.3



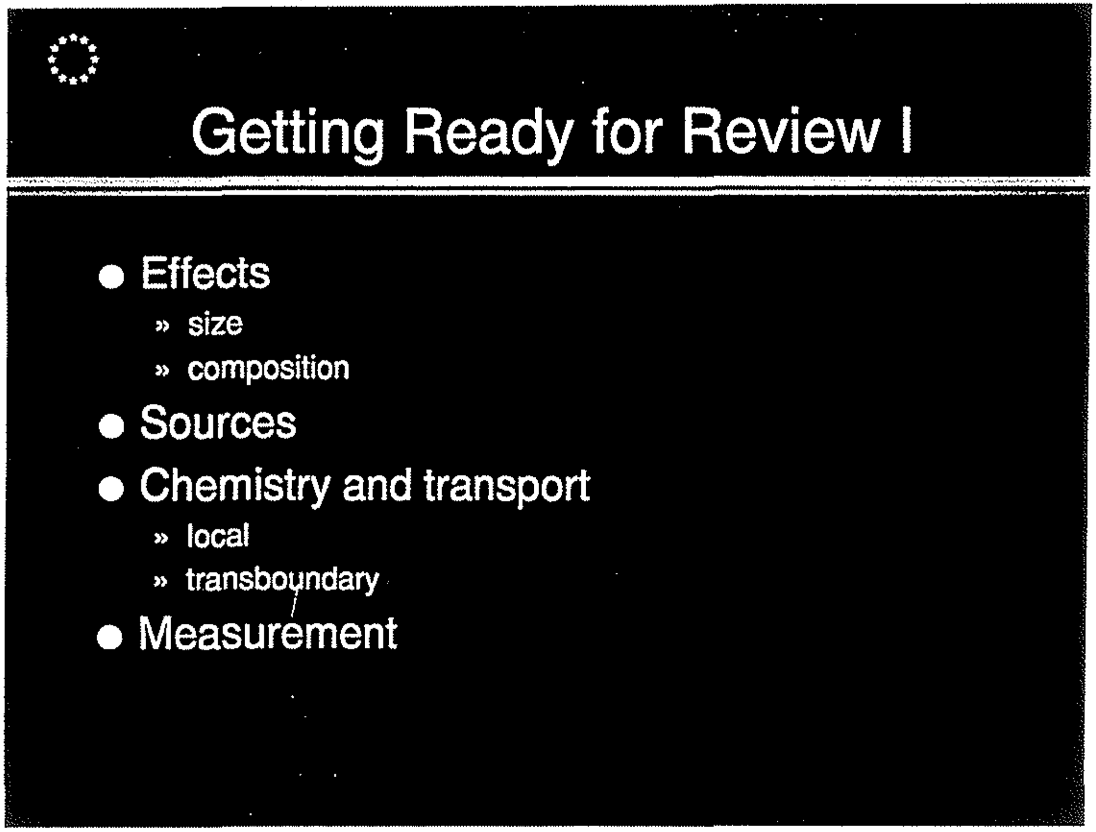
To Be Considered at Review

- Limit values for PM_{10} to be met by 2010
- Limit values for other fractions
- Alert thresholds?



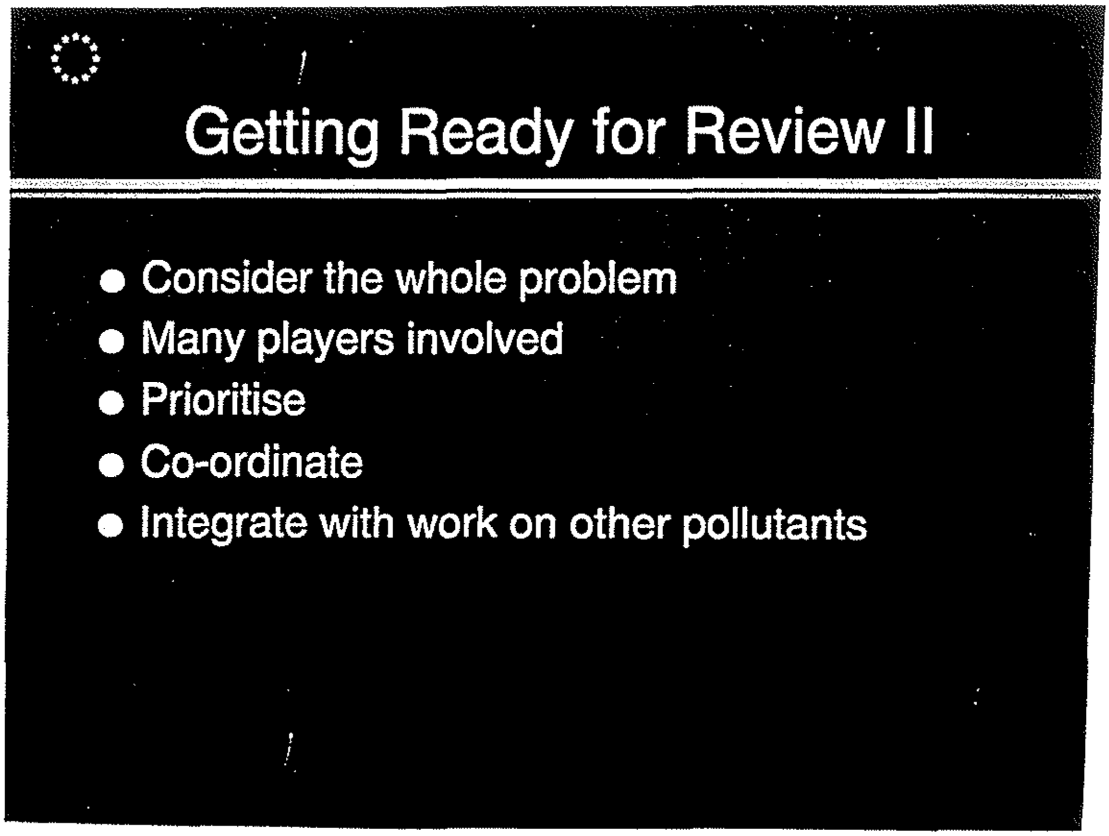
To Be Taken Into Account

- Effects of different fractions
- Progress in measurement techniques
  - concentrations in air
  - deposition
- Technical feasibility
- Experience of applying legislation



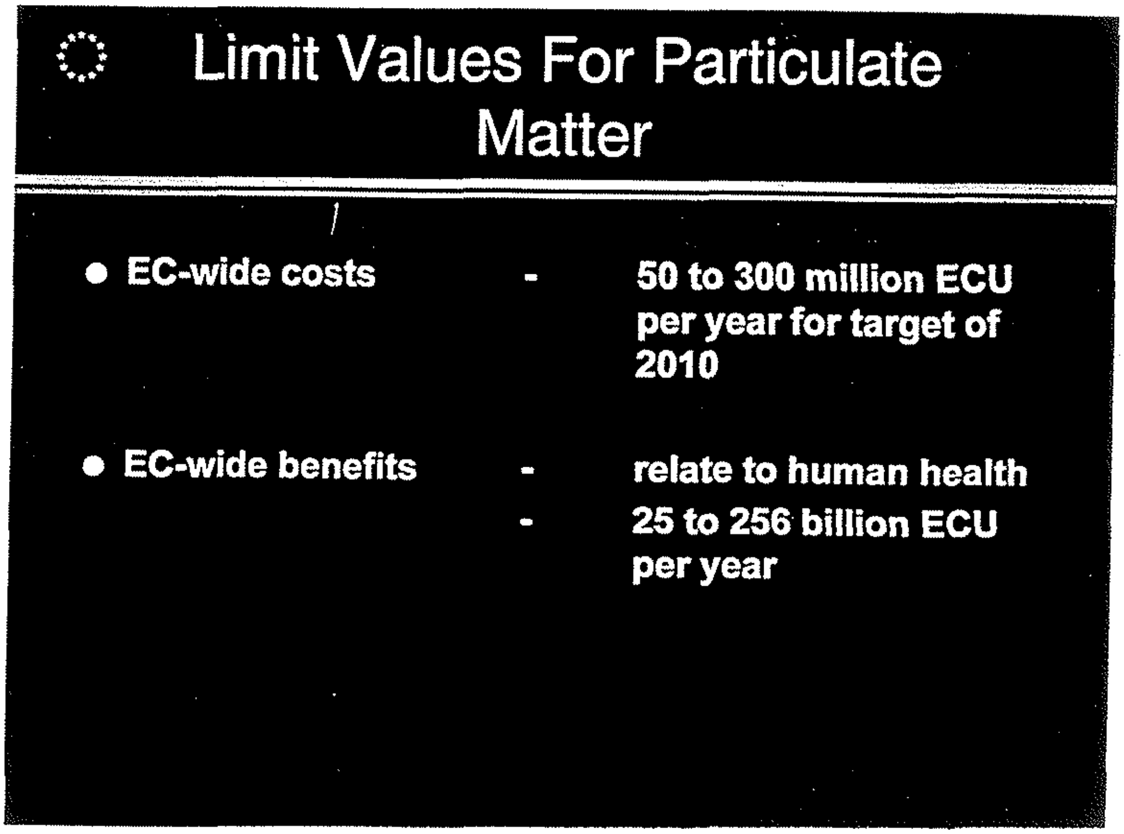
Getting Ready for Review I

- Effects
  - size
  - composition
- Sources
- Chemistry and transport
  - local
  - transboundary
- Measurement



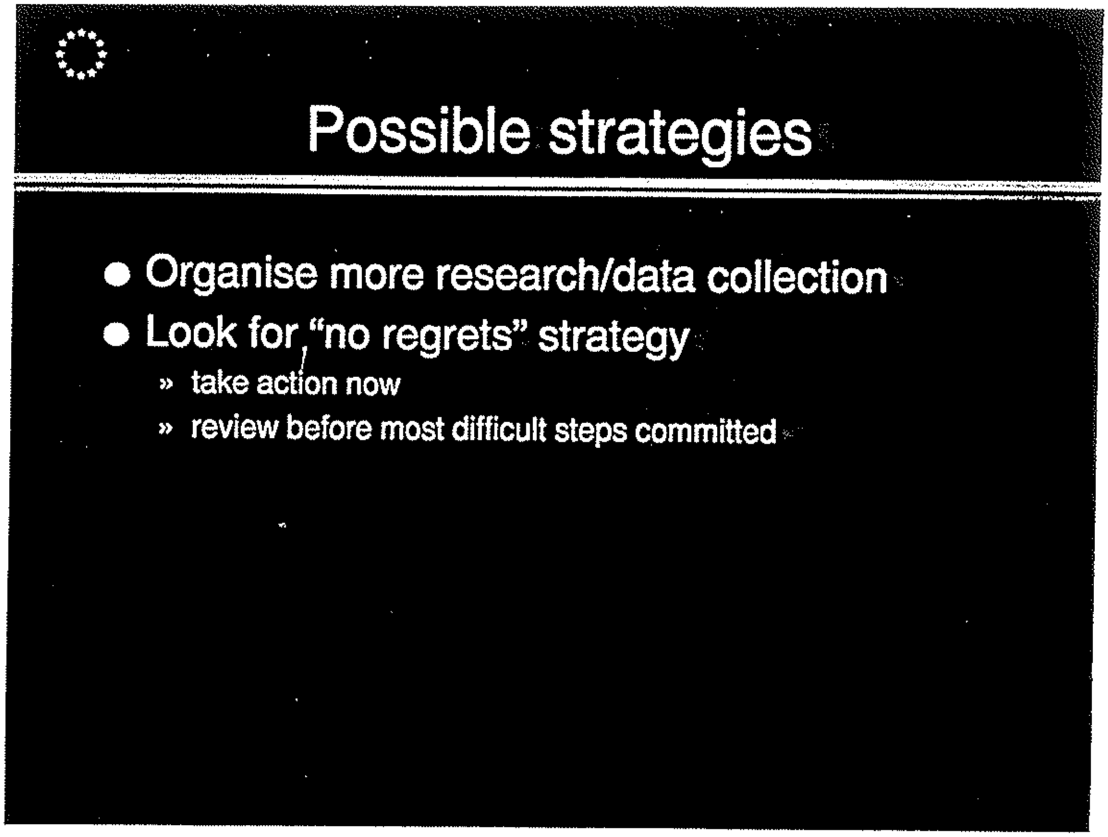
Getting Ready for Review II

- Consider the whole problem
- Many players involved
- Prioritise
- Co-ordinate
- Integrate with work on other pollutants



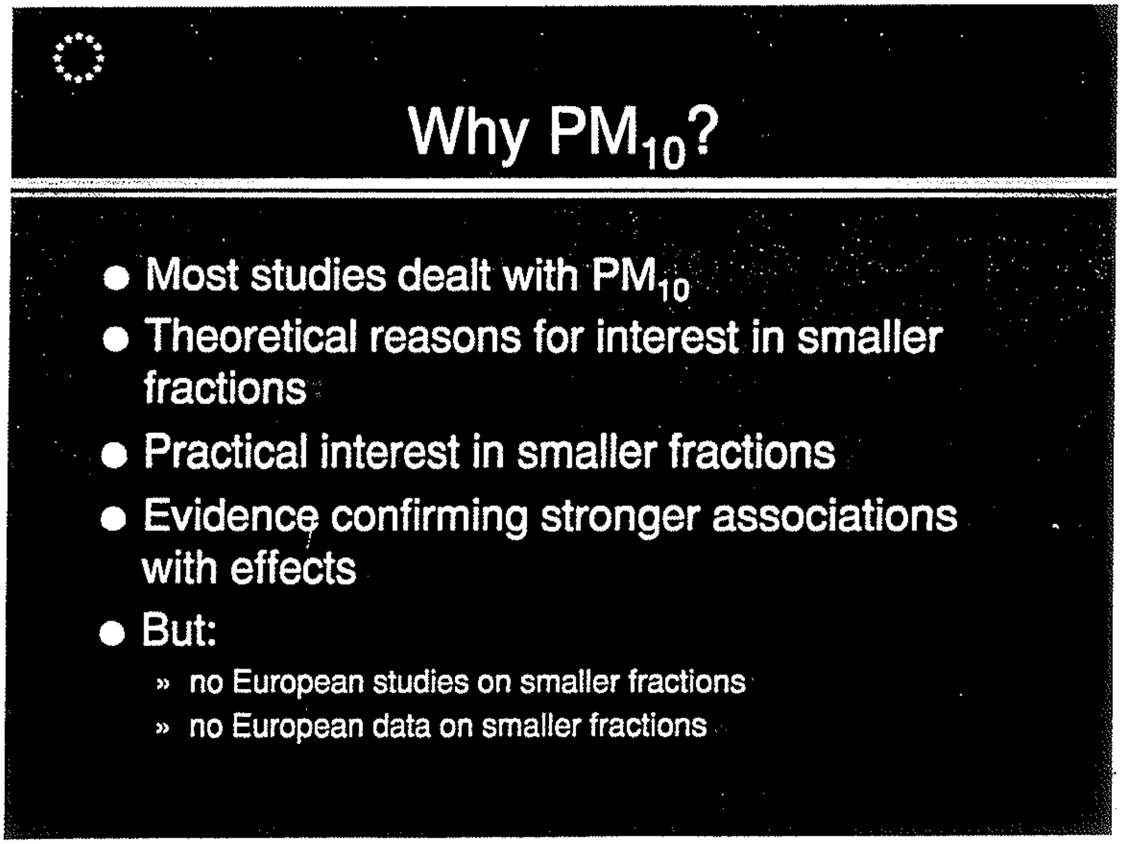
Limit Values For Particulate Matter

- **EC-wide costs** - **50 to 300 million ECU per year for target of 2010**
- **EC-wide benefits** - **relate to human health**
  - **25 to 256 billion ECU per year**



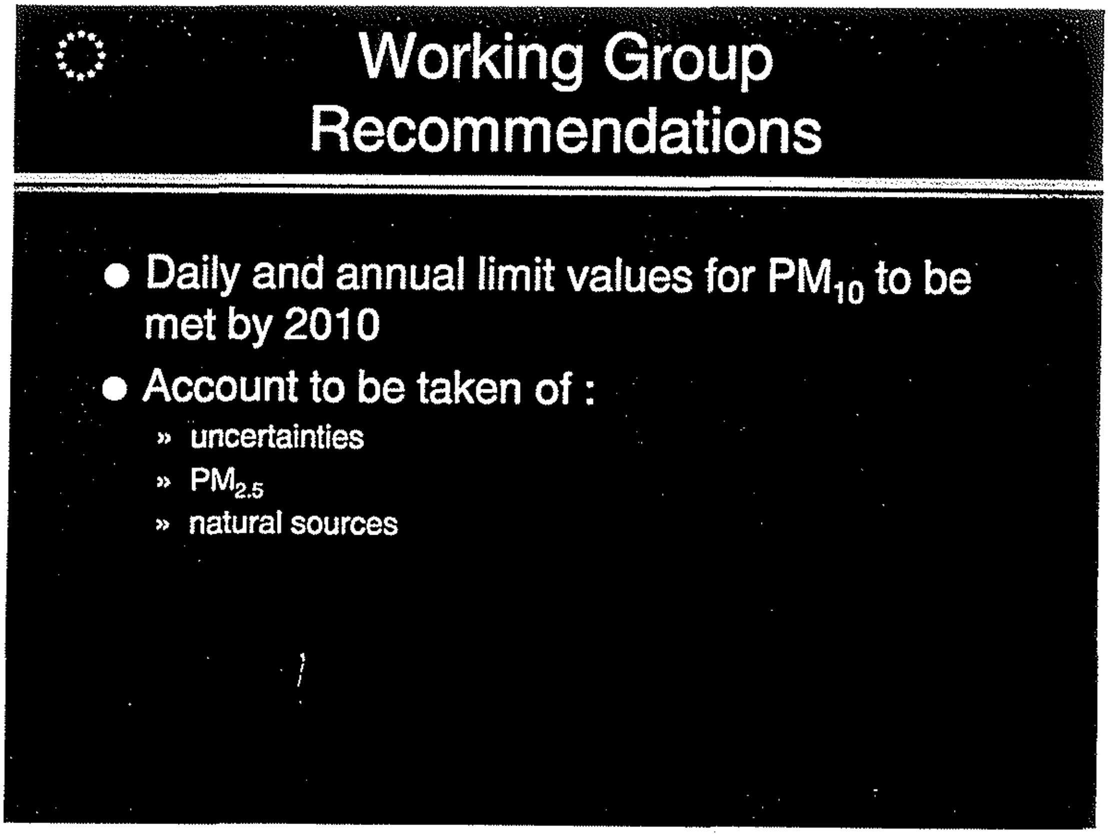
Possible strategies

- Organise more research/data collection
- Look for "no regrets" strategy
  - » take action now
  - » review before most difficult steps committed



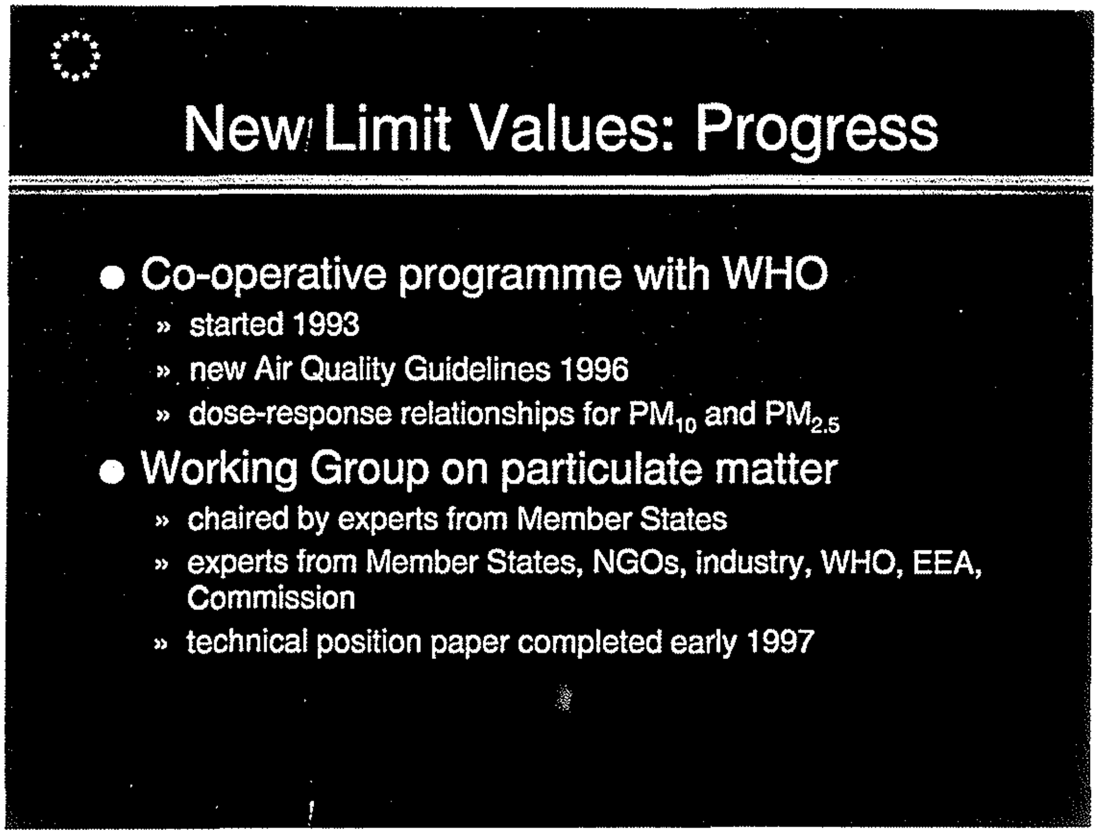
Why PM_{10}?

- Most studies dealt with PM_{10}
- Theoretical reasons for interest in smaller fractions
- Practical interest in smaller fractions
- Evidence confirming stronger associations with effects
- But:
  - » no European studies on smaller fractions
  - » no European data on smaller fractions



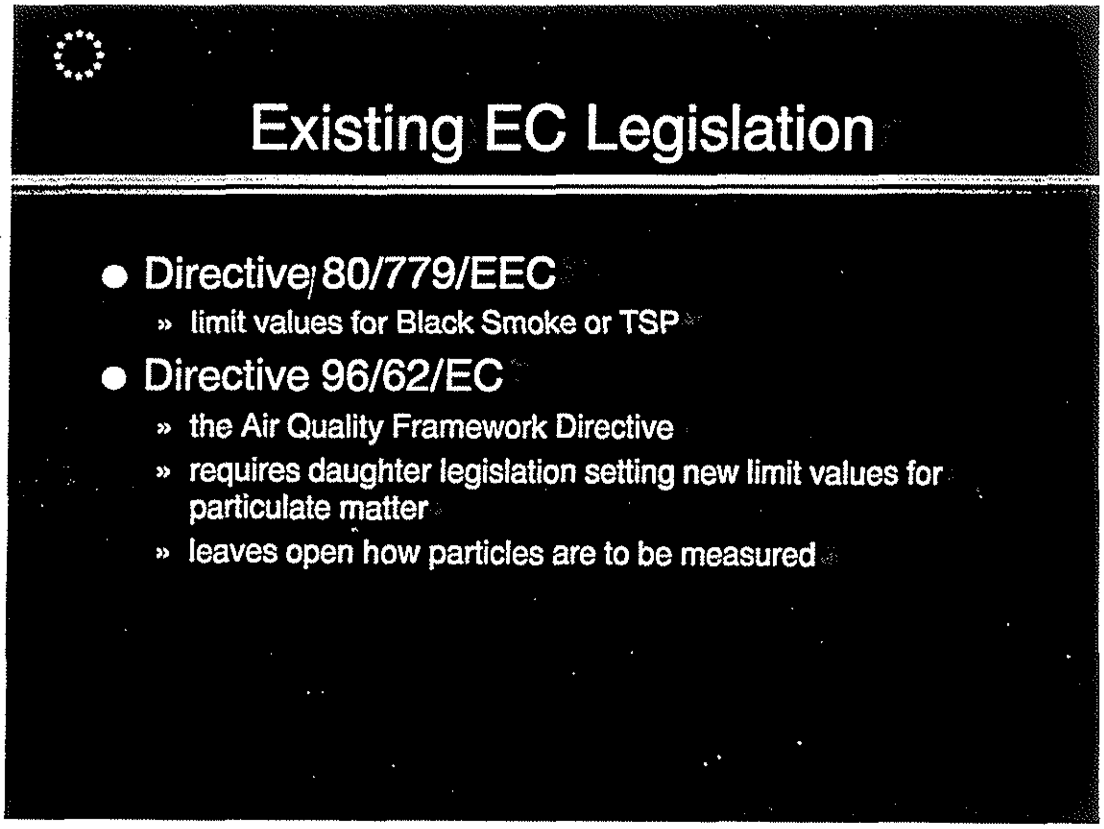
Working Group Recommendations

- Daily and annual limit values for PM_{10} to be met by 2010
- Account to be taken of :
  - » uncertainties
  - » $PM_{2.5}$
  - » natural sources



New Limit Values: Progress

- Co-operative programme with WHO
  - » started 1993
  - » new Air Quality Guidelines 1996
  - » dose-response relationships for PM_{10} and $PM_{2.5}$
- Working Group on particulate matter
  - » chaired by experts from Member States
  - » experts from Member States, NGOs, industry, WHO, EEA, Commission
  - » technical position paper completed early 1997



Existing EC Legislation

- Directive 80/779/EEC
  - » limit values for Black Smoke or TSP
- Directive 96/62/EC
  - » the Air Quality Framework Directive
  - » requires daughter legislation setting new limit values for particulate matter
  - » leaves open how particles are to be measured

Revised U.S. National Ambient Air Quality Standard for Particulate Matter

Bill Harnett
January 14, 1999

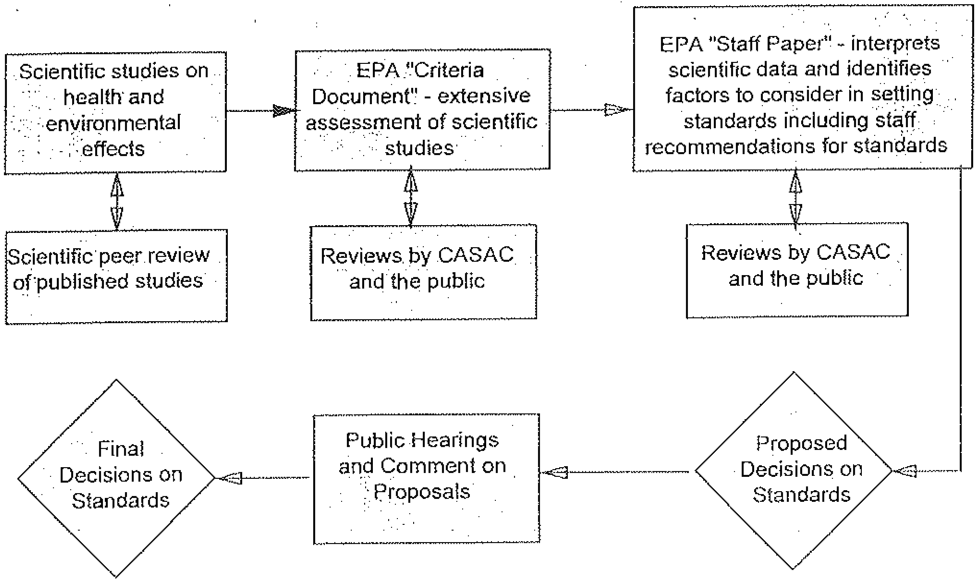
Overview of NAAQS Reviews

- The Clean Air Act calls for national ambient air quality standards (NAAQS) for "criteria" pollutants
  - "Primary" standards protect public health with an adequate margin of safety;
  - "Secondary" standards protect public welfare and the environment (crops, vegetation, wildlife, buildings & national monuments, visibility)
- EPA has set NAAQS for six common air pollutants: ground-level ozone (smog), particulate matter, carbon monoxide, lead, nitrogen dioxide, sulfur dioxide
- The Act requires EPA to review the scientific criteria and these standards at least once every five years, with advice from the Clean Air Scientific Advisory Committee (CASAC)

Different Considerations Used in Setting and Achieving NAAQS

Setting the Standards	*Achieving the Standards*
• Health Effects	• Costs
• Environmental Effects	• Time to attain the standards

Review Process for NAAQS



Scientific studies on health and environmental effects

Scientific peer review of published studies

EPA "Criteria Document" - extensive assessment of scientific studies

Reviews by CASAC and the public

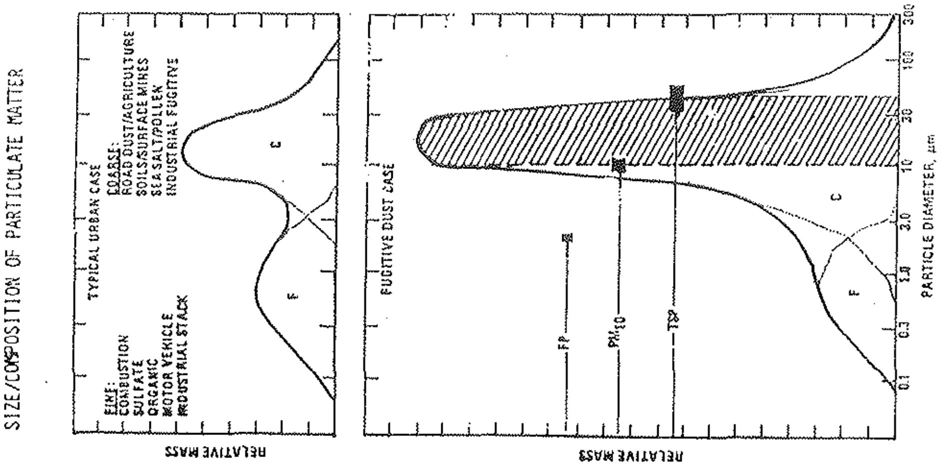
EPA "Staff Paper" - interprets scientific data and identifies factors to consider in setting standards including staff recommendations for standards

Reviews by CASAC and the public

Proposed Decisions on Standards

Public Hearings and Comment on Proposals

Final Decisions on Standards


SIZE/COMPOSITION OF PARTICULATE MATTER
TYPICAL URBAN CASE
FINE:
COMBUSTION
SULFATE
ORGANIC
MOTOR VEHICLE
INDUSTRIAL STACK
COARSE:
ROAD DUST/AGRICULTURE
SOILS/SURFACE MINES
SEA SALT/POLLEN
INDUSTRIAL FUGITIVE
F
C
RELATIVE MASS
FUGITIVE DUST CASE
FP
PM10
TSP
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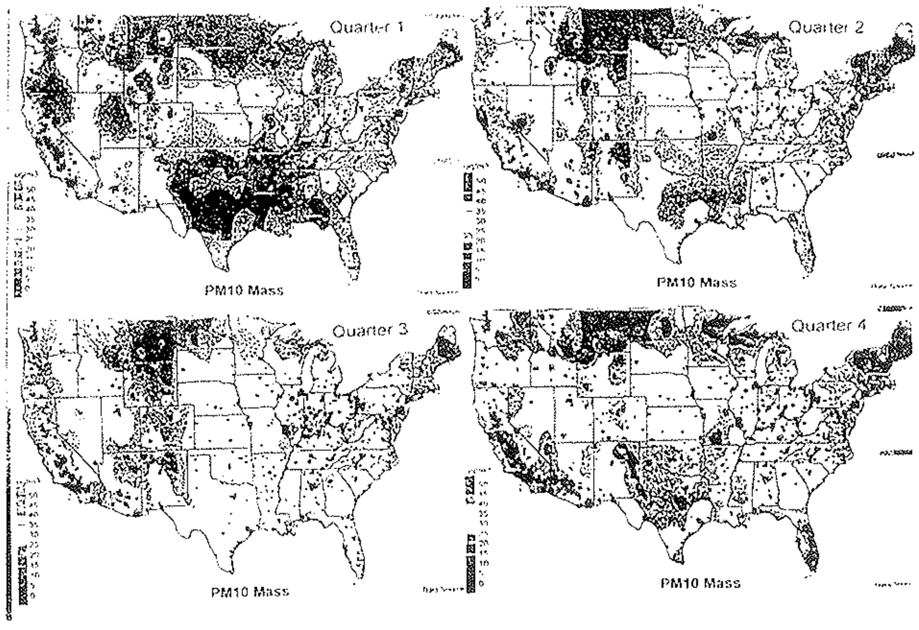
C
RELATIVE MASS
0.1
0.3
1.0
3.0
10
30
100
300
PARTICLE DIAMETER, µm


Quarter 1
PM10 Mass
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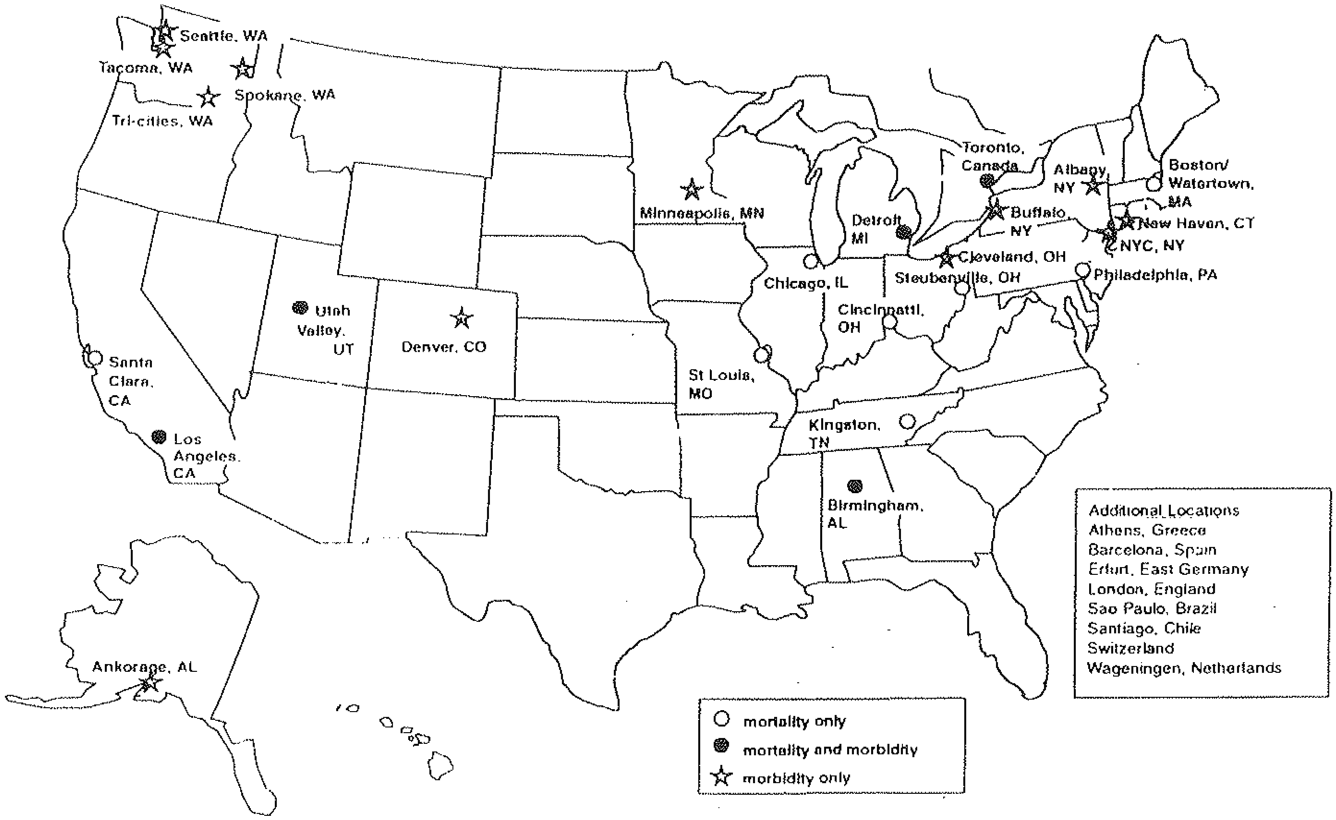
PM10 Mass
Quarter 3
PM10 Mass
Quarter 4
PM10 Mass

Locations of Recent PM Health Studies



Scientific Basis for Revisions to Particulate Matter Standards

- Extensive review of thousands of scientific studies highlighted over 80 key epidemiological studies as basis for revisions
- Over 60 epidemiological studies found significant links between particulate matter levels at or below the previous standards and premature death or serious illness
- Numerous studies indicate "fine" and "coarse" particles are fundamentally different pollutants
- 19 of 21 CASAC panel members recommended revising the PM10 standards by adding standards for fine particles (those smaller than 2.5 micrometers, or PM2.5)
- CASAC unanimously recommended retaining at least the annual PM10 standard to continue to address health concerns associated with coarse fraction particles (those with diameters between 2.5 and 10 micrometers)

Particulate Matter-Related Health Effects of Concern

- Increased premature deaths, primarily in the elderly and those with heart or lung disease
- Aggravation of respiratory and cardiovascular illness, leading to hospitalizations and emergency room visits in children and individuals with heart or lung disease
- Lung function decrements and symptomatic effects such as those associated with chronic bronchitis, particularly in children and asthmatics
- Increased work loss days and school absences
- Changes to lung structure and natural defense mechanisms

Revisions to Particulate Matter Standards

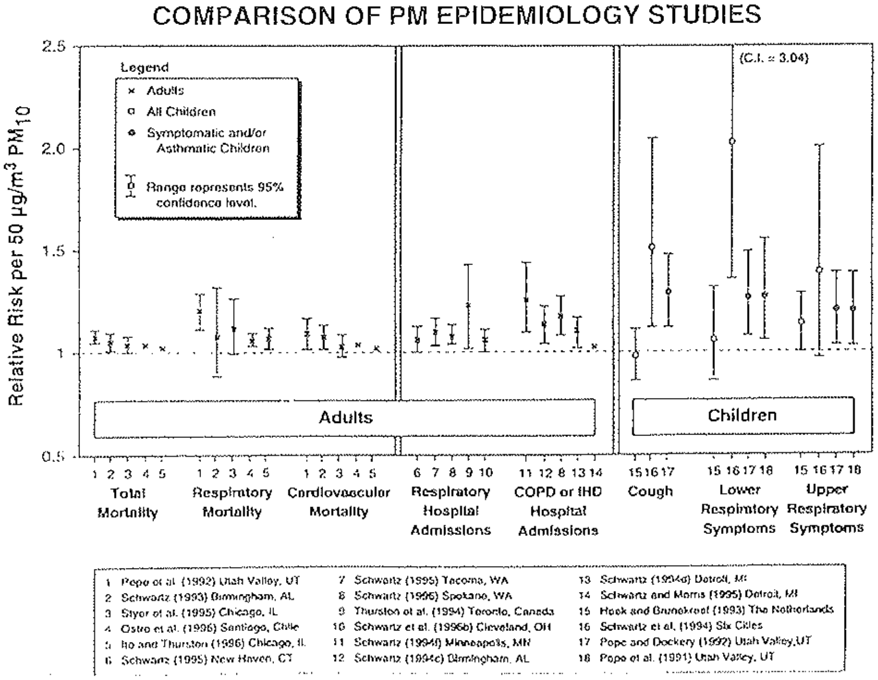
- Previous PM_{10} standards:
  - 50 ug/m3, annual arithmetic mean (3 year average)
  - 150 ug/m3, 24-hour average, 1 expected exceedance/year (3 year average)
- New $PM_{2.5}$ standards:
  - 15 ug/m3, annual arithmetic mean, spatial average of designated monitors (averaged over 3 years)
  - 65 ug/m3, 24-hour average, 98th percentile concentration (averaged over 3 years), maximum monitor in an area
    - Forms generally consistent with some CASAC panel members' recommendations for more stable forms
- Revised PM_{10} standards:
  - Retain current annual standard
  - Retain level of 24-hour standard but revise form to 99th percentile concentration (3 year average)

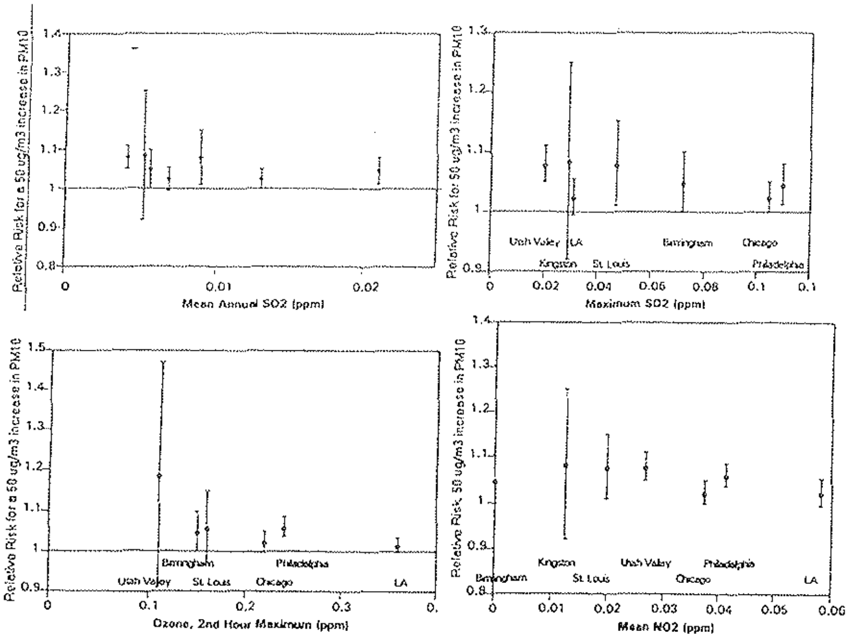
Increased Health Protection from New and Revised Particulate Matter Standards

- Tens of thousands fewer premature deaths per year, especially in the elderly and those with heart and lung disease
- Thousands of fewer respiratory-related hospital admissions per year
- Hundreds of thousands fewer incidences each year of aggravated asthma and respiratory symptoms
- Tens of thousands fewer cases each year of chronic bronchitis
- Reduced risks of more frequent childhood illnesses, which are of concern both in the short term as well as for the future development of healthy lungs in affected children

Considerations in Selecting Final PM Standards

- Fine and coarse fraction particles considered as separate pollutants, both of which are related to adverse health effects
- The consistency and coherence of the evidence of associations between PM at concentrations below the previous standards and serious health effects
- Community studies showing associations between health effects and both short- and long-term exposures to PM and analyses supporting a focus on the annual standard as the predominant standard with the 24-hour standard providing supplemental protection
- Analyses of key epidemiology studies linking health effects to measured fine particle concentrations in the study cities





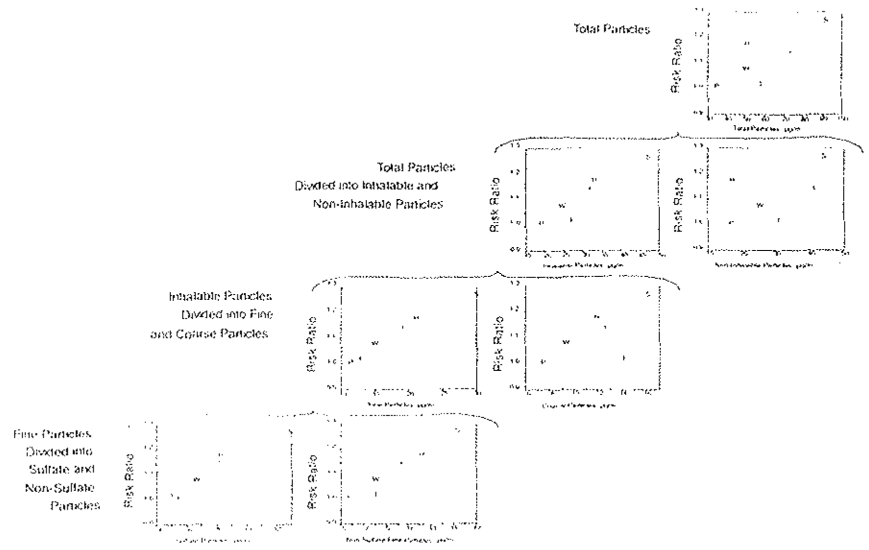
10/31/95 TUE 12:28 FAX 202 401 6537

To John Bachmann

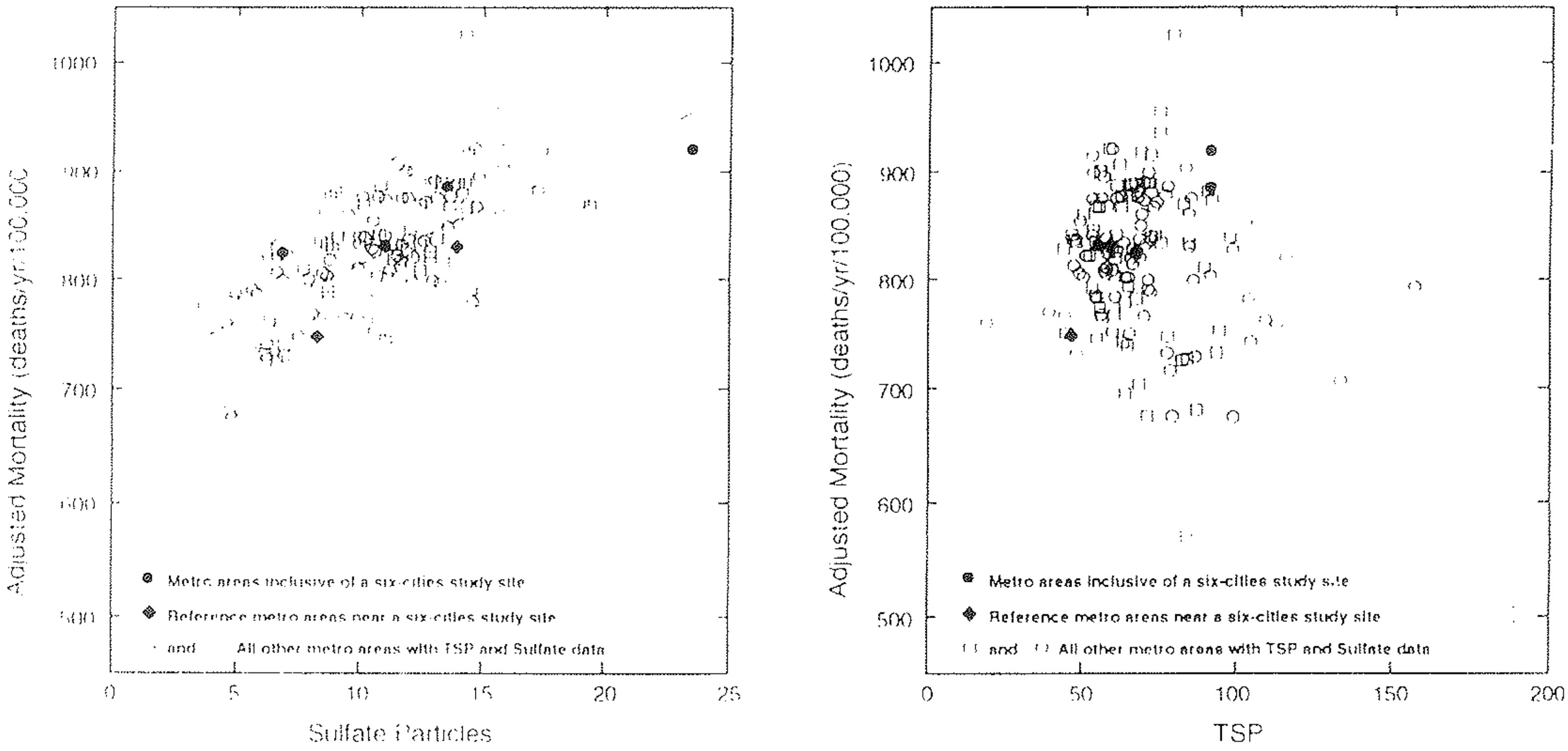
fr: Tracey

@001

6 City Long-Term Mortality vs PM Fractions



Long-Term Mortality vs. Fine Sulfates and TSP



Pope, 1995: ACS Study Data

Fine and Coarse Particles are Separate Pollutants

- Different sources
  - Coarse fraction mechanically generated from crushing and grinding
  - Fine particles formed secondarily and from combustion sources
- Fundamentally different chemistry
- Differences in size, aggregate number and surface area
- Different exposure characteristics

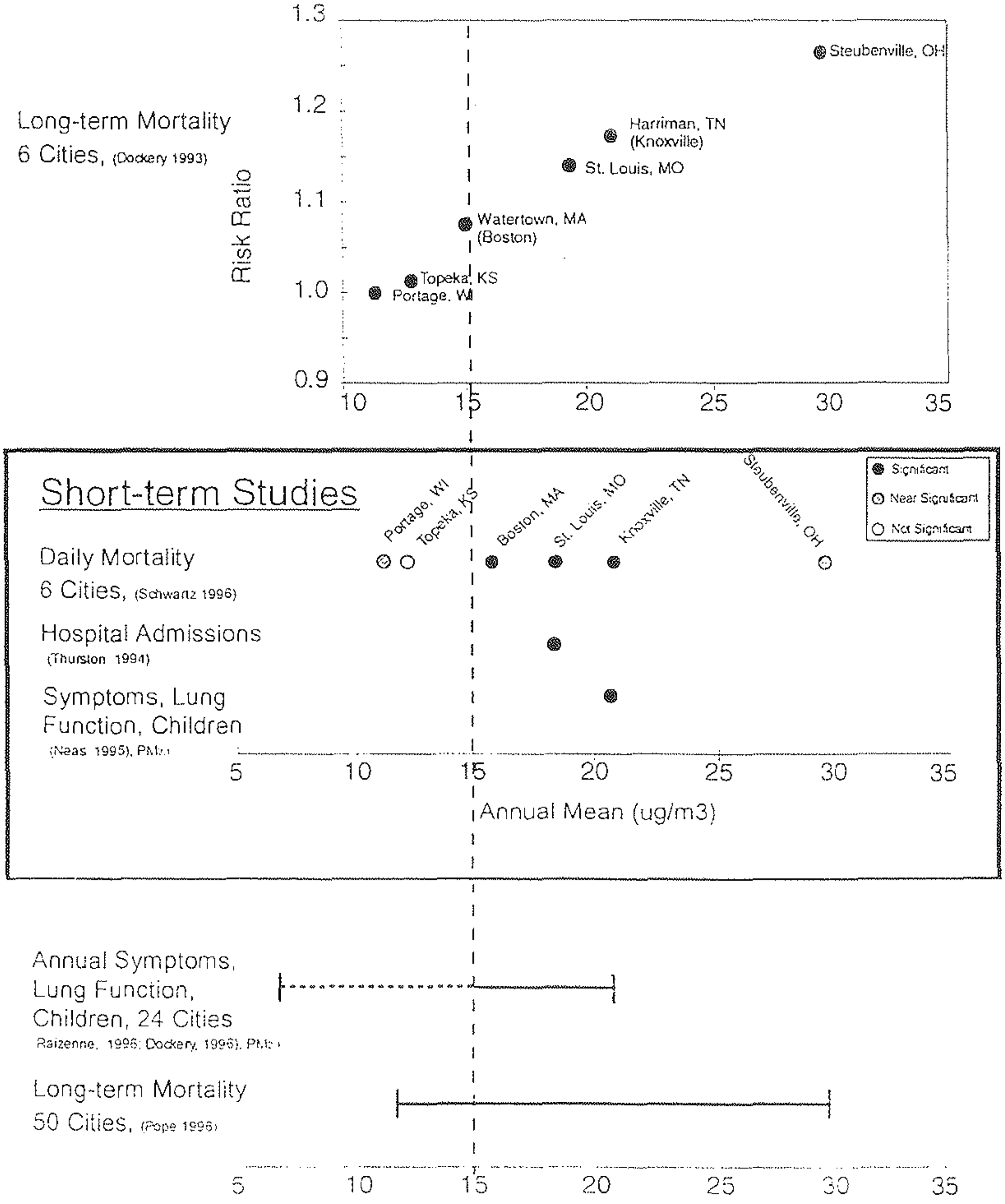
Focus on Annual Standard for Major Risk Reduction

- EPA considered the combined effects of standards on reducing risk of both short- and long-term exposures
  - Daily PM studies usually involve 1 to 8 years of aggregated daily data on relationships between community-wide exposures and effects
  - Most of the aggregate risk stems from mid- to lower range concentrations due to widespread potential exposures (more people experiencing days with lower concentrations)
- Annual standard can provide a stable target for protection against daily and longer term risk
- 24-hour standard still needed for locations with high daily or seasonal levels that would not be controlled by annual standard

Community Studies: Consistent and Coherent Effects Evidence

- Striking consistency in quantitative results of studies across multiple communities
  - Similar relative risk for PM despite differences in weather, presence of other pollutants, demographics and lifestyles between locations.
  - PM is most consistently linked to health effects -- Relative risks are consistent with high or low SO2, O3, CO, NOx concentrations
- Similar sensitive groups in multiple study locations, multiple effects observed in single locations (e.g., mortality and hospital admissions)
- Qualitative coherence in sensitive populations and in nature of effects enhances biological plausibility (e.g., the elderly and those with heart and lung disease with cause of death or reason for hospital admission relating to cardiopulmonary disease)

PM$_{2.5}$ Studies - Basis of Annual PM$_{2.5}$ Standard

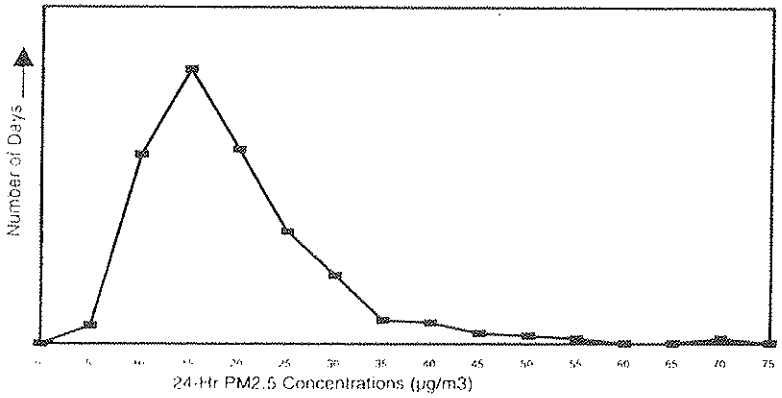


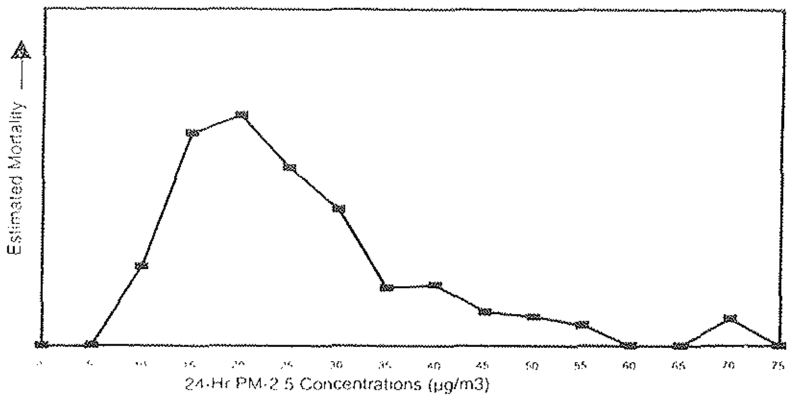
Basis for Annual $PM_{2.5}$ Level/Form

- Decisions on final level placed greater weight on studies reporting associations between health effects and direct measures of fine particles
  - No thresholds demonstrated, but uncertainties increase markedly at lower concentrations
  - Significant daily mortality/morbidity associations in cities with annual means from about 16 to 21 ug/m3
  - Significant long-term mortality associations in 2 studies with annual means of about 18 - 22 ug/m3; staff paper concluded evidence for increased risk was more apparent at annual means above 15 ug/m3

- Spatial average consistent with data from community studies used in selecting level and with the atmospheric behavior of fine particles

A. Illustrative Air Quality Distribution: 24-Hr PM Values





B. Estimated Mortality Risks Associated With Air Quality Distribution In A

Basis for 24-hour $PM_{2.5}$ Standard Level/Form

- Daily standard intended to provide supplemental protection against high exposures for short time periods or local peaks
- concentration-based percentile form
    - Risk more related to concentration than number of exceedences
    - Avoids complex data handling for missing data and less-than -every-day monitoring, more stable target for control programs
- Level of daily standard selected based on protection afforded by combination of both annual and daily standards
- Highest monitored value in an area consistent with need to protect against localized peaks

Coarse Fraction Particles Still of Health Concern

- Coarse fraction particles reach sensitive areas of the lung

- Health effects of concern
  - Aggravation of asthma
  - Increased respiratory illness
  - Children are particularly sensitive
  - Concerns about long-term accumulation

- Best evidence is from studies with higher concentrations

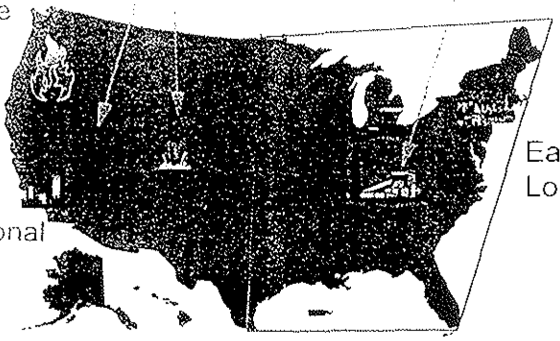
PM10 used to regulate coarse fraction particles

- Unanimous agreement by CASAC for continued coarse fraction particle control
  - Retain annual PM_{10} at 50 ug/m3
  - Keep daily PM_{10} at least as an option

- Many CASAC panelists suggested a more stable form for the daily PM_{10} standard

- 99th percentile form, averaged over 3 years
  - Avoids complex data manipulation

Fine PM Strategy Considerations



Isolated Woodsmoke

Isolated Urban FP Mix

Eastern Regional Summer SOx, FP

Eastern Urban, Local Scale FP

Urban Regional NOx, FP

MARTIN WILLIAMS

Department of Environment, United Kingdom

PARTICLE FORMATION AND CHARACTERISATION

by

Dr Martin L Williams

DETR, UK

This paper does not aspire to present a detailed scientific review of the formation and characterisation of particles, but rather presents the salient features of the subject appropriate to the context of the formulation of European air quality legislation. The paper will discuss different metrics for particle measurement and the different source contributions to PM_{10}. It will also discuss the implications for legislation and policy formulation of recent scientific advances in these areas.

The measurement of an air pollutant is a fundamental part of any regulatory system and it is at this basic level that problems arise for particles. This is because any atmospheric distribution of particles, unlike a gas such as sulphur dioxide, will be different from one location to another, and from one time to another at the same location. Any standard reference method will therefore necessarily be to some extent arbitrary and will depend on agreed sampling and analytical conventions. In Europe the position is that CEN, the European organisation responsible for formulating standard reference measurement methods, has recently agreed a standard for PM_{10}, but has only recently begun work on a method for $PM_{2.5}$. The existence of an agreed reference method is a crucial step in the development of air quality legislation, and progress in this area will be important in the period before the recently agreed Daughter Directive is reviewed in 2003.

The Daughter Directive on air quality set limit values for PM_{10}, and it is worth considering the various sources of PM_{10} concentrations.

Atmospheric levels of PM_{10} can be considered as being composed of three main categories. *Primary* particles are emitted directly by combustion processes, and are generally less than 2.5µm and often less than 1µm. *Secondary* particles are those formed in the atmosphere from chemical reaction and include sulphates and nitrates formed from the reactions of emissions of SO_2 and NO_x, and organic aerosols formed from reactions of VOCs; these are also generally less than 2.5µm, but the size could vary depending on humidity. The third category are the so-called *coarse* particles which are formed from a variety of non-combustion sources. These include natural events such as wind-blown dusts and soils, forest and other natural fires, and sources generated by human activities such as resuspended road dust and tyre debris, and construction and mining/quarrying activity. The particles generated by these sources mostly arise from mechanical attrition and are thus relatively large, that is they are generally greater than 2.5µm.

This categorisation of the sources of atmospheric particles is convenient from an air quality management point of view in that the *emission* source categories of the primary, secondary and coarse particle fractions are, generally different. There will clearly be exceptions, but very broadly, in a given urban area, the primary particles will arise from combustion sources in and around the urban area with a relatively small degree of long range transport from more distant areas; the secondary particles could however have a significant proportion of long-range transported material since the particle size and deposition velocities of sulphate and nitrate particles are small and their atmospheric lifetimes are long. The lifetimes of the coarse particles are short and their atmospheric transport distances are also short, except in some specific climatic conditions, especially in southern Europe. Coarse particle concentrations in an urban area will therefore normally arise predominantly from within that area.

In designing a rational control policy for atmospheric particles, it is important to attempt to quantify as far as possible the contributions of the different source categories to atmospheric levels. This is not a straightforward task, but some significant progress has been made in the last year or so in the UK by the Airborne Particles Expert Group. This has used receptor modelling involving a regression technique for apportioning the measured PM_{10} data on independent variables of black smoke or NO_x (as a surrogate for primary particles) and sulphate concentrations (as a surrogate for secondary particles). The 'constant' term in the linear regression represents the other source categories which are considered to be principally the coarse fraction of the PM_{10} referred to above. The method has been described more fully in the report of the Group. As a very broad rule of thumb, the method suggests that annual mean PM_{10} concentrations in UK urban areas are composed of roughly equal proportions of primary, secondary and coarse particles. The proportions will of course vary day by day, and even hour by hour.

These results have a significant implication for the formulation of legislation to control PM_{10} (and $PM_{2.5}$) concentrations. The formation mechanisms and lifetimes of the secondary particles means that they are essentially transboundary in nature, in the same way as ozone is in most areas of Europe. The extent to which secondary particles contribute to ambient levels of PM_{10} and $PM_{2.5}$ over the whole of Europe has still to be determined, but given the size of the modelled contribution to some large urban areas in the UK, the transboundary contribution to most other urban areas in Europe is likely to be significant.

Chemical analyses of the various fractions of PM_{10} have been carried out, and some typical results for the UK show that as long terms averages the fine fraction (less than 2.5 microns) is composed of roughly equal proportions of carbonaceous matter (primary combustion emissions) and secondary particles composed mostly of ammonium sulphate and nitrate aerosol. The coarse fraction (between 2.5 and 10 microns) is composed mostly of insoluble minerals (wind-blown dusts etc.) with smaller contributions from sea-salts and some primary and secondary aerosols. From a legislative viewpoint the significance of these results is that the major part of the coarse fraction does not arise from man-made combustion emissions and is inherently difficult, if not impossible to control. Virtually all of the $PM_{2.5}$ fraction arises from anthropogenic emissions which are controllable. It is likely that the broad features of the chemical composition of these fractions are similar throughout Europe, although more analyses are required. The results discussed above for the UK are broadly similar to results reported from the USA, with the slight difference that in some areas, notably Los Angeles, the aerosol contains proportionally more nitrate than sulphate than do European aerosols. This may well change in future as European sulphur emissions are decreasing faster than those of NO_x.

There is currently debate over the most appropriate metric for particle measurement and legislation. The primary determinant in this decision must be the health effect evidence, and this will be discussed elsewhere in the symposium. However, some comments on possible metrics other than PM_{10} are worth noting here. In particular it has been suggested that the number of particles is more important than their mass concentration, but there are as yet few studies of health effects to substantiate this. Measurement data on particle number concentrations are emerging, notably in Finland and the UK, and what is clear from these studies is that particle number concentration differences between roadside, urban background and rural monitoring sites are much larger than mass measures such as PM_{10} or even $PM_{2.5}$. Particle number appears to be a much more sensitive indicator of primary, particularly vehicle, emissions than the commonly used mass measures.

Surface area has also been suggested as a possible metric of particles most closely associated with health effects. It is too early to come to definitive conclusions on these issues, but it is worth noting from a regulatory point of view that the different metrics pose quite different policy problems. Looking at the atmospheric particle size distribution, and bearing in mind that the surface area of a particle is proportional to the square of the radius and that the volume, or mass, is proportional to the radius cubed, it is clear that information on *number* concentration is carried by the ultrafine particles (those less than about 0.1 microns) which are likely to be overwhelmingly primary particles emitted within urban areas, the *surface area* information is carried by the accumulation mode (between about 0.1 and 1 microns) which will contain significant amounts of secondary-and hence transboundary-particles. The *mass* information is carried by both the accumulation mode and the coarse mode particles, the latter being those larger than about 2.5 microns, and which contain significant amounts of uncontrollable natural components.

At the present time there is emerging evidence that the $PM_{2.5}$ fraction is the more important for public health, largely on the basis of studies carried out in the USA which lead to the USEPA promulgating an air quality standard for $PM_{2.5}$. $PM_{2.5}$ should penetrate further into the respiratory system, so there are reasons of principle for considering $PM_{2.5}$ to be more appropriate than PM_{10}, but there are two important points to note. Firstly, there are few European studies of the heath effects of $PM_{2.5}$, although there are several under way. Secondly, even if a body of evidence does build up demonstrating associations between health effects and $PM_{2.5}$, there is still the existing body of evidence on PM_{10} which cannot be ignored, and the question of the toxicity of the coarse fraction will need to be addressed.

There is also at present little information on ambient air concentrations of $PM_{2.5}$ in Europe, although now that the recent air quality Daughter Directive requires Member States to make such measurements a substantial body of data should emerge by the time that Directive is reviewed in 2003. On the basis of the currently available data, it appears that $PM_{2.5}$ and PM_{10} are closely correlated in urban areas, and that, very approximately $PM_{2.5}$ is about 0.6 times PM_{10}. Roadside levels of $PM_{2.5}$ tend to be enhanced proportionally more than those of $PM_{2.5}$ because of the contribution of resuspended roadside dust. It has already been noted that a major fraction of $PM_{2.5}$ is man-made, but it should further be noted that the *controllable* fraction of both $PM_{2.5}$ *and* PM_{10} are essentially the same. (The exception here of course is that the resuspended road dust which contributes to PM_{10} but not to $PM_{2.5}$ is potentially controllable through reductions in traffic volumes).

There are already policies and agreements in place which will reduce the concentrations of PM_{10} and $PM_{2.5}$ in Europe in the future. Motor vehicle emission limits and fuel quality standards have been agreed which will result in substantial reductions of primary particle emissions from diesel vehicles. Agreements in the UNECE on the Second Sulphur Protocol under the Convention on Long Range Transboundary Air Pollution will reduce sulphur emissions, and further reductions together with reductions in emissions of NO_x, VOCs and ammonia will further reduce the concentrations of secondary particles in Europe. Further agreements in the EU are likely via the National Emissions Ceilings Directive, a proposal for which is expected in 1999.

PARTICLE FORMATION AND CHARACTERIZATION

by

Dr Martin Williams

DETR, UK

14-15 January 1999

IN AN IDEAL WORLD WE WOULD KNOW :-

- Nature and extent of effects-risk
- Dose/exposure-response
- Mechanism of effects
- Exposures/ambient levels
- Sources of harmful concentrations
- Benefits of abatement
- Costs

MEASUREMENTS OF PM

- Policy is ahead of the science here
- There are good reasons for a standard for $PM_{2.5}$ on health grounds
- But we have very little data in Europe
- We don't have a reference method

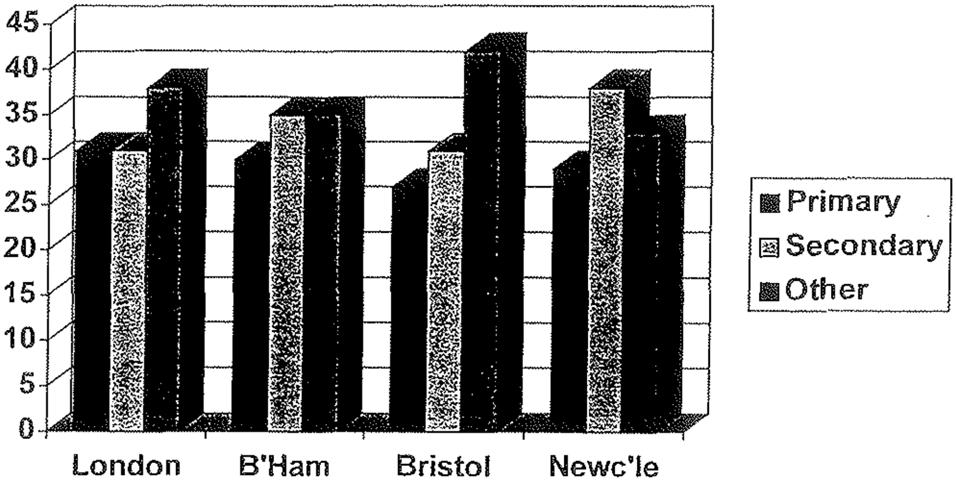
MEASUREMENT METHODS

- There is no absolute standard for particles
- Samples of particles are not unique like gaseous pollutants
- Measurement standards will therefore always be arbitrary to some degree

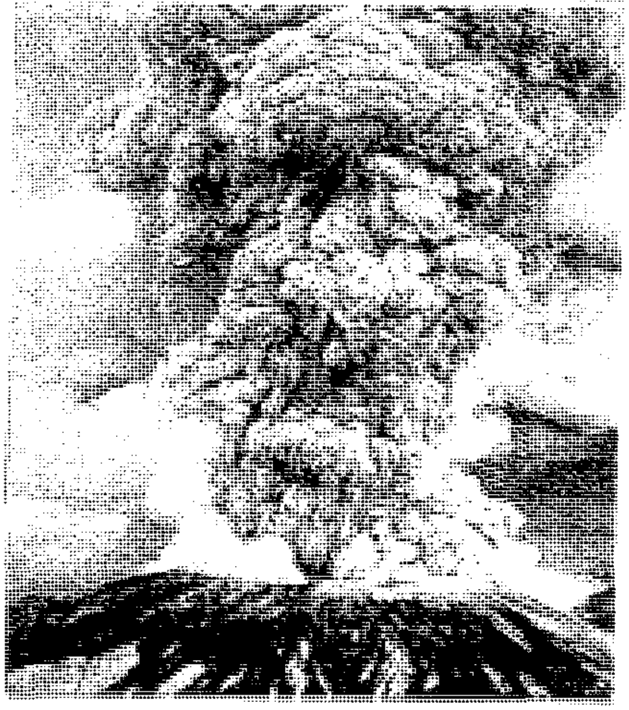
Primary and Secondary Particles

- Primary particles are *emitted* directly to the atmosphere
- Secondary particles are *formed* in the atmosphere by homogeneous and heterogeneous chemical reactions

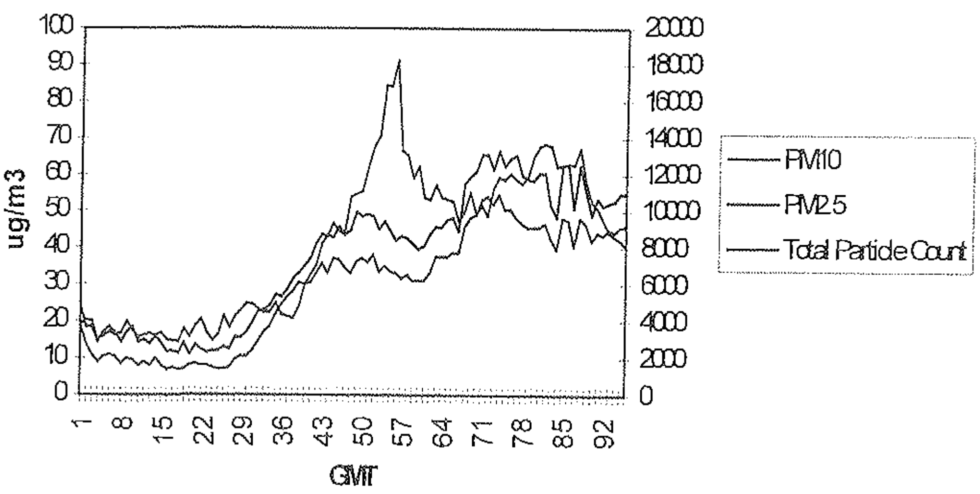
SOURCE APPORTIONMENT OF PM_{10} (% of Annual Mean) in UK



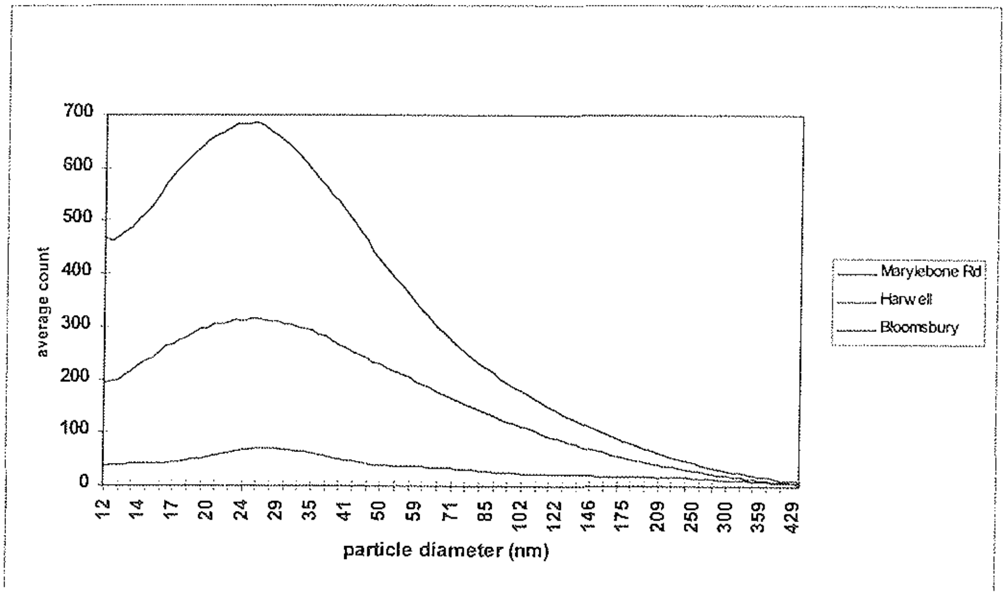
NATURAL SOURCES OF PM



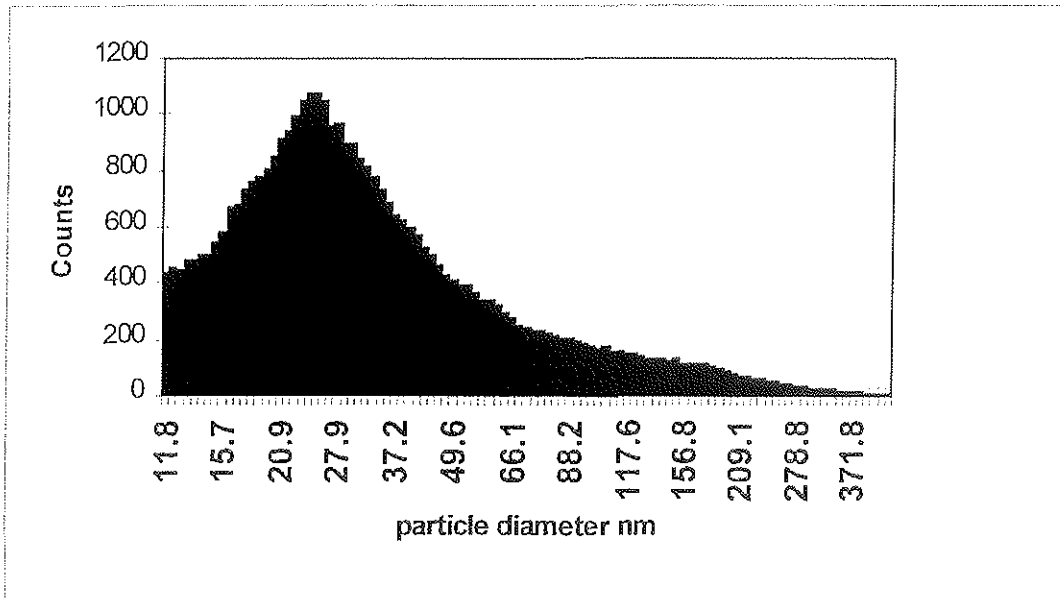
Particle Number, PM10 & PM2.5


Particle Mass vs Numbers - Harwell 13th May 1998

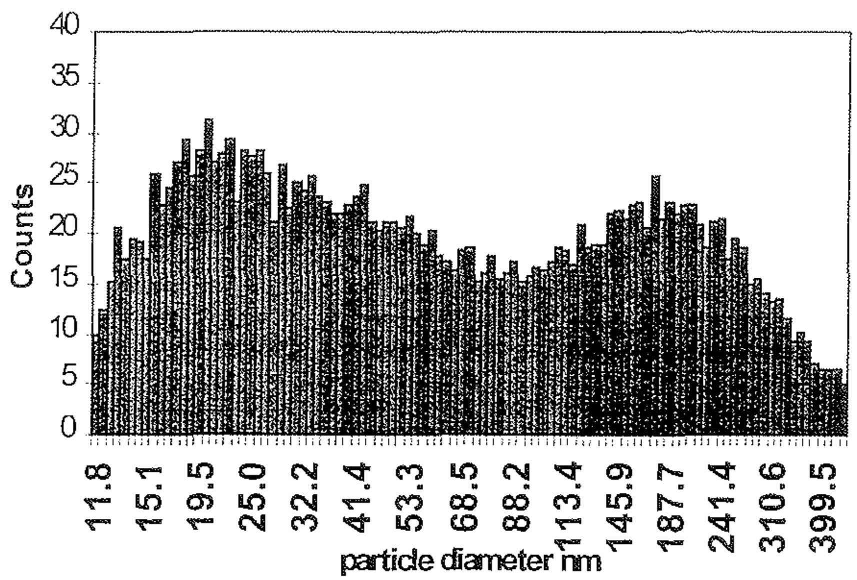
Number Count at Roadside, Urban Background and Rural Sites



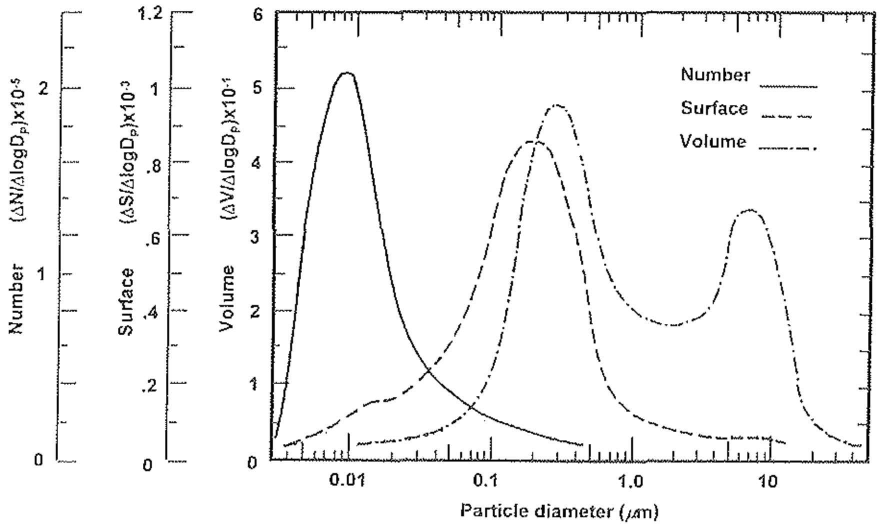
Particle Counts-Roadside site



Particle Counts-Rural Site



Size of urban particulate matter by category (μm)



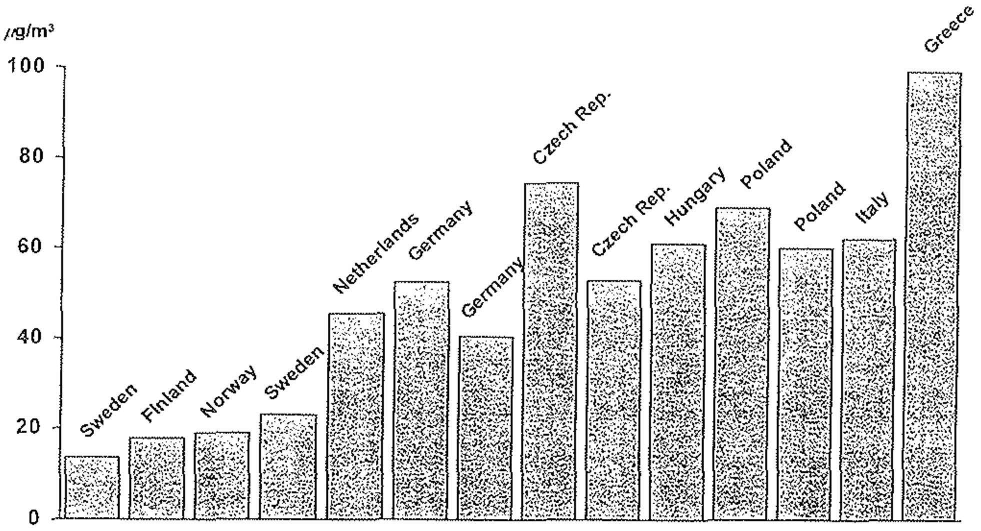
Number, Area or Mass ?

- **Number information is carried by the *Ultrafines***
- **Area information is carried in the *accumulation* mode**
- **Mass information is carried in the *accumulation and coarse* modes**

TRENDS IN PM10

- **Limited by lack of data**
- **Probably downward over last two decades**
- ***Future* trends are more important**

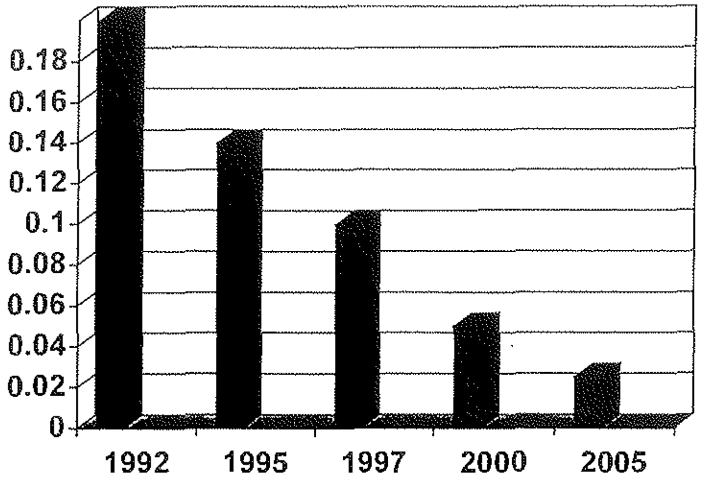
Winter mean PM_{10} levels 1993/94 in PEACE Study ($\mu g/m^3$)



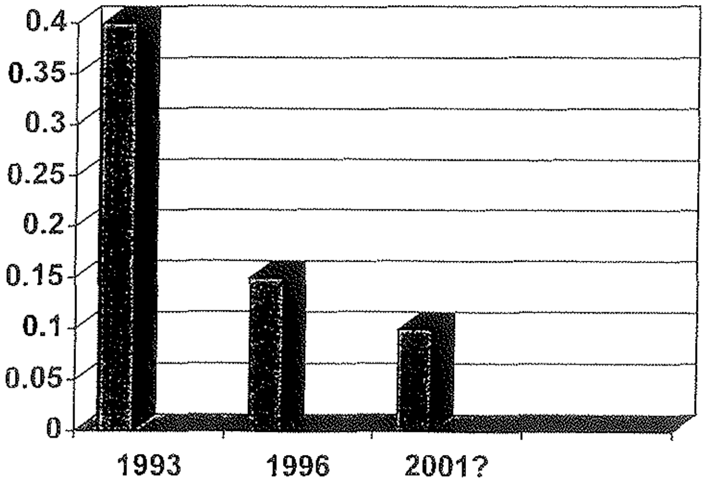
What Rules Connect Emissions and Concentrations of PM ?

- *Primary* particles should respond linearly (in mass metric) to mass emissions
- *Secondary* particles *should* also be linear, but this needs more work
- As precursors in Europe reduce, more distant and natural sources become significant

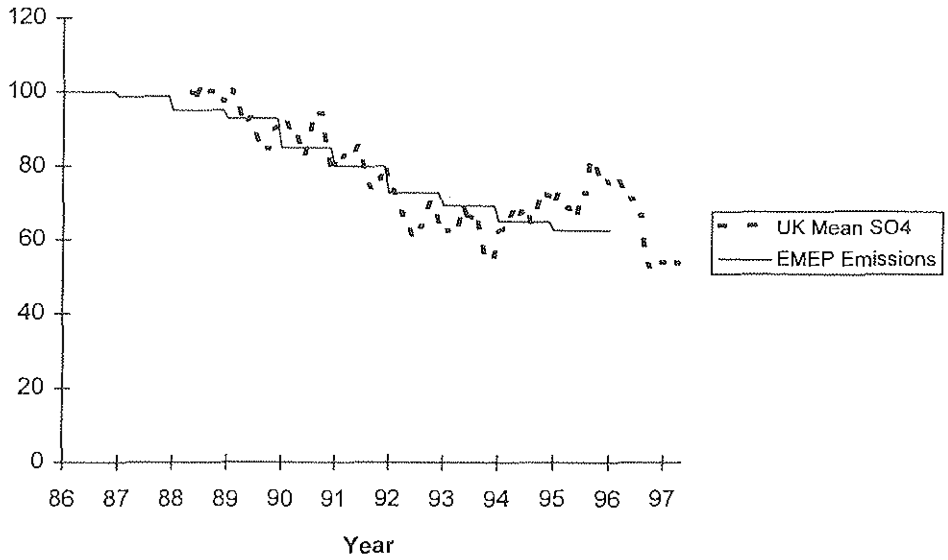
DIESEL CAR PM LIMITS (g/km)



HEAVY DUTY PM LIMITS (g/kWh) >85kW



UK Average aerosol sulphate vs European sulphur emissions



PM10 or PM2.5 ?

- **PM10 is confounded by coarse particles**
- **A very much higher fraction of PM2.5 is man-made**
- **The control levers we can pull on both pollutants *are the same***
- ***Does the coarse fraction of PM10 matter in health terms ?***

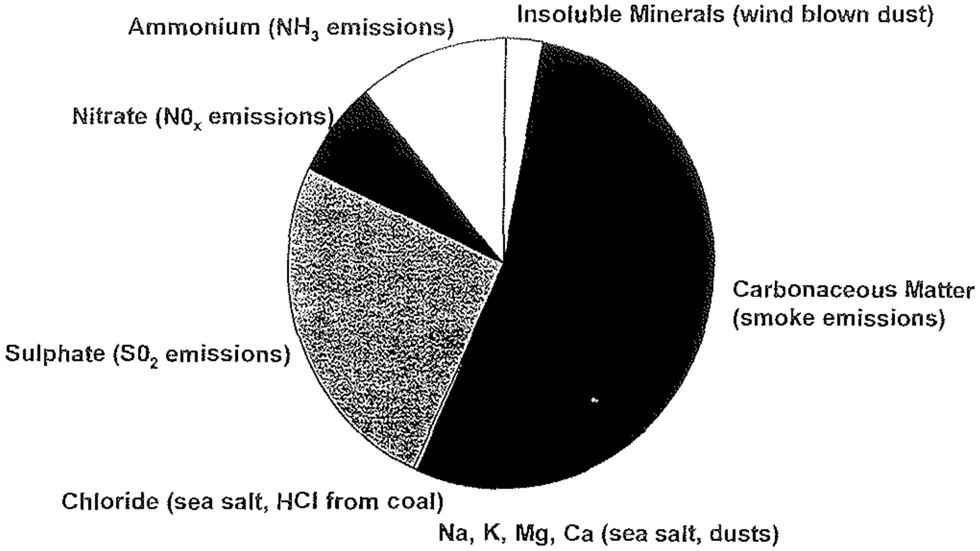
The Sources of PM10 and PM2.5 (based on UK APEG Report)

- In larger urban areas PM10 arises in roughly similar proportions from Primary, Secondary and Coarse particles
- Secondary PM10 is not controllable by one Member State alone
- Coarse fraction is probably not controllable at all

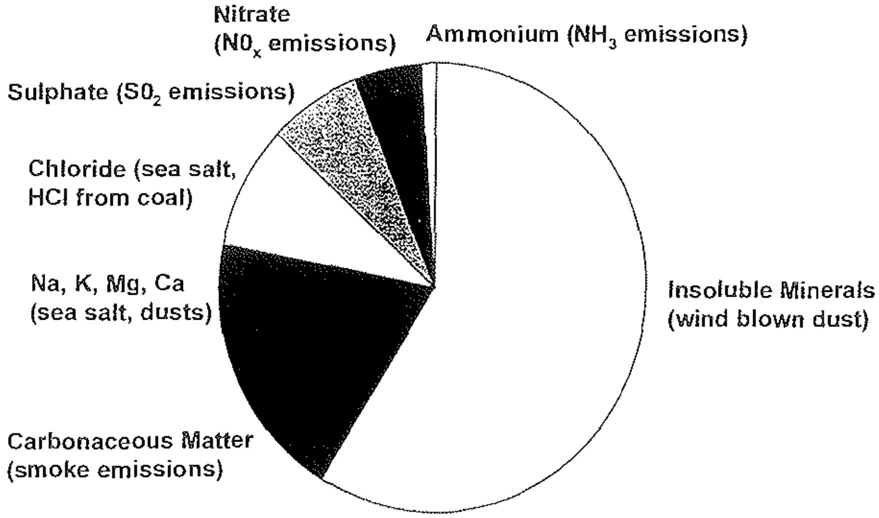
PM10 and PM2.5 Concentrations

- Closely correlated in urban and rural areas
- Very roughly PM2.5 ~ 0.6 PM10 on annual average
- Roadside levels of PM10 enhanced proportionally more, because of resuspended dust

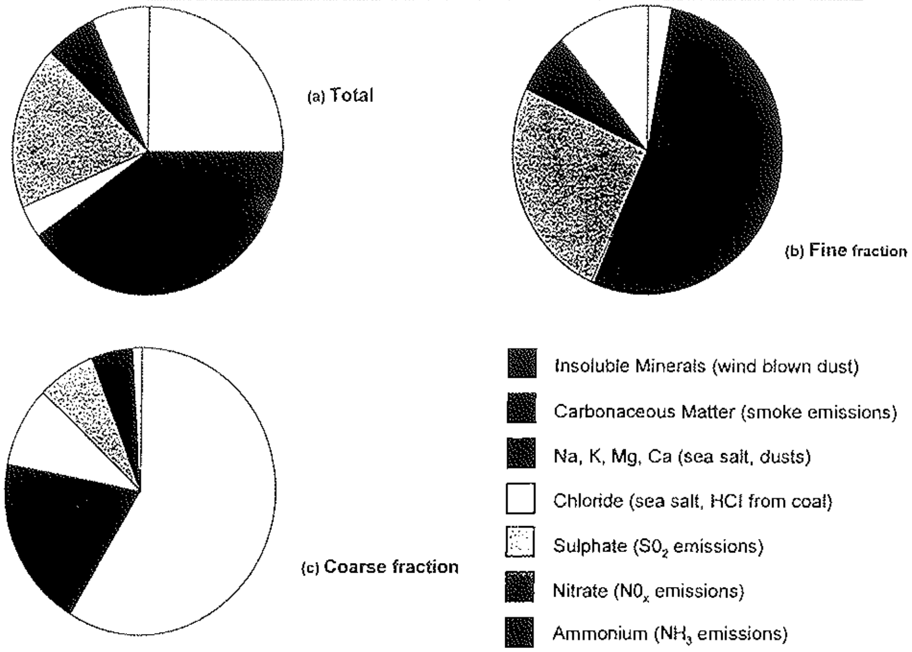
Fine fraction (< 2.5μm) (from Clarke, 1992)



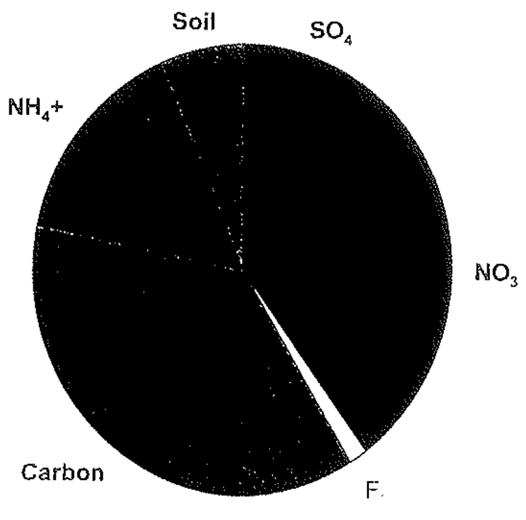
Coarse fraction (2.5 -15μm) (from Clarke, 1992)



Approximate composition of urban particles



Average fine particulate (aerodynamic diameter < 2.5μm) composition for two inversion episodes in February 1993, USA data



Sources of PM10 and PM2.5

- **In larger urban areas PM10 arises in roughly equal proportions from Primary, Secondary and Coarse particles**
- **Secondary PM is not controllable by one Member State alone**
- **Primary and Secondary particles have *different* optimum control strategies**

What do we need ? (I)-Atmospheric Data

- **More measurements of PM2.5 *and* PM10**
- **A Reference Method for PM2.5**
- **Chemical composition *by size fraction*, informed by toxicological/health research**
- **Measurements of *number, size distributions* co-located with PM10 and PM2.5 at more sites**

What do we need ? (II) - Understanding the Rules

- Understanding and modelling the chemical processes forming secondary aerosols
- More data on primary emissions from new technologies and fuels-no regrets?
- Understanding the dynamics of primary emissions - especially ultrafines

PM10 or PM2.5 ?

- PM10 is confounded by coarse particles
- A major fraction of PM2.5 is man-made
- The *controllable* sources of both metrics are the same
- PM2.5 penetrates further into the respiratory system-but need more European health studies
- Does the coarse fraction matter for health?

What Are People Exposed To and Where Do Particles Come From?

Chairs: *Giovanni Angeletti,* European Commission, DG XII; and *Robert Sawyer,* University of California at Berkeley, United States, and University College of London, United Kingdom

PETROS KOUTRAKIS
Harvard School of Public Health, United States

THE RELATIONSHIP BETWEEN PERSONAL PARTICULATE EXPOSURES AND THE CORRESPONDING OUTDOOR CONCENTRATIONS*

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Harvard University, School of Public Health
665 Huntington Avenue, Boston MA 02115
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Presented below is an overview of the current understanding of the relationship between personal particulate exposures and corresponding outdoor concentrations. This information is important to understand the results of the epidemiological studies which have used ambient particle concentrations as surrogates for personal exposures. Results from the US EPA PTEAM study showed that PM_{10} outdoor concentrations explained only 16% of the variability in personal exposures. For fine particles ($PM_{2.5}$ and $PM_{3.5}$), the results were conflicting, as correlations ranged from low and non-significant (Spengler et al, 1985 and Sexton et al, 1984) to significant, r^2=0.69, (Dockery and Spengler, 1985). These poor correlations were attributed to the importance of indoor activities, such as smoking, cooking, dusting, and vacuuming. As such, Tamura et al (1996) found strong correlations between personal and outdoor concentrations when the masking influences of indoor combustion sources were removed and resuspension of particles was eliminated.

Wallace, upon re-examination of existing personal exposure data, demonstrated that pooling the data together for cross-sectional analysis may be another factor influencing the strength of the observed correlations between personal exposures and outdoor concentrations (Wallace, 1996). Janssen et al (1998) conducted a personal particulate monitoring study in the Netherlands. Four to eight measurements were obtained for 13 non-smoking adults in Amsterdam and 15 children in Wageningen (a semi-rural environment). The results of these studies indicated that longitudinal analyses yielded a reasonably high correlation between personal and ambient PM_{10} within individuals (median r=0.57). Re-analysis of data from Phillispsburg, NJ, where personal PM_{10} exposures of 14 individuals were measured for 14 days, also yielded improved correlations on an individual level (median r^2=0.46), as compared to an r^2=0.037 in the original cross sectional analysis (Lioy and Waldman, 1990). These studies indicated that cross-sectional analyses result in lower correlations between personal and outdoor concentrations because the inter-personal variability masks the intra-personal correlations (Wallace, 1996 and Janssen et al, 1998).

A study was performed in Boston, MA, during the summer and winter of 1996 to characterize the personal PM_{10} and $PM_{2.5}$ exposures of individuals with Chronic Obstructive Pulmonary Disease (COPD) (Rojas et al, 1997). This study included individuals with moderate to severe COPD who lived in non-smoking private households. 12-hr simultaneous measurements of personal, indoor, and outdoor particulate levels were conducted. 18 participants were measured over 6- to 18-day periods in the winter and summer seasons. Results showed that the association between personal particulate exposures and outdoor concentrations varied with both size fraction and season when data were analyzed cross-sectionally. Personal $PM_{2.5}$ exposures were significantly associated with the corresponding outdoor concentrations. Spearman correlation coefficients were 0.30 and 0.63 for the winter and the summer seasons, respectively. Personal PM_{10} exposures were significant only in the summer season (Spearman correlation coefficient was 0.45). Observed seasonal differences in the personal-outdoor relationships were attributed to variability in home ventilation characteristics and air exchange rates. The personal-outdoor concentration association was stronger when data were analyzed by individual. Subject–specific linear regressions of personal-outdoor $PM_{2.5}$ levels showed that 10 out of 18 subjects had significant r^2 values, ranging between 0.32 and 0.86. The slopes of the regression lines varied widely, ranging between 0.47 and 1.64. Regressions for PM_{10} levels showed that only 3 out of 18 subjects had significant r^2 values, ranging between 0.23 and 0.40. The regression slopes ranged between 0.72 and 0.88. This inter-individual variability was attributed to differences in activity patterns and housing characteristics. Together, these results suggest that personal-outdoor associations vary substantially by individual.

A large number of exposure studies are underway in the U.S. (NRC, 1999). The data from these studies, along with those of the multinational European exposure study EXPOLIS (Jantunen et al, 1998), will provide sufficient information to elucidate the factors influencing relationships between personal particulate exposures and corresponding outdoor concentrations.

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Janssen N., Hoek G., Brunekreef B., Harssema H., Mensink I., Zuidhof A. (1998). "Personal Sampling of Particles in Adults: Relation among Personal, Indoor, and Outdoor Air Concentrations." *Am. J. Epidemiol.*, 147: 537-547.

Jantunen M., et al. (1998). "Air Pollution Exposure in Europen Cities: The "EXPOLIS" Study." *J. of Exposure Anal. and Env. Epid.* 8: 495-518.

Rojas L., Koutrakis P., Suh H. (1998). "Personal Exposure to Particulate Matter and its Relationship to Outdoor Concentrations in a Group of COPD Patients Living in the Boston Area." Presented at *PM2.5: A Fine Particulate Standard Conference*, Jan 28-30, Long Beach, CA.

Sexton K., Spengler J., Treitman R. (1984). "Personal Exposure to Respirable Particles: A Case Study in Waterbury, Vermont." *Atmos. Environ.*, 18: 1385-1398.

Spengler J., Treitman R., Tosteson T., Mage D., Soczek M. (1985). "Personal Exposures to Respirable Particulates and Implications for Air Pollution Epidemiology." *Environ. Sci. Technol.*, 19: 700-707.

Tamura K., Ando M., Sagai M., Matsumoto Y. (1996). "Estimation of Levels of Personal Exposure to Suspended Particulate Matter and Nitrogen Dioxide in Tokyo." *Environ. Sci. (Tokyo)*, 4: 37-51.

Wallace L. (1996). "Indoor Particles: A Review." *J. Air & Waste Manage. Assoc.*, 46: 98-126.

BACKGROUND

Epidemiological Studies

- Associations between CV admissions and 24-h mean particle levels shown with pollution lagged 0-2 days
- A. Peters found an association between the onset of MIs and $PM_{2.5}$ concentrations both 2 hours and one-day (24-48 hours) prior to the onset of symptoms

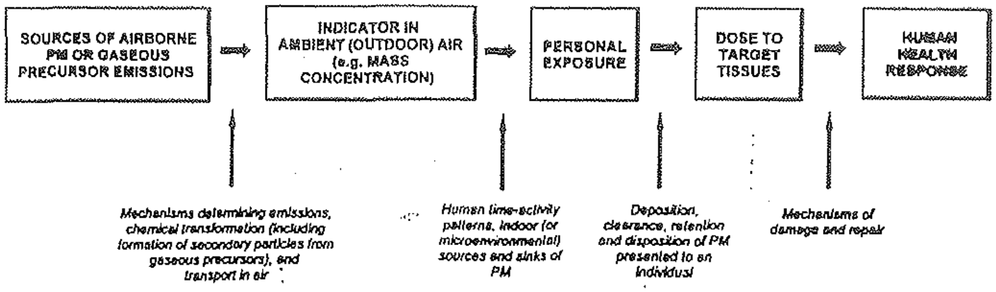
Exposure Studies

- Relationship between personal exposures-outdoor concentrations is individual-specific
- Activity patterns and home ventilation characteristics are important determinants of personal exposures

OBJECTIVES

- Improve our understanding of particle exposures
  - Use findings from our previous particulate exposure studies to design new study
  - Combine data from particle super-site with additional data on personal exposures to particle components
- Develop improved exposure estimates for use by epidemiogical study
  - Marry design of exposure assessment study with epidemiological study(-ies)
  - Improve our ability to examine the association between exposure and effect

(Figure 3.1) **A General Framework for Integrating PM Research**



SOURCES OF AIRBORNE PM OR GASEOUS PRECURSOR EMISSIONS → INDICATOR IN AMBIENT (OUTDOOR) AIR (e.g. MASS CONCENTRATION) → PERSONAL EXPOSURE → DOSE TO TARGET TISSUES → HUMAN HEALTH RESPONSE

Mechanisms determining emissions, chemical transformation (including formation of secondary particles from gaseous precursors), and transport in air

Human time-activity patterns, indoor (or microenvironmental) sources and sinks of PM

Deposition, clearance, retention and disposition of PM presented to an individual

Mechanisms of damage and repair

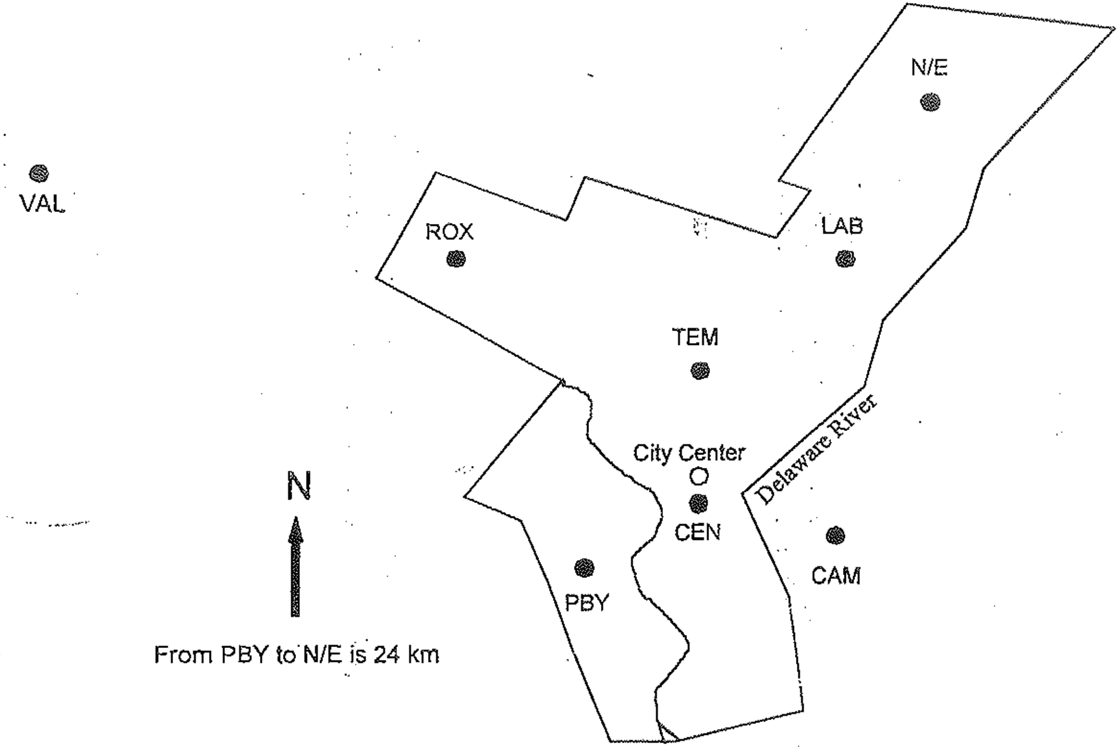
Modified from NRC 1983, 1994; Lioy 1990; and Saxton 1992.

METROPOLITAN ACID AEROSOL CHARACTERIZATION STUDY

- **Objectives**
  - to examine spatial and temporal variation in acid aerosol and particle levels
  - to determine the composition of particulate matter during high air pollution periods
- **Study Locations**
  - Philadelphia, PA: 1992 and 1993
  - Washington, D.C.: 1994
  - Nashville, TN: 1995
  - Boston, MA: 1996

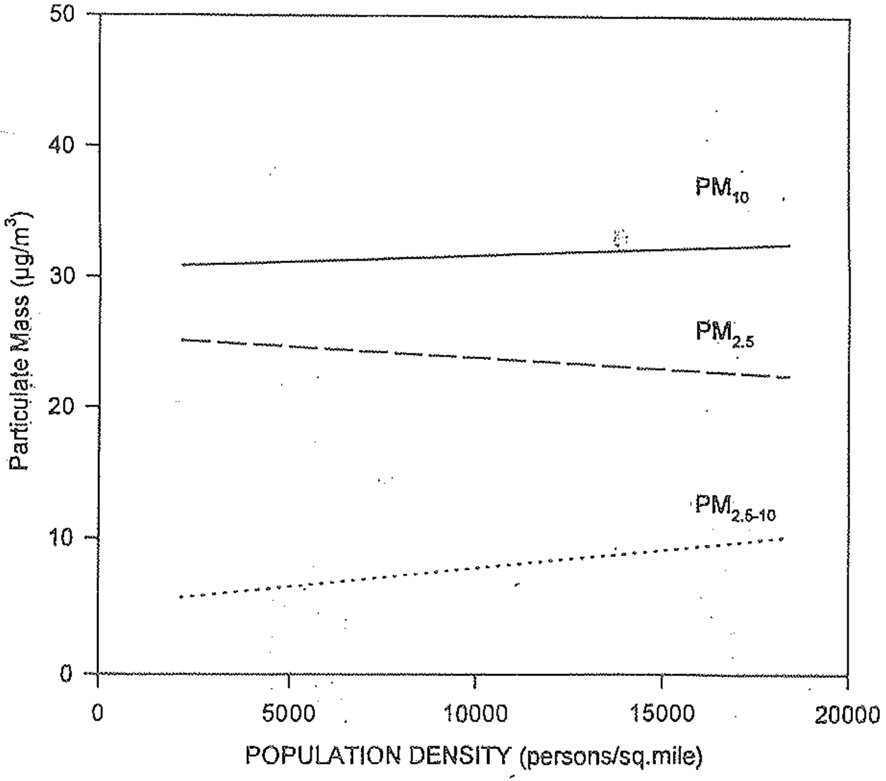
Philadelphia Monitoring Sites (1992 and 1993)

Metropolitan Acid Aerosol Characterization Study



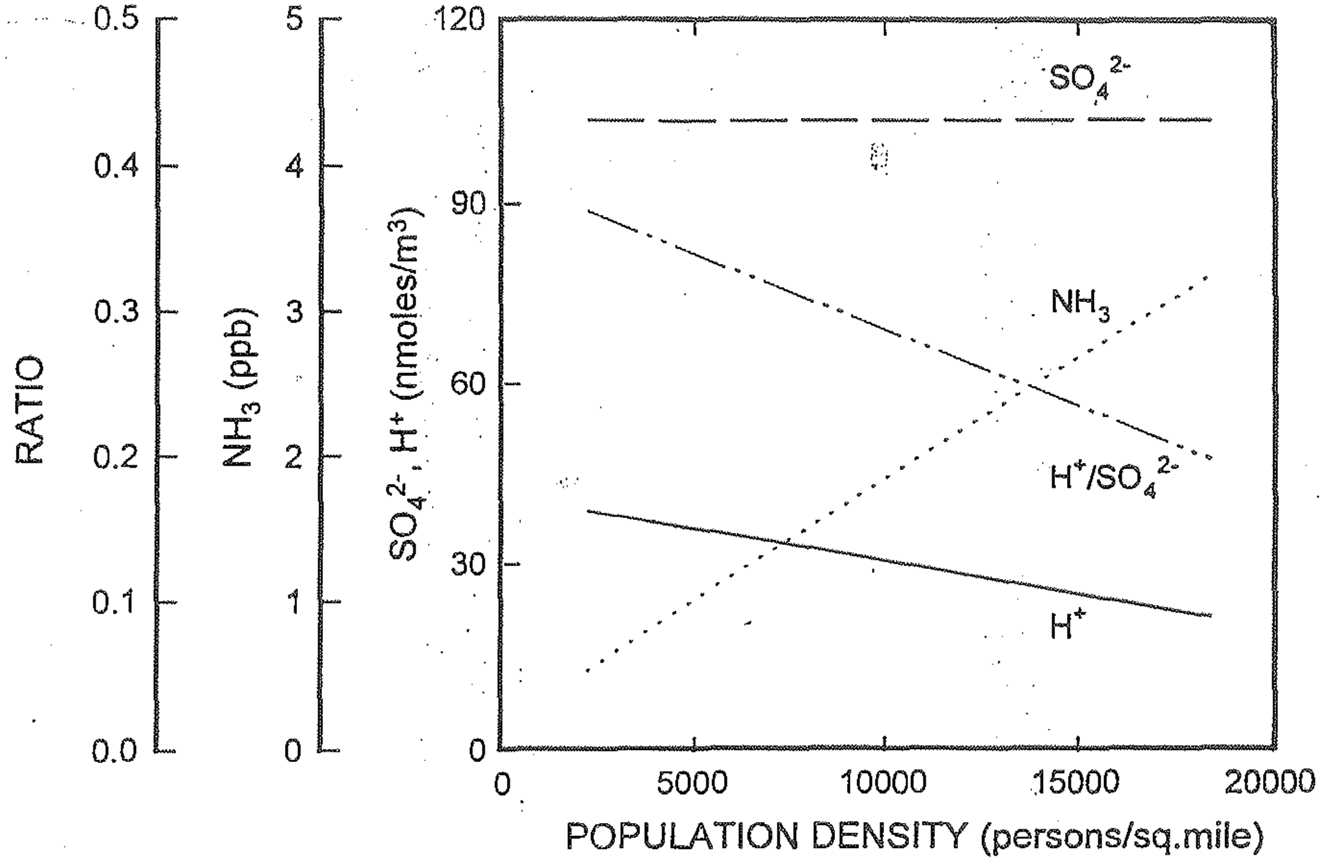
PARTICULATE MASS VS. POPULATION DENSITY

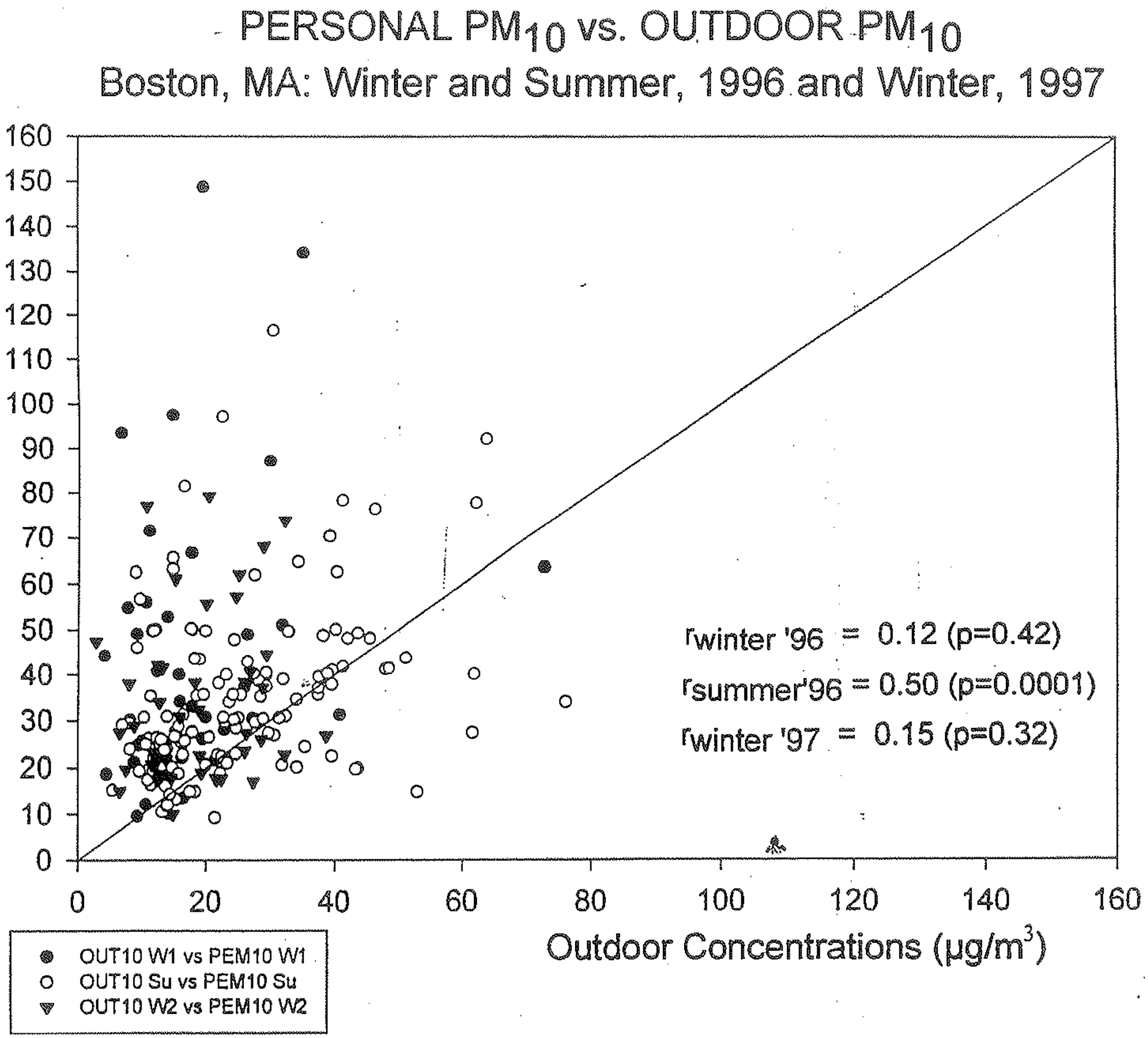
Philadelphia, PA -- Summers 1992 and 1993

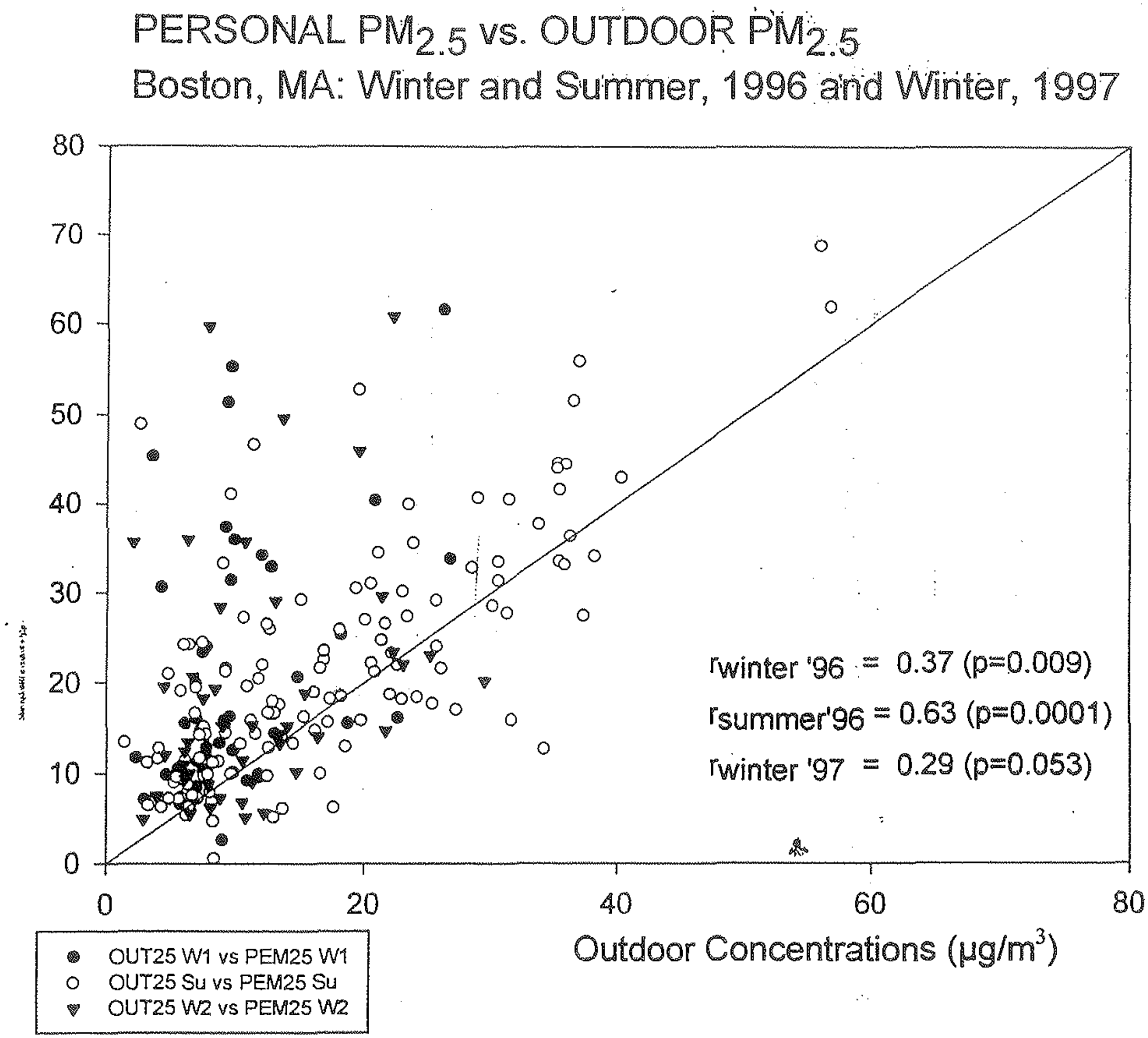


MEAN SO_4^{2-}, H^+, AND NH_3

Philadelphia, PA, Summer 1992, 1993: South Winds




PERSONAL PM10 vs. OUTDOOR PM10
Boston, MA: Winter and Summer, 1996 and Winter, 1997
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
0
20
40
60
80
100
120
140
160
Outdoor Concentrations (µg/m³)
r winter '96 = 0.12 (p=0.42)
r summer'96 = 0.50 (p=0.0001)
r winter '97 = 0.15 (p=0.32)
● OUT10 W1 vs PEM10 W1
○ OUT10 Su vs PEM10 Su
▼ OUT10 W2 vs PEM10 W2



PERSONAL $PM_{2.5}$ vs. OUTDOOR $PM_{2.5}$
Boston, MA: Winter and Summer, 1996 and Winter, 1997

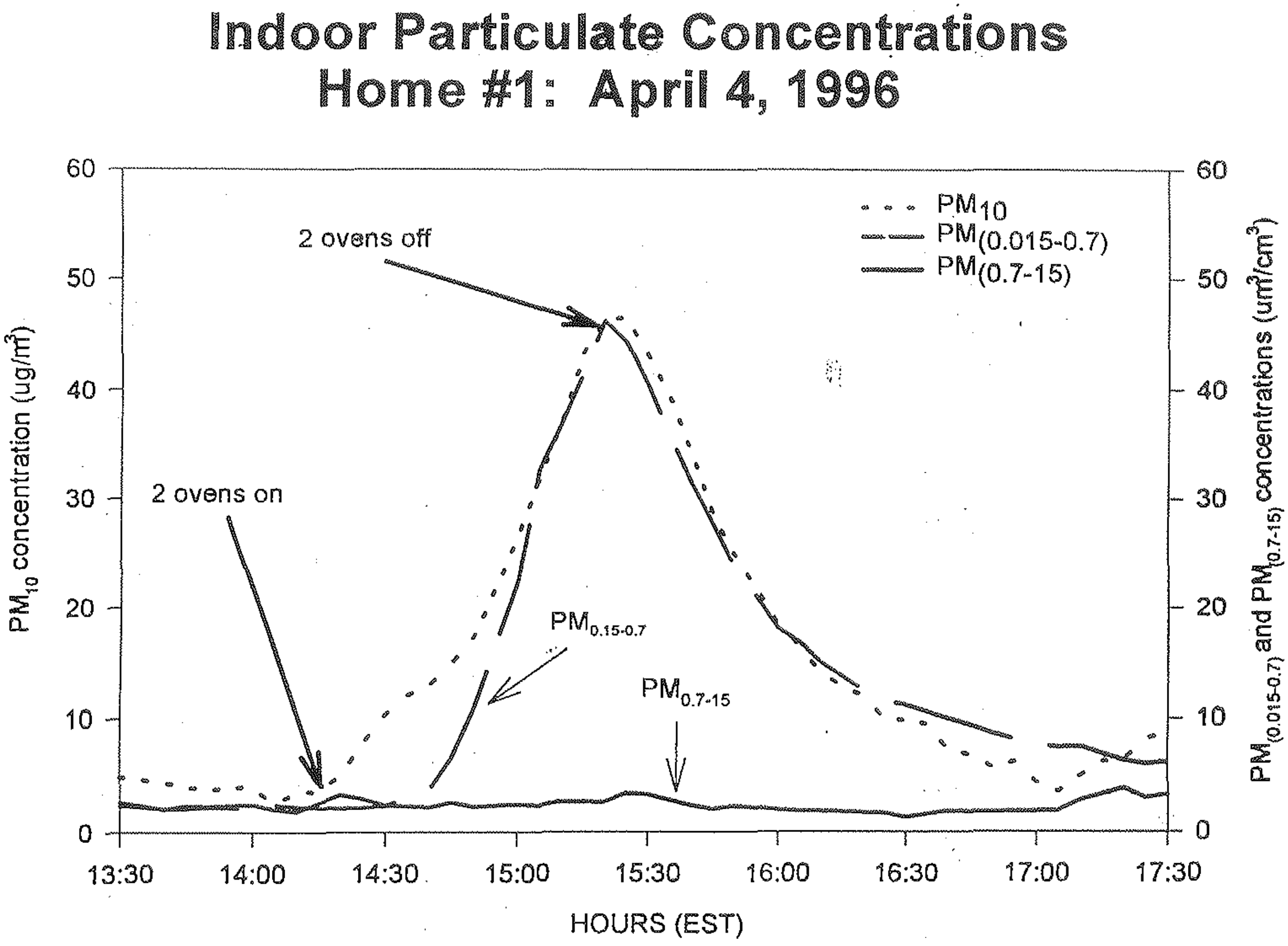
Subject-by-Subject Linear Regression Results

$PEM_{2.5}$ vs $OUT_{2.5}$			PEM_{10} vs OUT_{10}		
n	R^2	p-value	n	R^2	p-value
10	**0.87**	**0.0001**	10	0.13	0.32
6	**0.86**	**0.0075**	6	0.40	0.18
17	**0.83**	**0.0000**	17	**0.40**	**0.0068**
17	**0.65**	**0.0001**	18	**0.35**	**0.0102**
10	**0.58**	**0.0109**	11	0.12	0.30
12	**0.52**	**0.0080**	12	0.23	0.12
12	**0.44**	**0.0187**	12	0.02	0.66
18	**0.35**	**0.0102**	18	**0.22**	**0.0507**
17	**0.32**	**0.0186**	17	0.12	0.17
11	0.28	0.0963	11	0.03	0.62
6	0.10	0.54	6	0.01	0.88
18	0.08	0.25	18	0.12	0.16
11	0.02	0.70	11	0.00	0.87
11	0.02	0.71	11	0.00	0.96
16	0.01	0.67	16	0.03	0.52
17	0.01	0.66	17	0.12	0.17
12	0.01	0.79	12	0.03	0.62
6	0.00	0.97	5	0.09	0.62
Med	0.30			0.12	

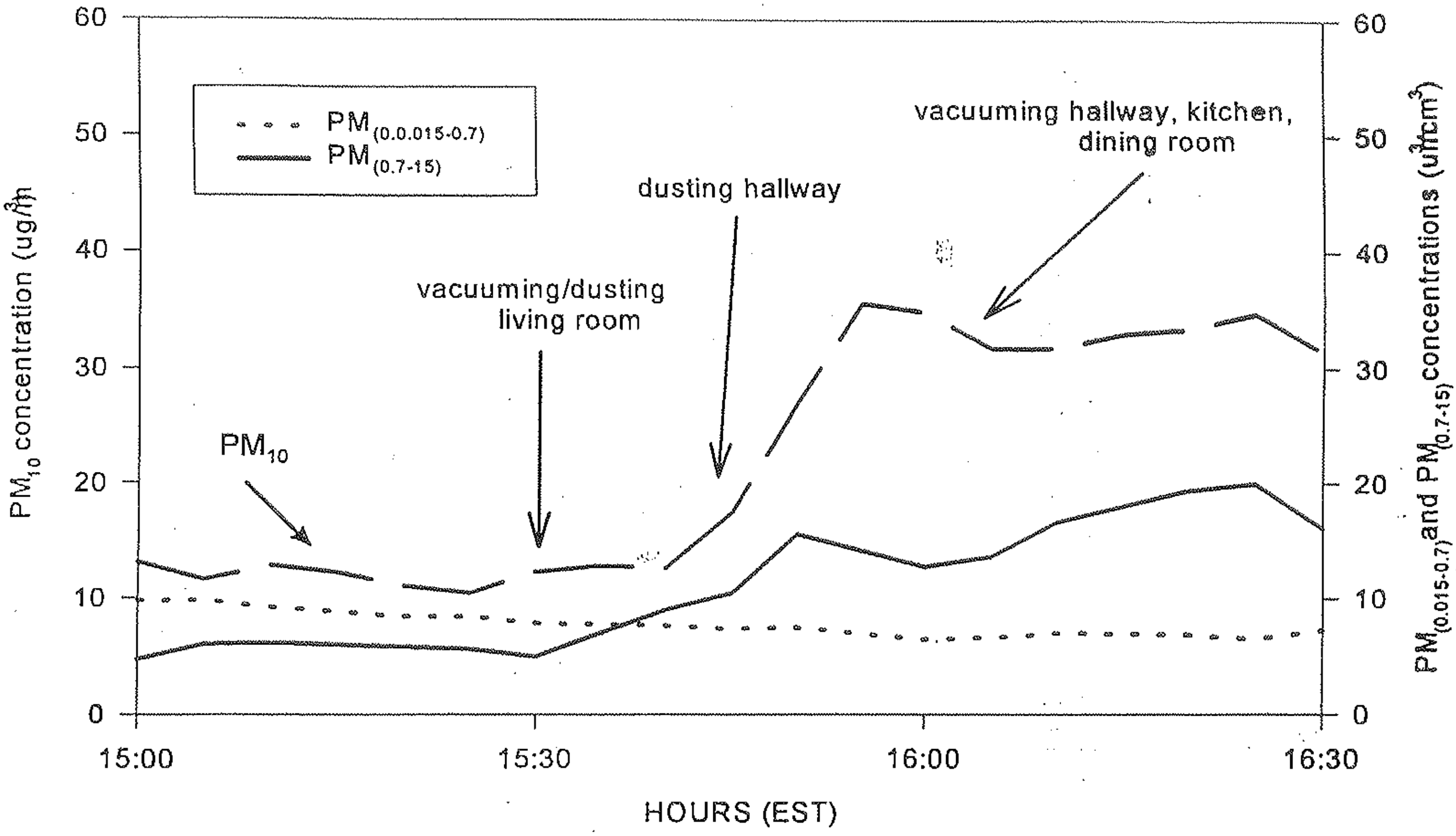
DESIGN AND METHODS

- Four homes sampled
  - Two homes sampled in Winter and Summer of 1996, while two homes sampled in Summer 1996 only.
  - Each home sampled for six days in a season.

- Data collection methods
  - Size distribution measurements
    - Scanning Mobility Particle Sizer (SMPS)
    - Aerodynamic Particle Sizer (APS)
  - Air exchange rates
  - Time activity information


Indoor Particulate Concentrations
Home #1: April 4, 1996
PM10
PM(0.015-0.7)
PM(0.7-15)
2 ovens off
2 ovens on
PM0.15-0.7
PM0.7-15
PM10 concentration (ug/m3)
PM(0.015-0.7) and PM(0.7-15) concentrations (um3/cm3)
0
10
20
30
40
50
60
13:30
14:00
14:30
15:00
15:30
16:00
16:30
17:00
17:30
HOURS (EST)

Indoor Particulate Concentrations
Home #01: March 30, 1996



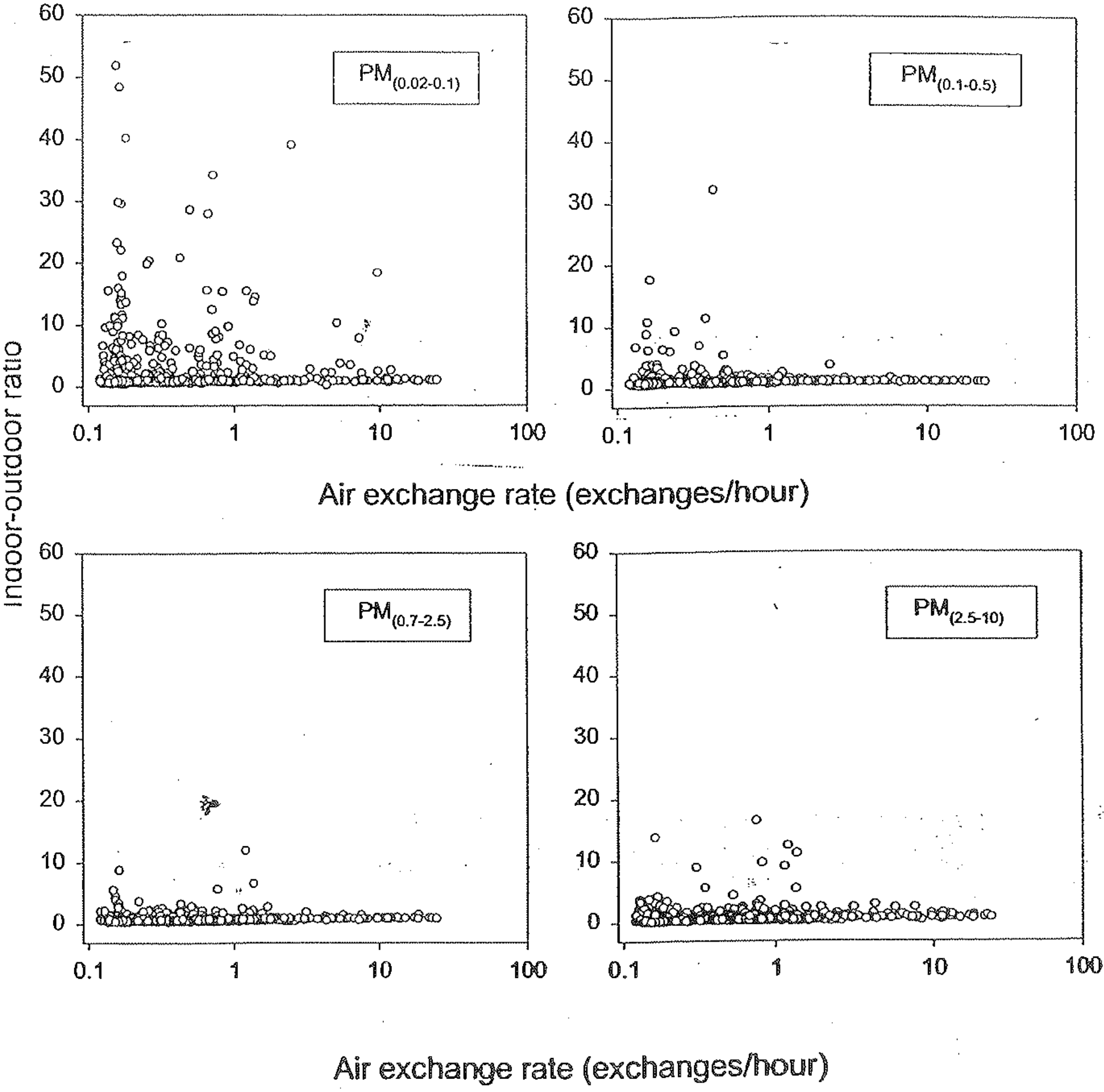

PM(0.02-0.1)
PM(0.1-0.5)
PM(0.7-2.5)
PM(2.5-10)
Indoor-outdoor ratio
0
10
20
30
40
50
60
0.1
1
10
100
Air exchange rate (exchanges/hour)

Figure 1. Multi-Pollutant Personal Sampler

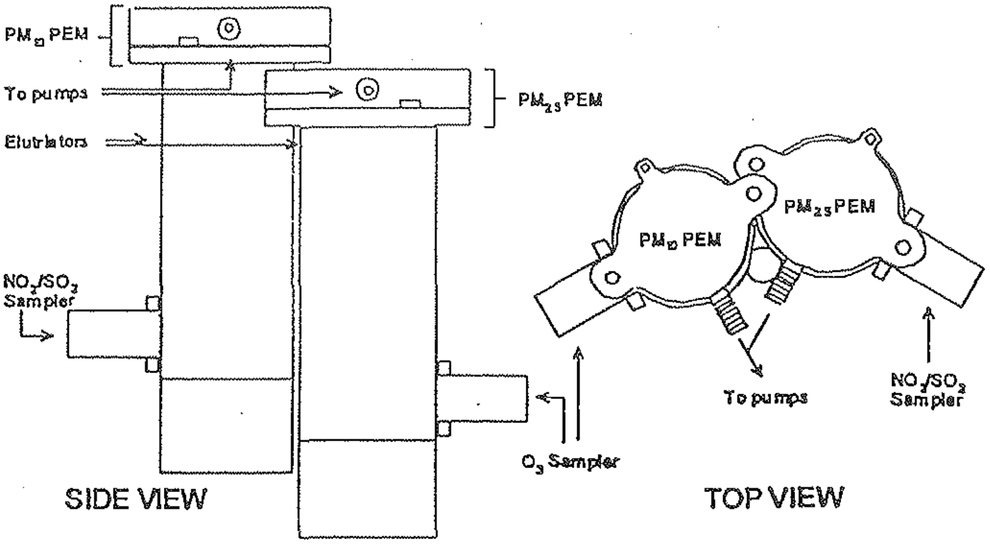


Figure 3c

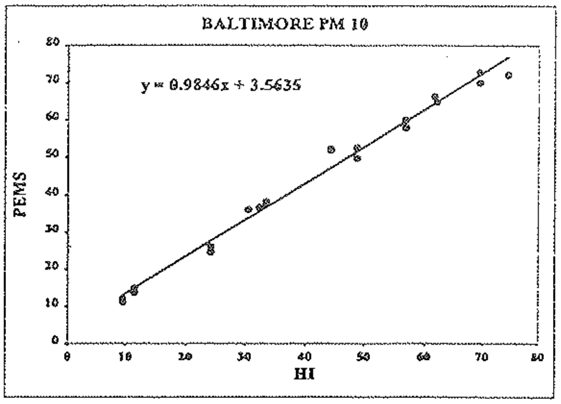


Figure 3d

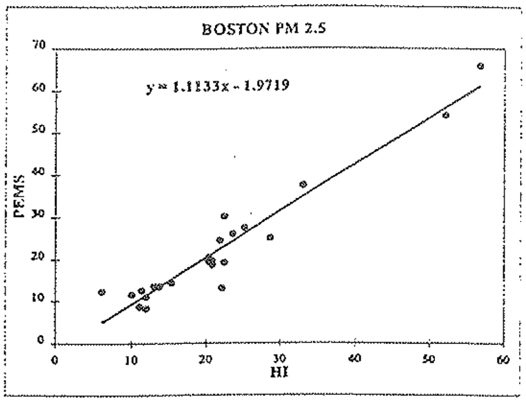


Figure 4a

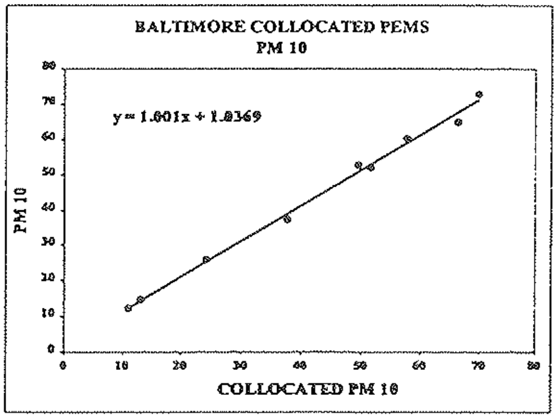


Figure 4b

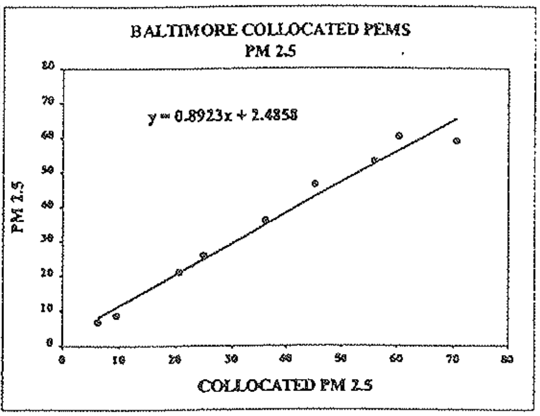


Figure 2a

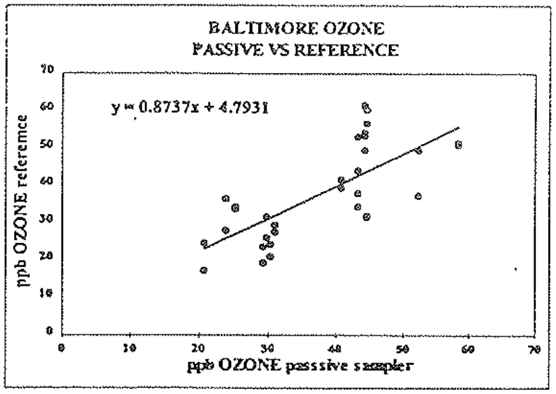


Figure 2b

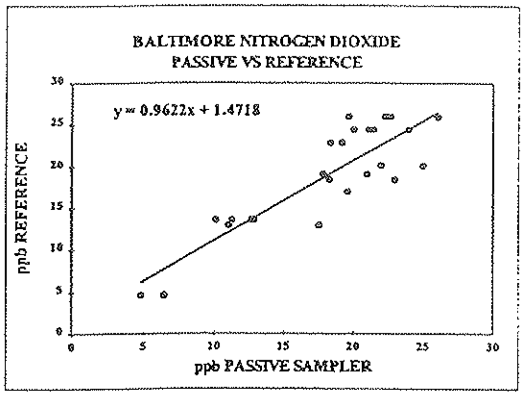


Figure 3a

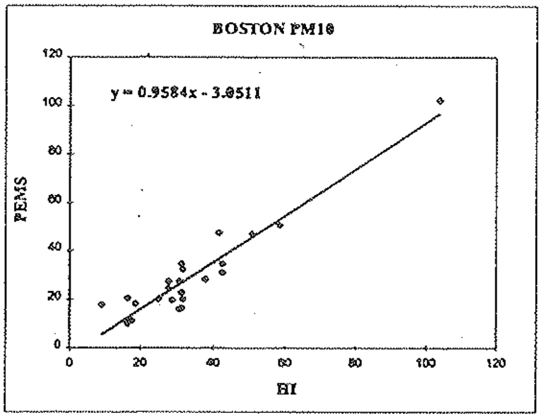
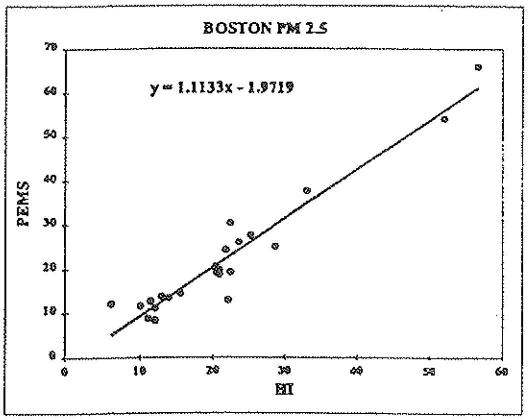


Figure 3b



EMILE DE SAEGER

Joint Research Centre ISPRA, Italy

Joint Meeting of the EC and HEI
"The Health Effects of Fine Particles: Key Questions and the 2003 Review "
Brussels, Belgium, 14-15 January 1999

CAPABILITIES AND LIMITATIONS OF AVAILABLE PARTICLE MEASUREMENT TECHNOLOGIES

E. De Saeger

European Reference Laboratory of Air Pollution
European Commission - Joint Research Centre / Ispra - Environment Institute

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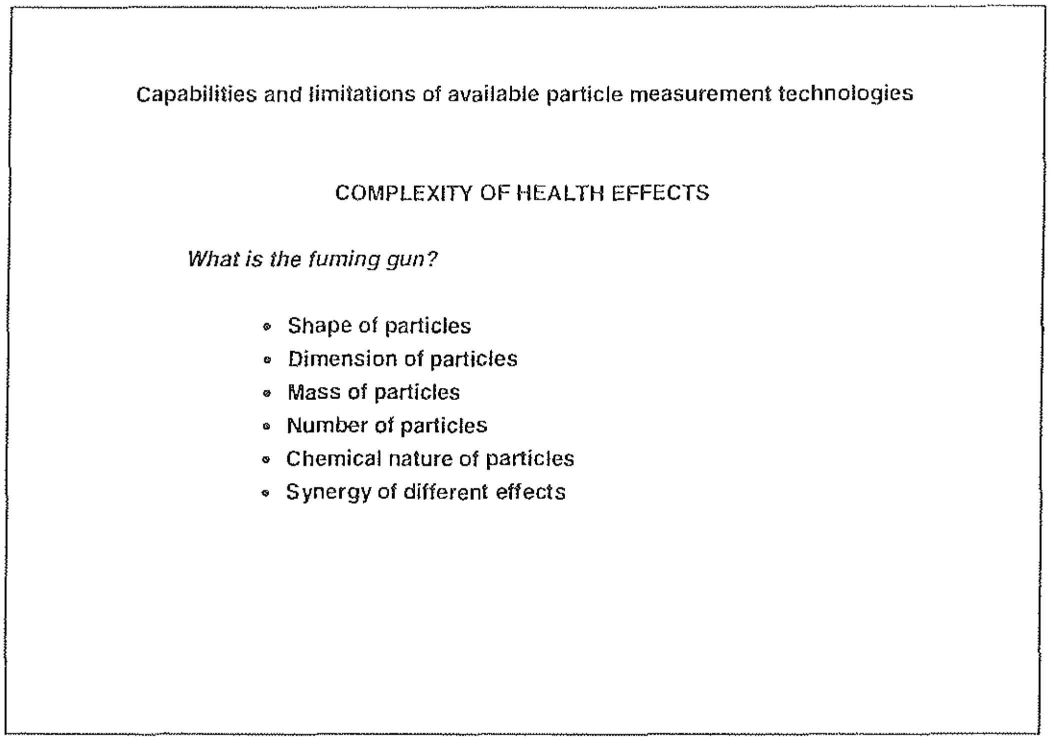
Capabilities and limitations of available particle measurement technologies

COMPLEX NATURE OF PARTICULATE MATTER

What are we really measuring?

- PM from various natural and anthropogenic sources
- Particles of different diameters
- Cocktail of different chemical substances
- Primary and secondary aerosols
- Core material and adsorbed substances

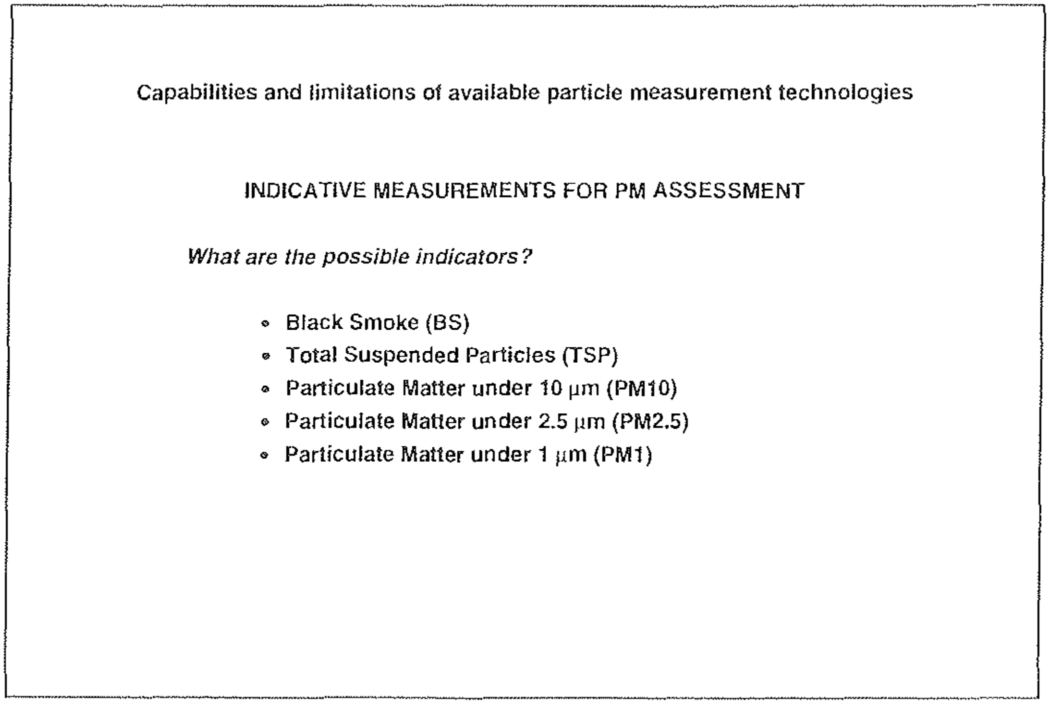


Capabilities and limitations of available particle measurement technologies

COMPLEXITY OF HEALTH EFFECTS

What is the fuming gun?

- Shape of particles
- Dimension of particles
- Mass of particles
- Number of particles
- Chemical nature of particles
- Synergy of different effects



Capabilities and limitations of available particle measurement technologies

INDICATIVE MEASUREMENTS FOR PM ASSESSMENT

What are the possible indicators?

- Black Smoke (BS)
- Total Suspended Particles (TSP)
- Particulate Matter under 10 µm (PM10)
- Particulate Matter under 2.5 µm (PM2.5)
- Particulate Matter under 1 µm (PM1)

Capabilities and limitations of available particle measurement technologies

SPECIATION OF PARTICULATE MATTER

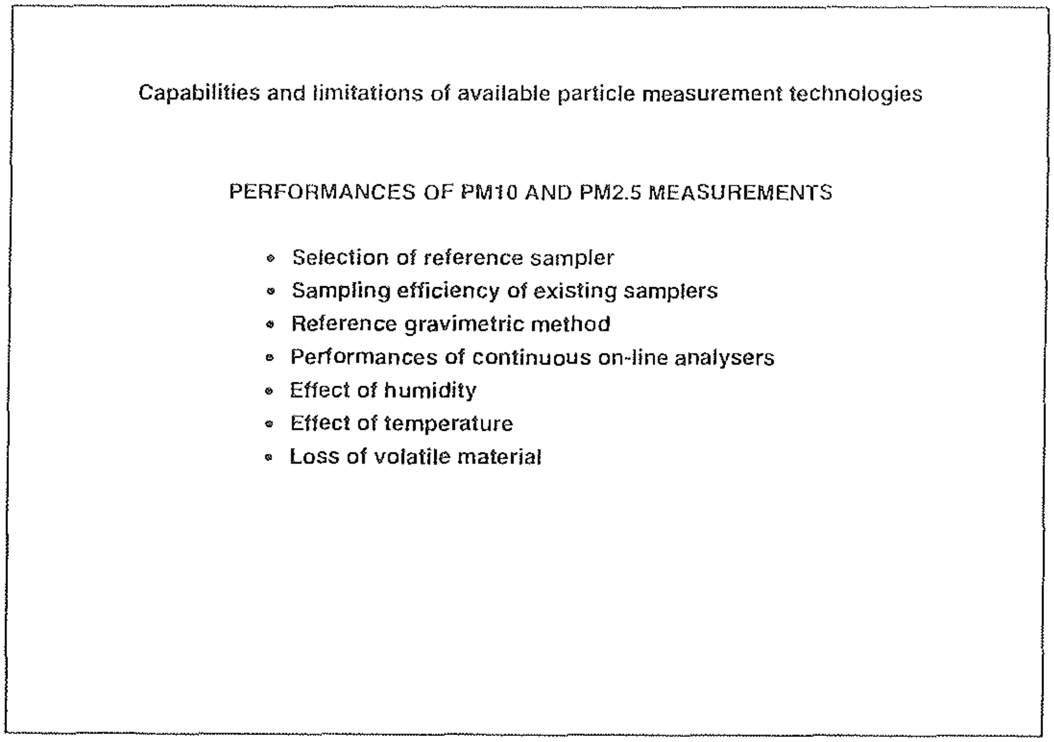
Let's go for the real culprits!

- Total acidity, tar
- Asbestos fibres
- Nitrates, sulphates, ammonium
- Elemental analysis
- Heavy Metals (Pb, Cd, Ni, As, Hg, ...)
- Poly-Aromatic Hydrocarbons (PAHs)
- Nitro Poly-Aromatic Hydrocarbons
- Elemental Carbon (EC)

Capabilities and limitations of available particle measurement technologies

PERFORMANCES OF BLACK SMOKE MEASUREMENTS

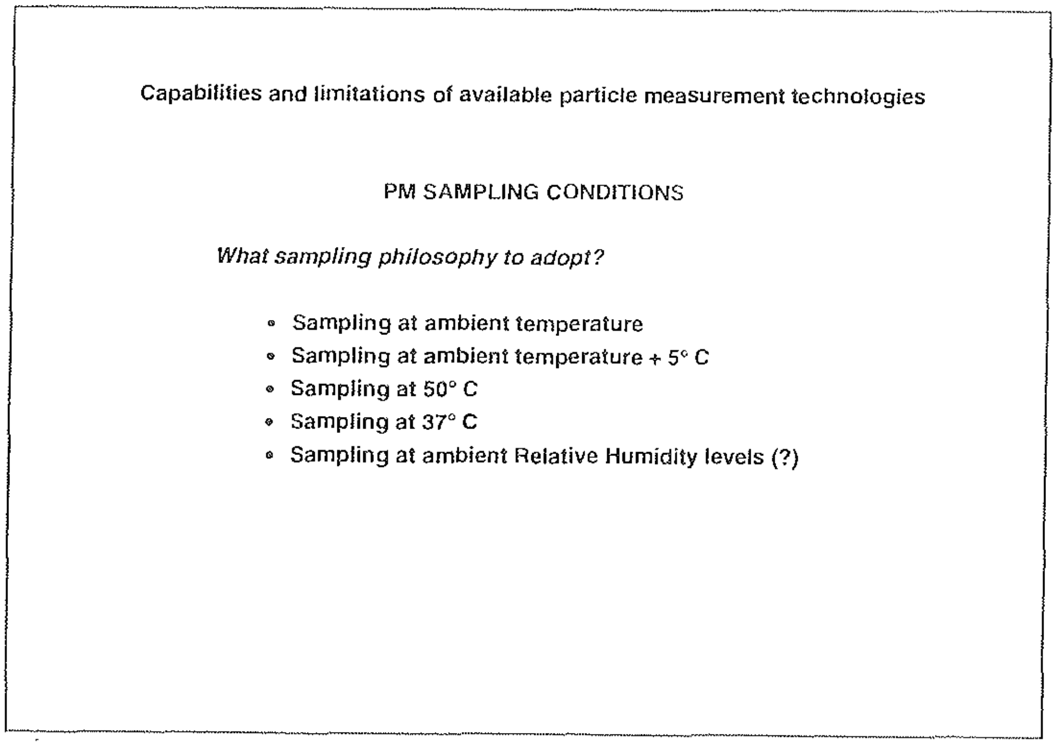
- Measurement of filter reflectance
- Simple and cost effective method
- Good comparability of data between laboratories
- BS indicator of black PM only
- Poor correlation with gravimetric PM determination
- Long historical time series available
- BS indicator for Elemental Carbon (?)



Capabilities and limitations of available particle measurement technologies

PERFORMANCES OF PM10 AND PM2.5 MEASUREMENTS

- Selection of reference sampler
- Sampling efficiency of existing samplers
- Reference gravimetric method
- Performances of continuous on-line analysers
- Effect of humidity
- Effect of temperature
- Loss of volatile material



Capabilities and limitations of available particle measurement technologies

PM SAMPLING CONDITIONS

What sampling philosophy to adopt?

- Sampling at ambient temperature
- Sampling at ambient temperature + 5° C
- Sampling at 50° C
- Sampling at 37° C
- Sampling at ambient Relative Humidity levels (?)

Capabilities and limitations of available particle measurement technologies

NETWORK DESIGN AND CRITERIA FOR MEASURING SITE SELECTION

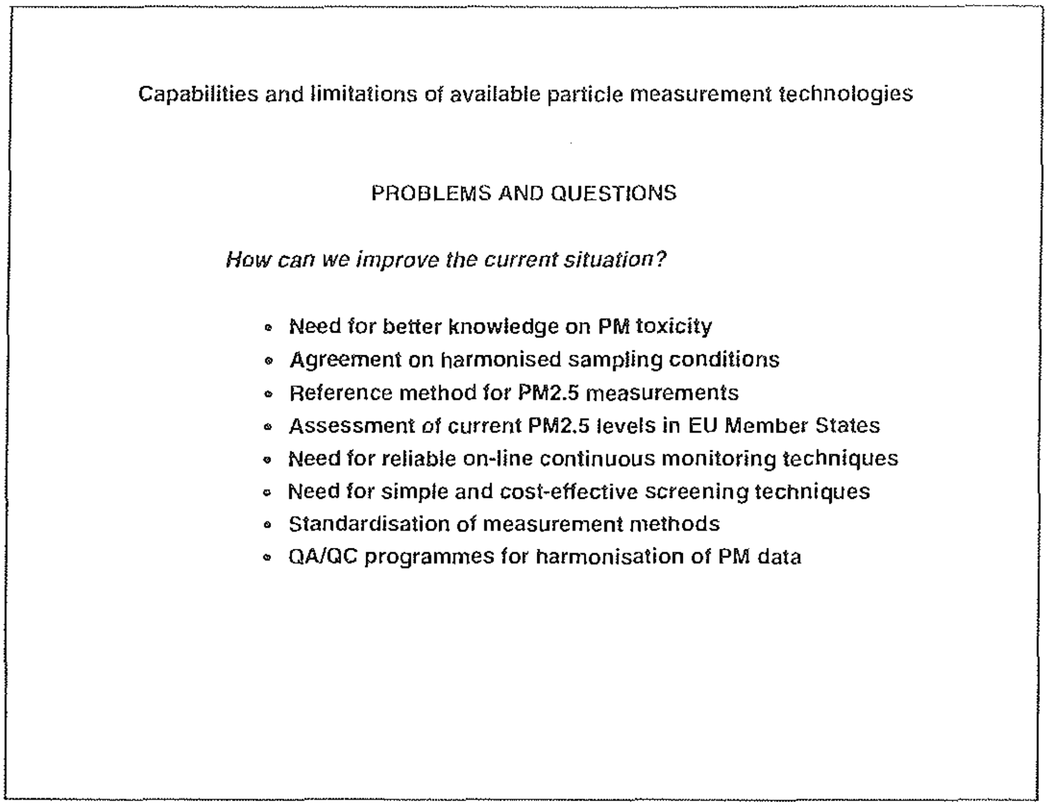
- Site selection criteria
    - Areas with maximum pollution levels
    - Areas representative of population exposure
- Minimum number of sites
- Further reduction of site number possible
- Low spatial variability of PM levels

Capabilities and limitations of available particle measurement technologies

EXPOSURE OF POPULATION

What are possible assessment strategies?

- Assessment of spatial distribution of PM levels
- Assessment of PM levels in areas of specific interest
- Personal exposure measurements



Capabilities and limitations of available particle measurement technologies

PROBLEMS AND QUESTIONS

How can we improve the current situation?

- Need for better knowledge on PM toxicity
- Agreement on harmonised sampling conditions
- Reference method for PM2.5 measurements
- Assessment of current PM2.5 levels in EU Member States
- Need for reliable on-line continuous monitoring techniques
- Need for simple and cost-effective screening techniques
- Standardisation of measurement methods
- QA/QC programmes for harmonisation of PM data

APPORTIONING PARTICLES TO THEIR SOURCES

JAN BERDOWSKI

TNO Institute, The Netherlands

Joint Meeting of the EC and HEI, Brussels 14 and 15 January 1999:
The Health Effects of Fine Particles: Key Questions and the 2003 Review

Particulate Matter Emission Estimates for several European Countries

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Abstract

For the outlining of policy on ambient air fine particulate concentration by the Ministry of Environment of the Netherlands additional knowledge on particulate emissions was required on a European scale. This information had to be suitable as input for atmospheric dispersion modelling, for scenario calculation and for presentation purposes. A sector-split European emission inventory has been prepared for PM_{10}, $PM_{2.5}$ and $PM_{0.1}$ for the years 1990 and 1993 on a 0.5° x 1° latitude - longitude grid scale [1]. The inventory has been restricted to primary aerosols from a number of contributing sectors, which are expected to cover the bulk of the anthropogenic sources: stationary combustion, industrial process emissions, transport (including maintenance, tyre wear, road abrasion and re-suspension for road transport), agricultural practice (pig and chicken factory farming) and waste incineration.

After the definition of the source groups relevant for the inventory, general and country specific information was collected on emission control for a range of combustion and production processes [1,2]. Subsequently, the emissions were calculated by applying emission factors for each process to a certain activity rate. The emission factors for PM_{10} were derived from open literature and specific measurement reports. Activity rates for combustion and production processes for each country were used from official statistics or when not available from other international emission databases. The $PM_{2.5}$ and the $PM_{0.1}$ emissions were calculated by characterizing the PM_{10} emissions with respect to the $PM_{2.5}$ and the $PM_{0.1}$ fractions. After the emission estimates were set for the individual source groups for the individual European countries, the country totals per sector were spatially distributed using point source information (location, capacity) for a large number of plants, population density distribution data or land use information.

For the year 2010, three types of emission predictions have been prepared [2]: a 'Business as usual' scenario, assuming no emission reduction activity in Europe and applying the RAINS 7.2 Official Energy Pathway Scenario (OEP); a 'Processes in Pipeline'scenario, applying OEP and assuming a range of agreed emission reduction activities (e.g. 2nd Sulphur Protocol); a 'Best Available Technology'scenario, applying OEP and BAT.

The total European (excl. the former SU) anthropogenic PM_{10}, $PM_{2.5}$ and $PM_{0.1}$ emissions were about 5100, 2900 and 430 ktonnes for 1990 respectively. Including the European part of the former SU would roughly double these emission figures. For PM_{10} and $PM_{2.5}$, stationary combustion processes contributed about 55% of total emissions, while industrial processes and transport contributed between 14-19% of total emissions. For $PM_{0.1}$, stationary combustion and industrial processes and transport each contributed about 30% of total emissions.

Power generation caused about 55% of total stationary combustion emissions. The contribution of residential combustion is more important for the most fine PM fractions. The industrial process emissions were dominated by activities from the iron and steel industry and refineries. The contribution of refineries emissions are more important for the finer PM fractions. For transport, the PM emissions were dominated by the contribution of heavy duty vehicles (60-70%).

Between 1990 and 1993 the PM emissions decreased because of economic circumstances and change in especially the type of fuel used for residential combustion in a number of countries.

When no emission reduction activities are applied the 2010 PM_{10} emission would increase by 20-25% compared to the 1990 emission values. Especially, transport emissions would increase strongly. When a series of reduction measures will be applied, the result for 2010 will probably be an emission level comparable to that in 1990, indicating an actual reduction by 20-25% compared to the BAU situation. When all Best Available Technology would be applied as well, an emission reduction of about 55% could be achieved, especially by reducing the emissions from stationary combustion and industrial production processes.

A very first approach to calculate the uncertainties in the inventory data, has been based on the statistical analysis of the information on emission factors. For the sources considered, the uncertainty in emissions for total Europe (without the former Soviet Union) range between 10 - 20% for the three PM fractions. The modelled annual average PM_{10} concentrations in the Netherlands (among others based on these inventory results) are about 50, 60 and 75% of the measured concentrations for respectively rural, urban and industrial area [3]. This could indicate an underestimate of the emissions in this inventory. This can be expected as a number of sources was not covered by this inventory (e.g. non-anthropogenic sources, incomplete re-suspension). On the other hand, it should be realized that both the measurement data and the model applied and the emission estimates contribute to the overall uncertainty observed so that the discrepancy between measured and modelled concentrations cannot be attributed to only the inaccuracy of emission data.

It is clear that the emission inventory results as presented here can be improved further and become more complete by applying more up-to-date information, by evaluating the results with national experts and by adding non-anthropogenic sources and more complete re-suspension information. Also, the results could be improved by using verification methods, e.g. by applying the combination of measurements of concentration, size distribution and chemical speciation of PM in the ambient air, atmospheric models and chemical characterization of the emissions of different sources.

References

1 *Berdowski, J.J.M., W. Mulder, C. Veldt, A.J.H. Visschedijk & P.Y.J. Zandveld (1997). Particulate matter emissions in Europe in 1990 and 1993. TNO-MEP-R 96/472, Apeldoorn, The Netherlands.*

2 *Berdowski, J.J.M., M.P.J. Pulles & A.J.H. Visschedijk (1998). Incremental cost and remaining emission in 2010 of Heavy Metals resulting from the implementation of the draft HM Protocol under the UN/ECE Convention on Long Range Transboundary Air Pollution. TNO-MEP-R 98/020, Apeldoorn, The Netherlands.*

3 *Bloemen, H.J.Th., L. van Bree, E. Bruringh, P.H. Fischer, S. de Loos, M. Marra & P.J.A. Rombout (1998). Fine dust in the Netherlands. A Preliminary balance (In Dutch). RIVM R 650010 006, Bilthoven, The Netherlands.*

Particulate Matter Emission Estimates for several European Countries

Jan Berdowski

TNO, The Netherlands

Introduction

Methodology

Results
- Emissions
- Source categories
- Countries

Trends 1990 – 1993

Predictions 2010

Conclusions

Frame

- Policy outlining Ministry of Environment, the Netherlands
- Aspects of ambient fine matter concentrations

Aim

Emission data suitable for

- atmospheric dispersion models
- scenario calculation
- data presentation

Approach

- Prepare emission inventory PM10
  - Definition of relevant source groups
  - General and countries experts knowledge on emission control
  - Emission factors, derived from literature or specific measurement reports
  - Activity rates from official statistics (e.g. IEA)
- Prepare emission inventory PM2.5, PM0.1
  - Fraction of PM10 emission
- Prepare projections
  - General and countries expert knowledge on specific technological or autonomous developments
  - Protocol agreements
  - IIASA activities projections

Major source groups

- Stationary combustion
    - Power generation
    - Industrial combustion
    - Residential combustion
- Production processes
    - Iron and Steel
    - Aluminium
    - Cement
    - Fertilizers
    - Petroleum refining
    - Beer
    - PVC
    - Sulphuric acid
- Storage and handling
    - Cereals
    - Other products
- Transport
    - Road transport
    - LDV - 4-stroke
    - LDV - 2-stroke
    - HDV
    - Motorised bikes > 50 cc
    - Motorised bikes < 50 cc
    - Non-road transport
- Agriculture
    - Pigs
    - Chickens
- Controlled Municipal Waste incineration

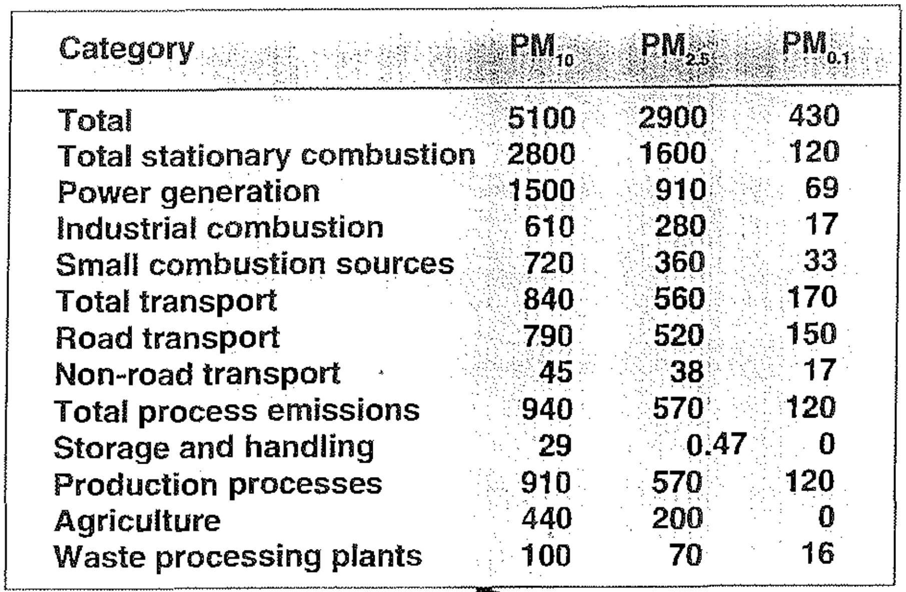
Removal efficiencies for particulate matter emission abatement in Europe

Sector	Fuel type	Western Europe			Eastern Europe		
		Eff10 (%)	Eff2.5 (%)	Eff0.1 (%)	Eff10 (%)	Eff2.5 (%)	Eff0.1 (%)
Power plants	Hard coal	98.9	96.5	95	80	45	0
	Brown coal	97.5	94.5	94.8	80	50	50
	Oil shale	-	-		80	70	0
Industrial combustion	Hard coal	81	79	80	0	0	0
	Brown coal	76	73	65	0	0	0

Caveats

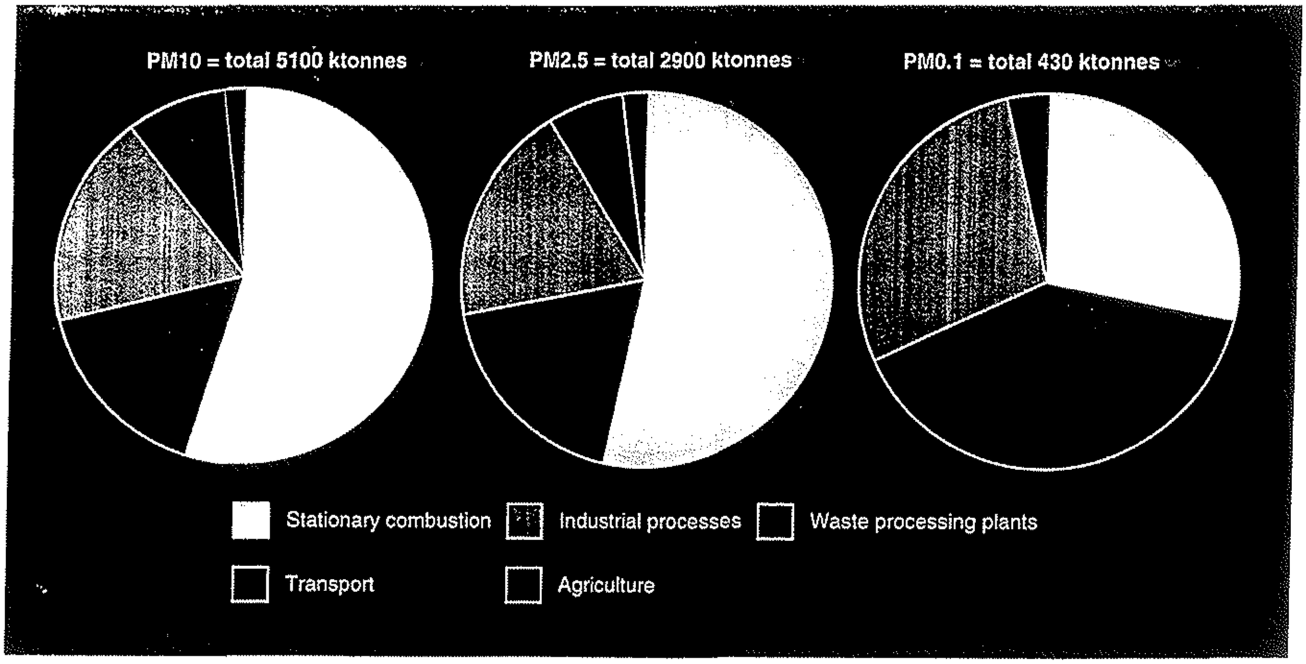
- **Anthropogenic sources**
- **Primary aerosols**
- **Limited resuspension processes involved**
- **Are all relevant sources involved?**
- **Are the sources under/overestimated?**
- **No verification by e.g. inverse modelling**

PM-emissions in Europe for 1990 (ktonnes/year) by source category

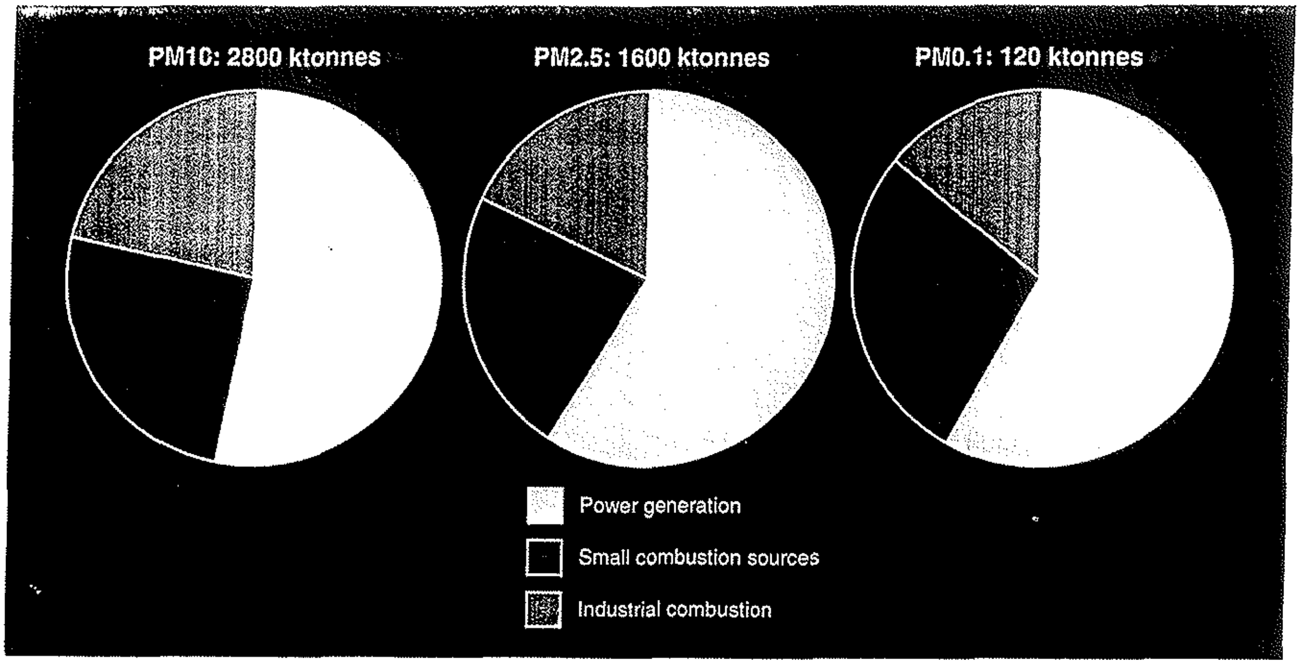


Category	PM_{10}	$PM_{2.5}$	$PM_{0.1}$
Total	5100	2900	430
Total stationary combustion	2800	1600	120
Power generation	1500	910	69
Industrial combustion	610	280	17
Small combustion sources	720	360	33
Total transport	840	560	170
Road transport	790	520	150
Non-road transport	45	38	17
Total process emissions	940	570	120
Storage and handling	29	0.47	0
Production processes	910	570	120
Agriculture	440	200	0
Waste processing plants	100	70	16

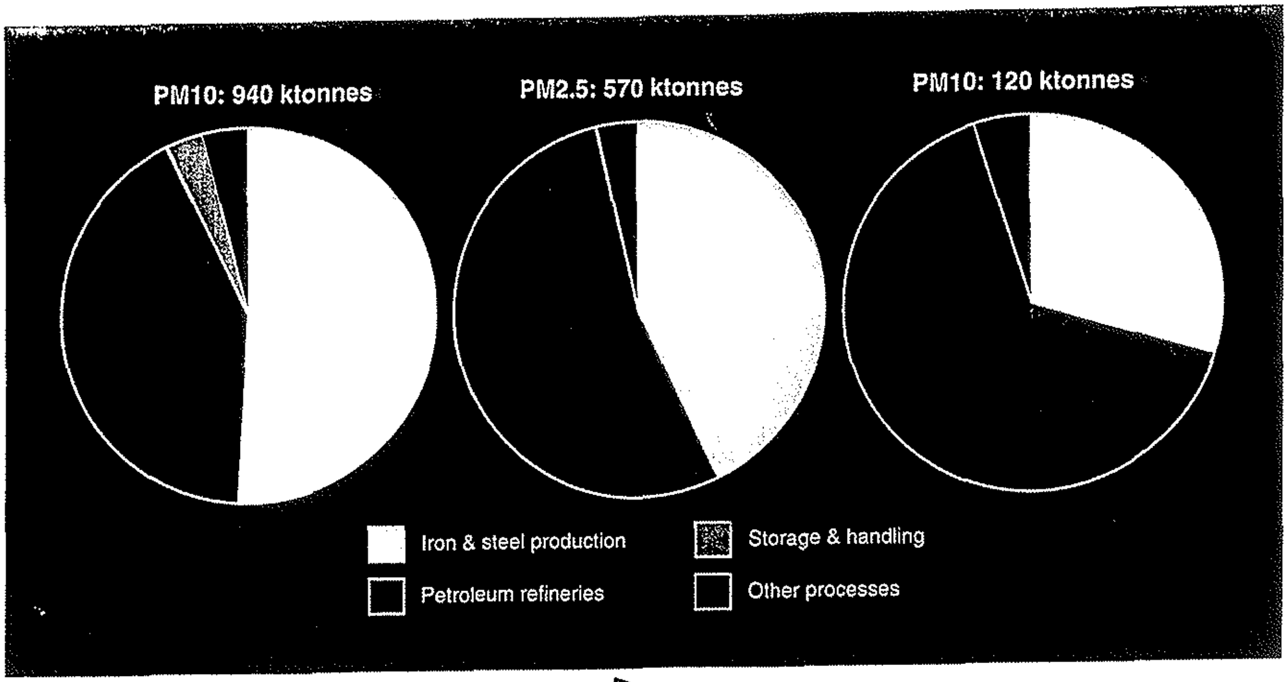
PM-emissions in Europe for 1990 by source category



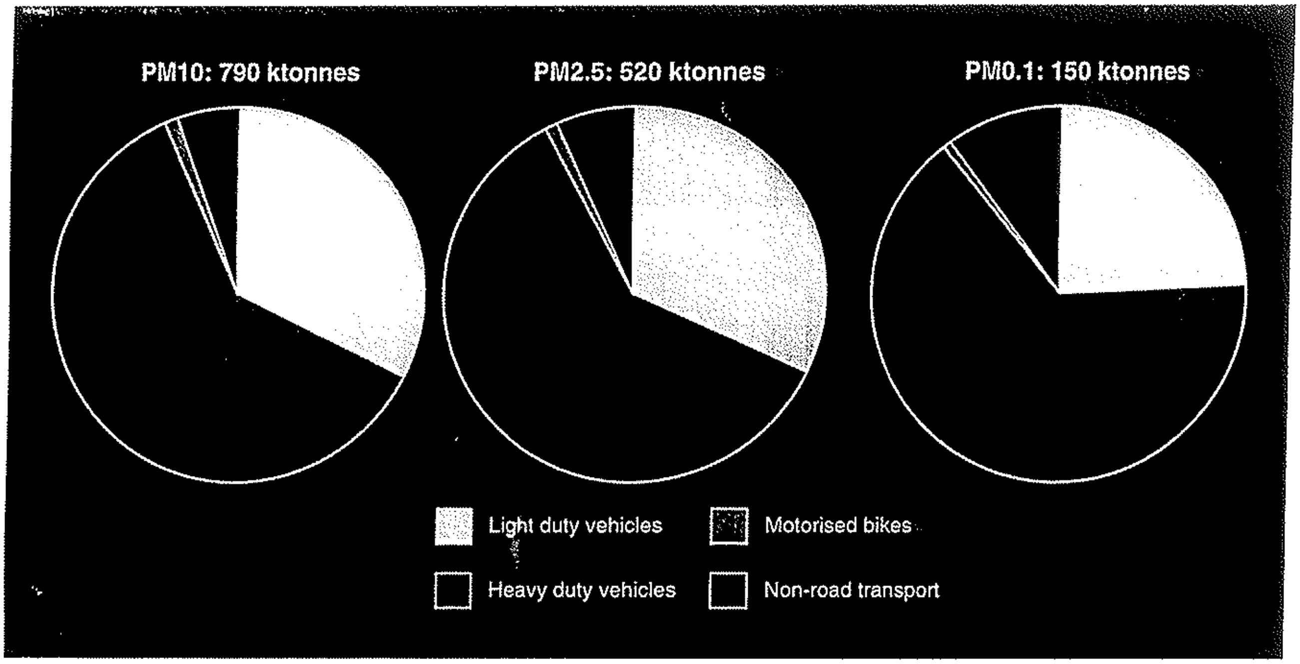
PM-emissions in Europe in 1990 for stationary combustion



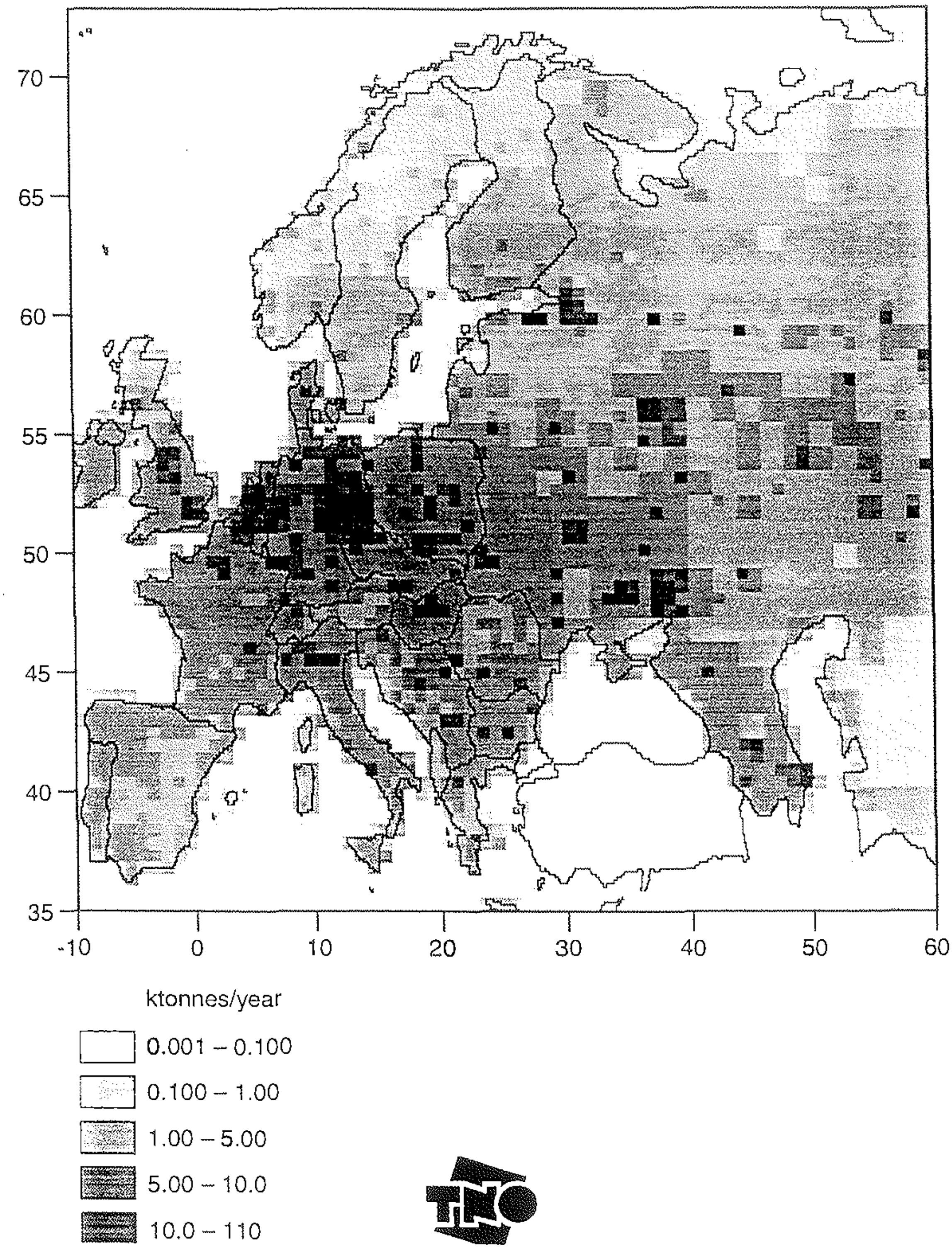
PM-emissions in Europe in 1990 for industrial processes



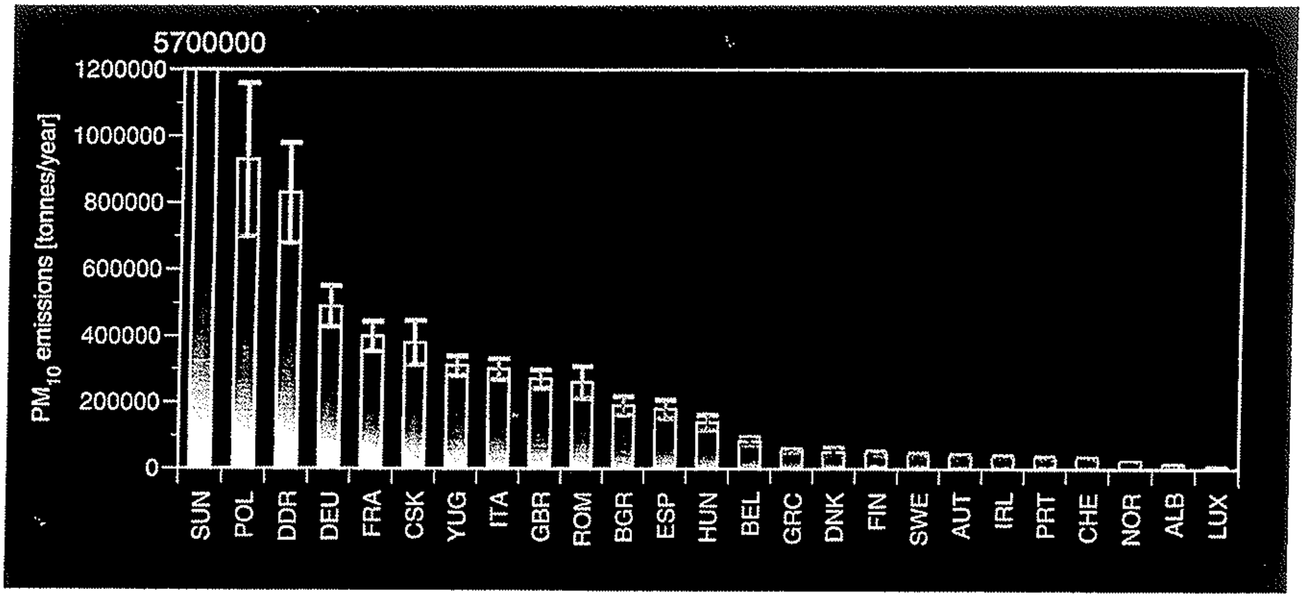
PM-emissions in Europe in 1990 for transport



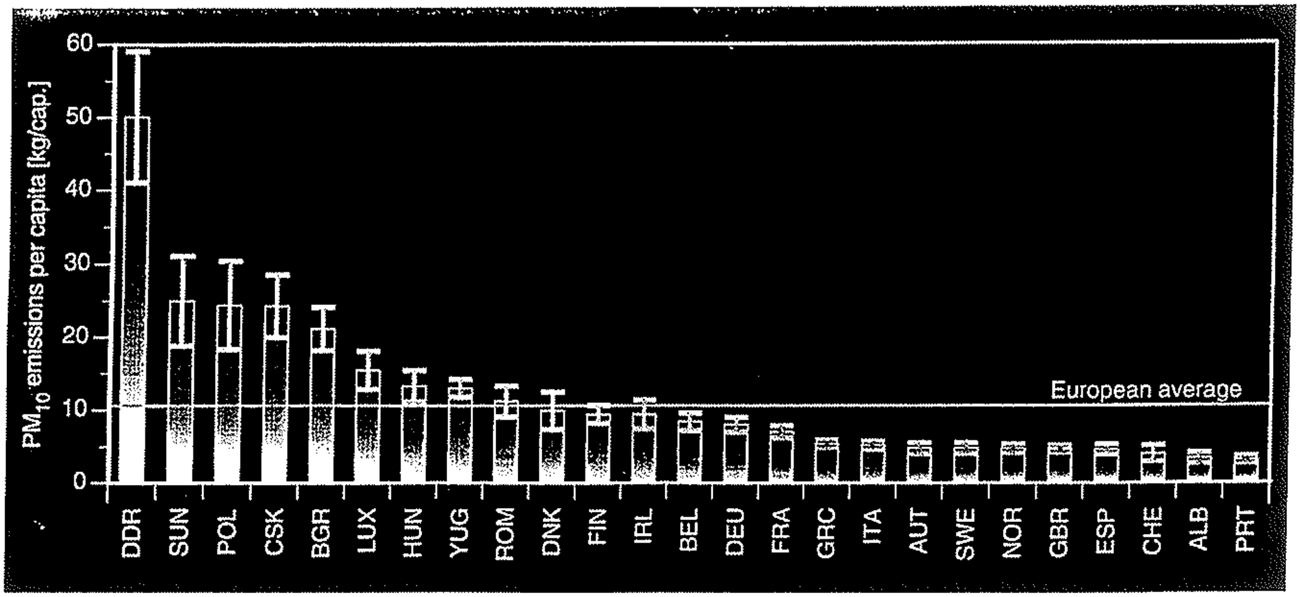
Spatial distribution of PM_{10} emissions in 1990 for Europe on a 0.5x1° grid



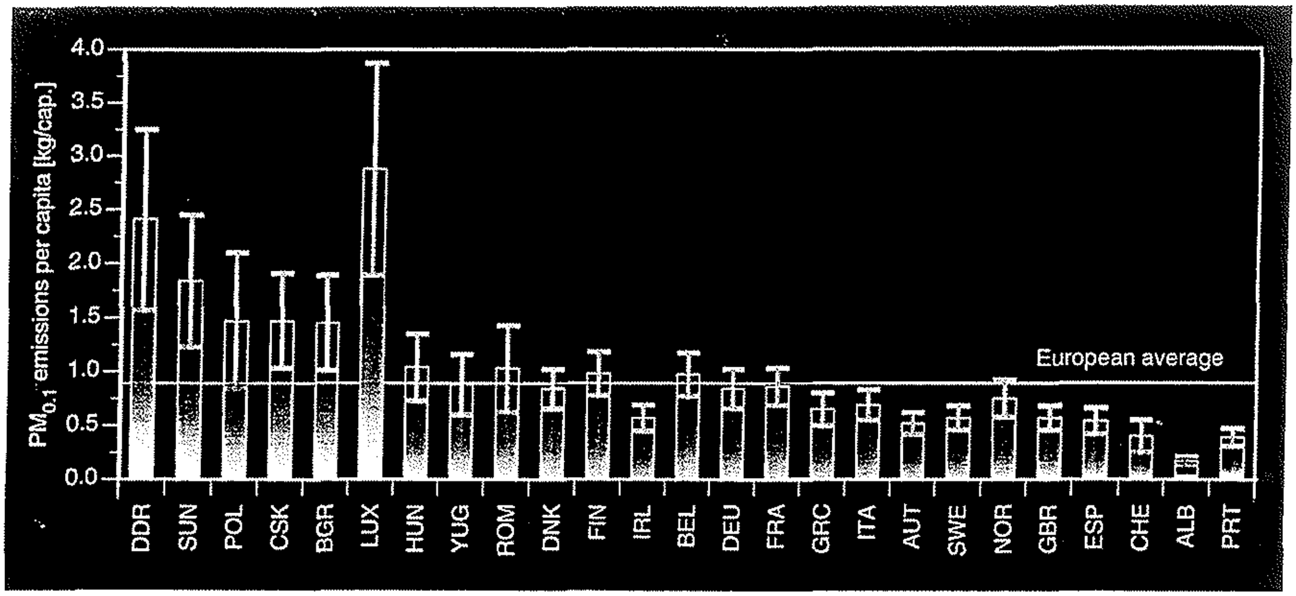
PM_{10}-emissions and standard deviation by country in Europe for 1990



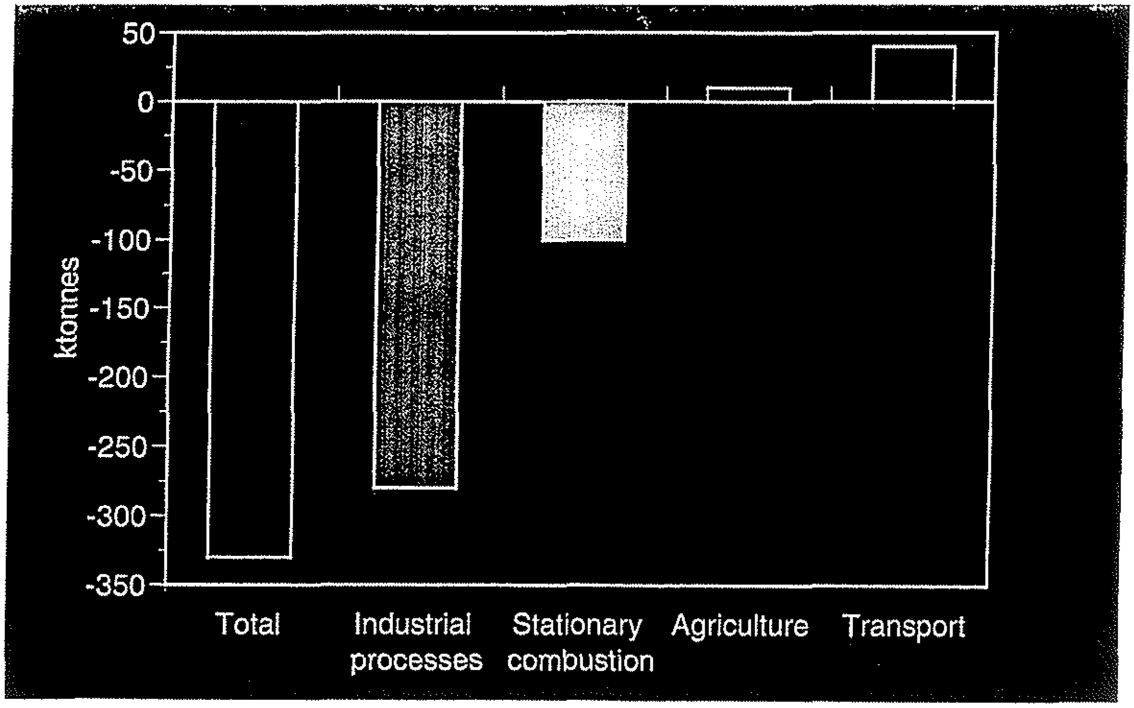
PM_{10}-emissions per capita and standard deviation



PM$_{0.1}$-emissions per capita and standard deviation



Changes in PM10-emisisons in Europe between 1990 and 1993



Emission Limit Values for Stationary Sources - Draft HM Protocol

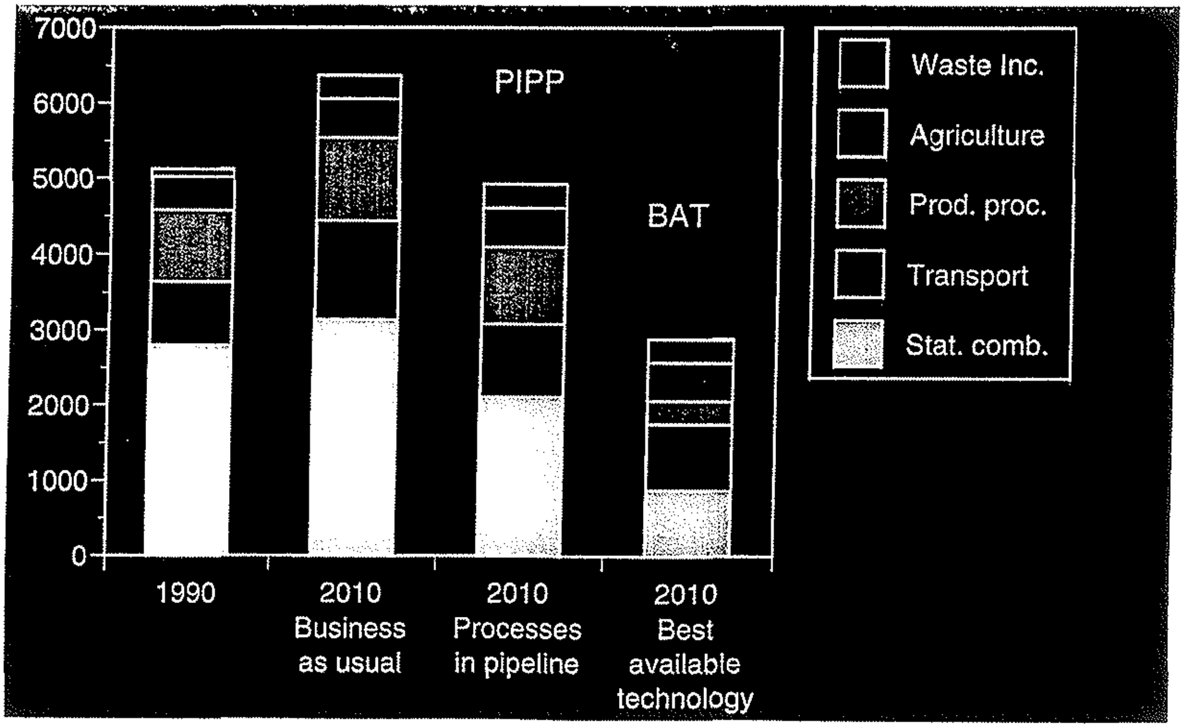
General (independent of source type):
- Cd: 0.2 mg/Nm^3 (mass flow threshold 1 g Cd/h)
- Hg: 0.2 mg/Nm^3 (mass flow threshold 1 g Hg/h)
- Pb: 5 mg/Nm^3 (mass flow threshold 25 g Pb/h)
- Dust: 150 mg/Nm^3 (for mass flows < 0.5 kg dust/h)
- Dust: 50 mg/Nm^3 (for mass flows > 0.5 kg dust/h)

Combustion plants in power generation and industry:
- Dust: 50 mg/Nm^3 (thermal output threshold 50 MWth)

Industrial process emissions:
- Dust, Basic oxygen furnaces in the iron and steel industry: 50 mg/Nm^3
  Dust, Electric arc furnaces in the iron and steel industry: 20 mg/Nm^3
  Dust, Sinter agglomeration plants in the iron and steel industry: 100 mg/Nm^3
  Dust, Cupola furnaces in the iron and steel industry: 20 mg/Nm^3
  Dust, non-Ferrous metals production plants (excl. Lead works): 20 mg/Nm^3
  Dust, Lead works: 10 mg/Nm^3
  Dust, Clinker coolers in cement production: 100 mg/Nm^3
  Dust, Cement grinding in cement production: 75 mg/Nm^3
  Dust, Glass production plants: [illegible] mg/Nm^3
  Hg, Chloro-alkali industry: [illegible] g/Mg Cl_2 production capacity

Waste incinerators:
  Dust, 10 mg/Nm^3
  Cd, 0.05 mg/Nm^3
  Hg, 0.05 mg/Nm^3
  [illegible] mg/Nm^3

PM$_{10}$ emission predictions for Europe in 2010 [ktonnes/year]



Conclusions

- Data set available suitable for modelling and scenario analysis
- Quality
  - Overall uncertainty sources covered
  - Results of RIVM modelling
- Source contribution
- Trends
- Research needs
  - Improvement for e.g. agriculture, traffic
  - Evaluate results with national expertise
  - Addition non-anthropogenic sources
  - Verification of results

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ABSTRACT

Characterization and Source Apportionment of Airborne Particles

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Urban and regional air pollution problems in densely populated areas arise from the combined effects of the emissions from hundreds of different types of air pollution sources. Emissions from stationary fuel combustion sources combine in the atmosphere with the emissions from motor vehicles. To these combustion source effluents are added the emissions from widely dispersed small-scale activities in the community such as emissions from food cooking, and the generation of paved road dust as vehicle traffic moves over the streets. As the emissions from these sources mix in the atmosphere and are transported downwind, atmospheric chemical reactions take place. The secondary air pollutants that are formed by atmospheric chemical reactions include secondary particulate sulfates, nitrates and organics that are formed from the low vapor pressure reaction products of directly emitted pollutant gases. This mixture of directly emitted particles and gases, combined with their secondary atmospheric reaction products can adversely affect human health and damage materials. Fine particle air pollutants scatter and absorb light and thus lead to visibility reduction.

In this presentation, methods for the characterization of fine particle concentrations in the atmosphere and fine particle emissions from sources first will be described. Then air quality modeling methods that can be used to determine how the various emissions sources present in a city combine to produce the observed airborne particle mixture will be discussed. In the source-oriented modeling approach, an atmospheric transport model is used to track pollutant emissions through a simulation of atmospheric fluid motion and chemical reaction as the pollutants are transported from their sources to community air monitoring sites. In the second modeling approach, use of organic compounds that act as tracers for the presence of the effluent from the particular sources that contribute to an an atmospheric sample will be described. From these models, the contributions that specific emissions sources make to ambient fine particle concentrations can be determined in a way that assists the formulation of regional plans for the control of atmospheric particulate matter concentrations. Methods developed will be illustrated through analysis of the causes of the Los Angeles fine particle air pollution problem.

References:

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M. J. Kleeman, A. Eldering, and G. R. Cass. Modeling the Airborne Particle Complex As a Source-Oriented External Mixture, Journal Of Geophysical

Research - Atmospheres 102, 21355-21372, 1997.

M. J. Kleeman and G. R. Cass. Source Contributions to the Size and Composition Distribution of Urban Particulate Air Pollution, Atmospheric Environment, 32, 2803-2816, 1998.

J.J. Schauer, W.F. Rogge, L.M. Hildemann, M.A. Mazurek, G. R. Cass and B.R.T. Simoneit. Source Apportionment of Airborne Particulate Matter Using Organic Compounds as Tracers, Atmospheric Environment, 30 (22), 3837-3855, 1996.

CHARACTERIZATION AND SOURCE APPORTIONMENT OF AIRBORNE PARTICLES

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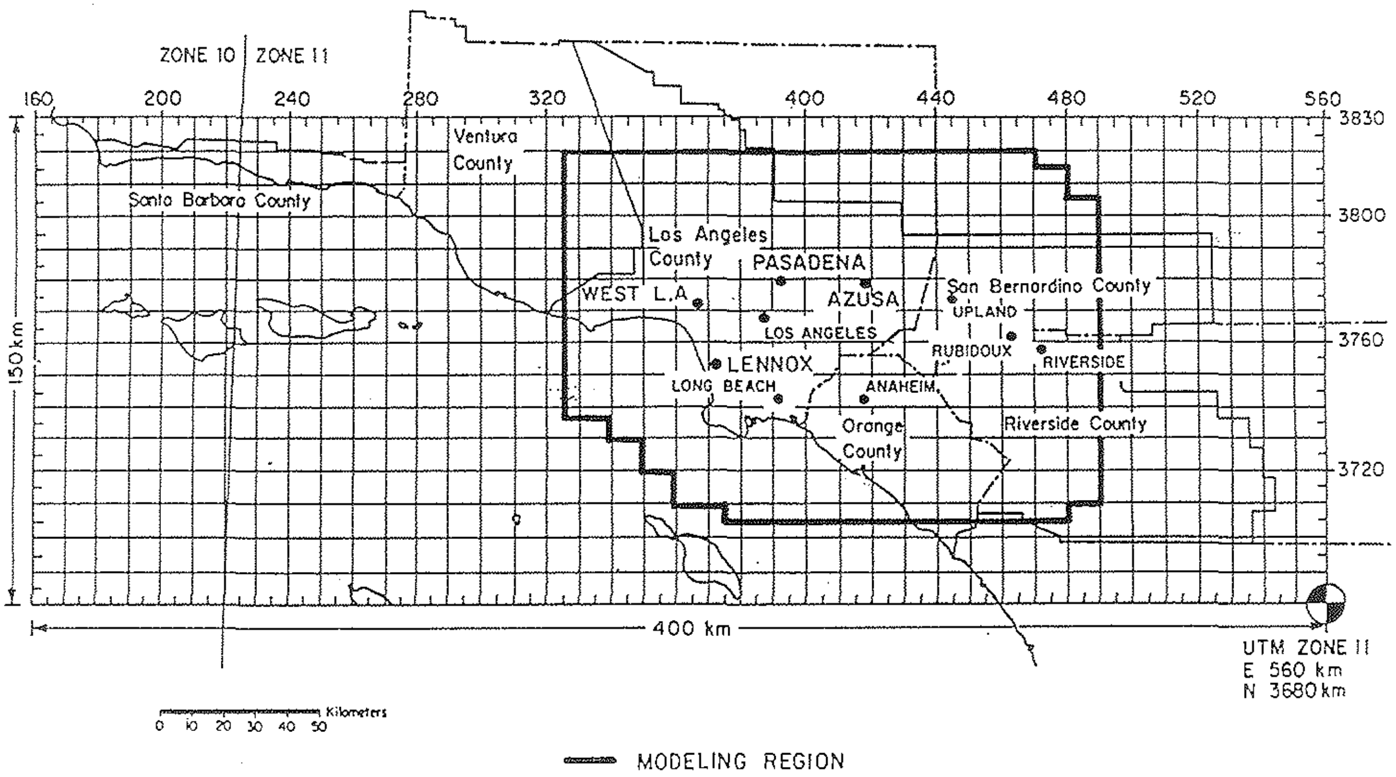
RELATING EMISSIONS TO AIR QUALITY: 2 APPROACHES

- SIMULATION OF ATMOSPHERIC TRANSPORT AND CHEMICAL REACTION
- ORGANIC CHEMICAL TRACER TECHNIQUES

TRANSPORT AND REACTION MODELS

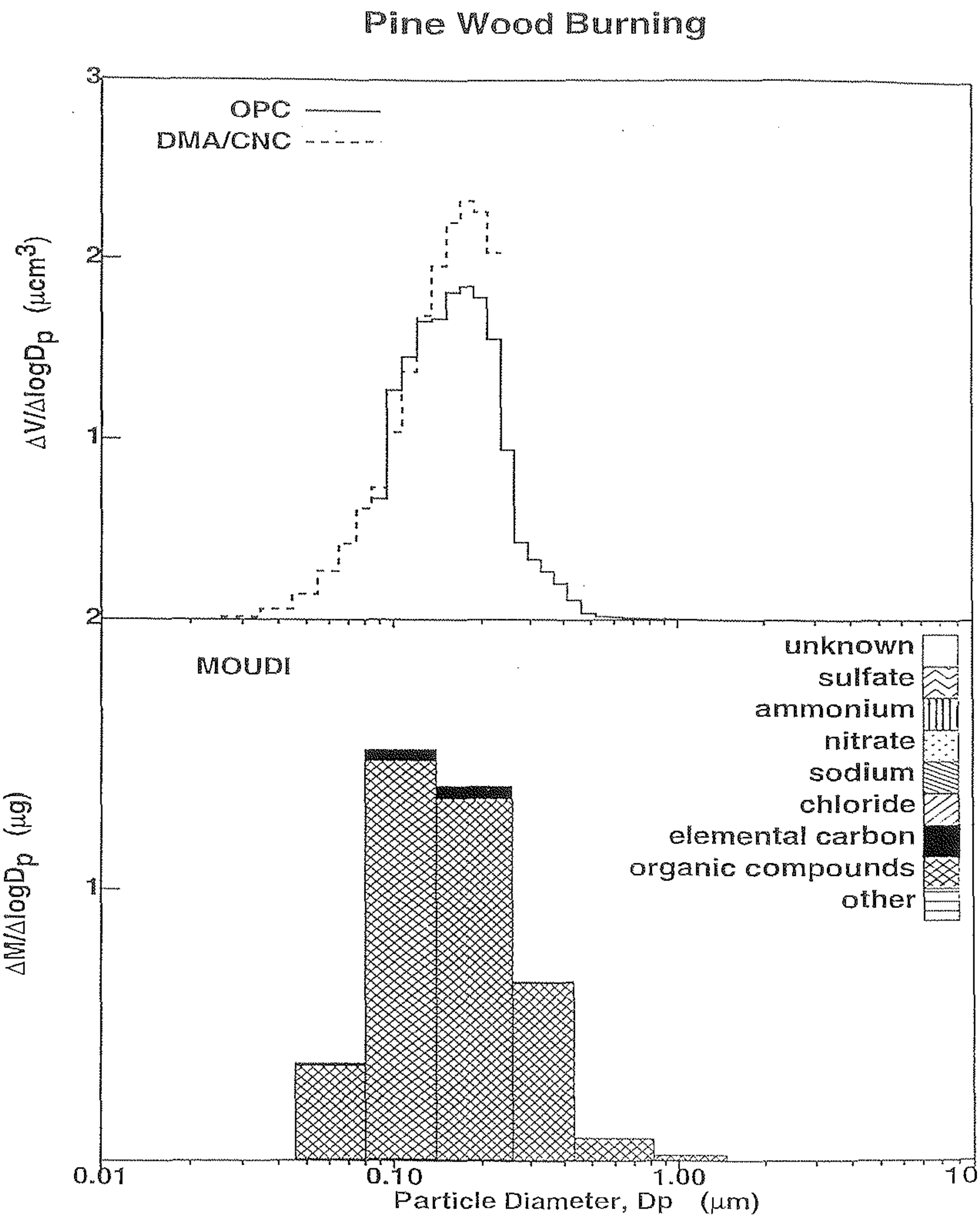
- LAY GRID SYSTEM OVER GEOGRAPHIC AREA
- MEASURE SIZE DISTRIBUTION AND CHEMICAL COMPOSITION OF PARTICLE EMISSIONS AS WELL AS REACTIVE POLLUTANT GASES
- TRACK PARTICLES AS THEY ARE TRANSPORTED BY THE WIND AND AS THEY ACCUMULATE COATINGS OF ATMOSPHERIC REACTION PRODUCTS
    - SULFATE AND NITRATE SALTS
    - SECONDARY ORGANIC COMPOUNDS
- COMPUTE ATMOSPHERIC CONCENTRATIONS
- COMPUTE SOURCE CONTRIBUTIONS TO ATMOSPHERIC CONCENTRATIONS

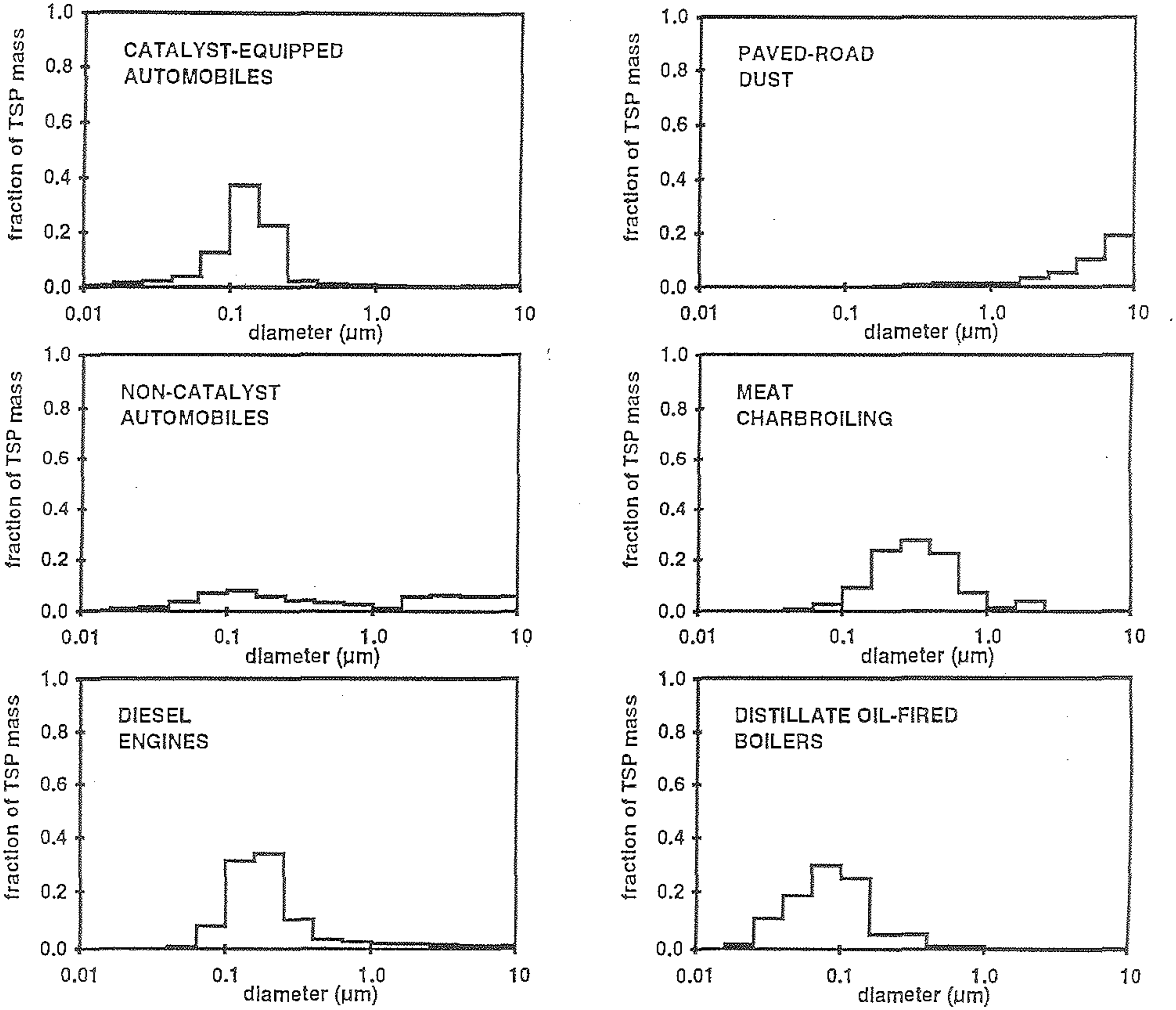
GRID SYSTEM FOR LOS ANGELES MODELING AREA



Sources Tested

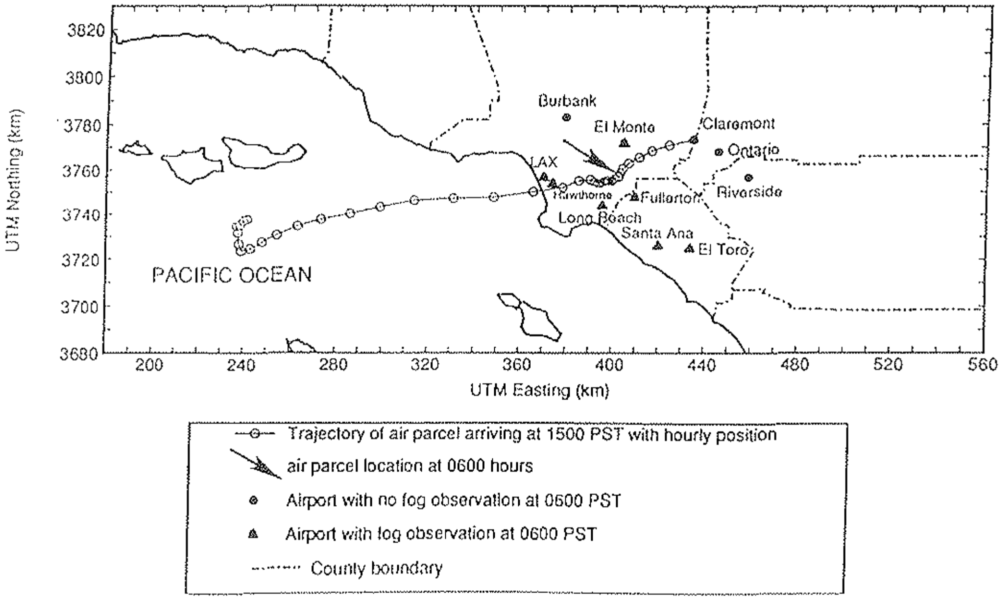
- Catalyst-Equipped Autos
- Non-catalyst Autos
- Diesel Trucks
- Fireplace Combustion of Wood
- Oil-Fired Boilers
- Meat Charbroilers
- Natural Gas Home Appliances
- Roofing Tar Pots
- Cigarette Smoke
- Tire Dust
- Brake Dust
- Paved Road Dust
- Vegetative Detritus

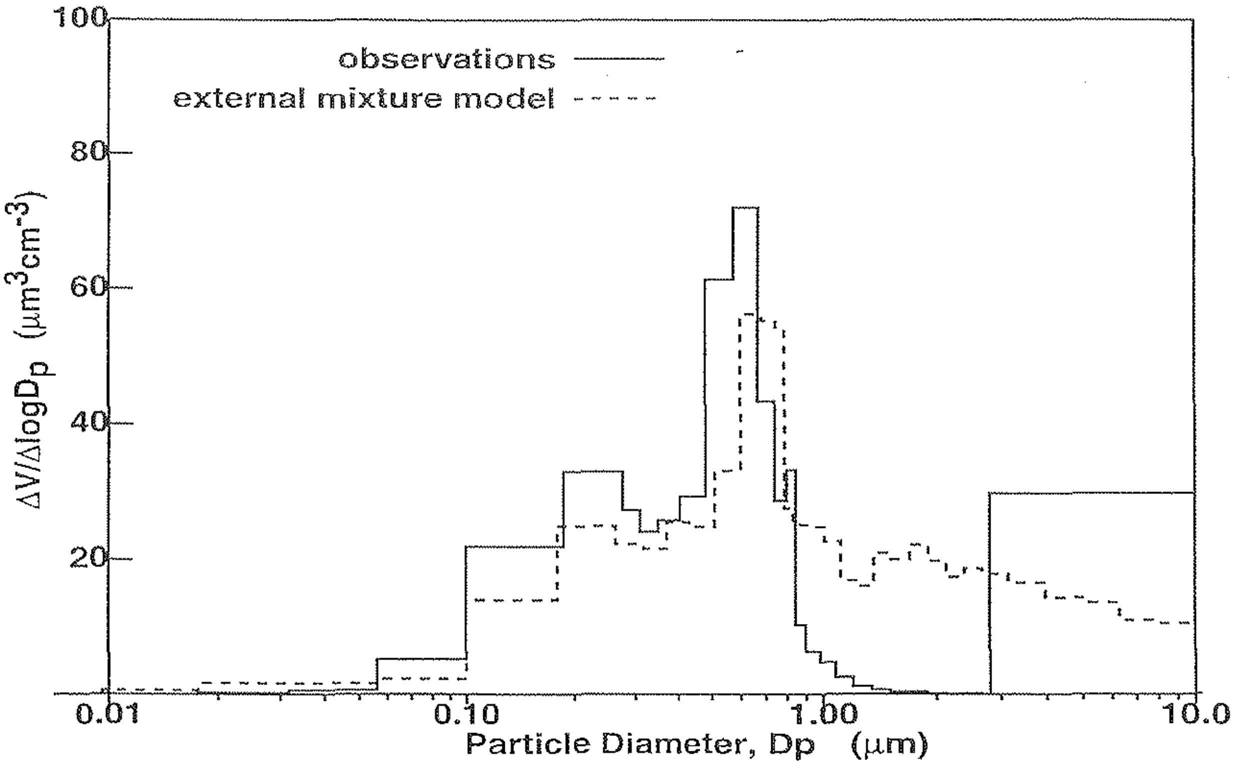

Pine Wood Burning
OPC
DMA/CNC
ΔV/ΔlogDp (μcm³)
3
2
1
2
MOUDI
unknown
sulfate
ammonium
nitrate
sodium
chloride
elemental carbon
organic compounds
other
ΔM/ΔlogDp (μg)
1
0.01
0.10
1.00
10.0
Particle Diameter, Dp (μm)

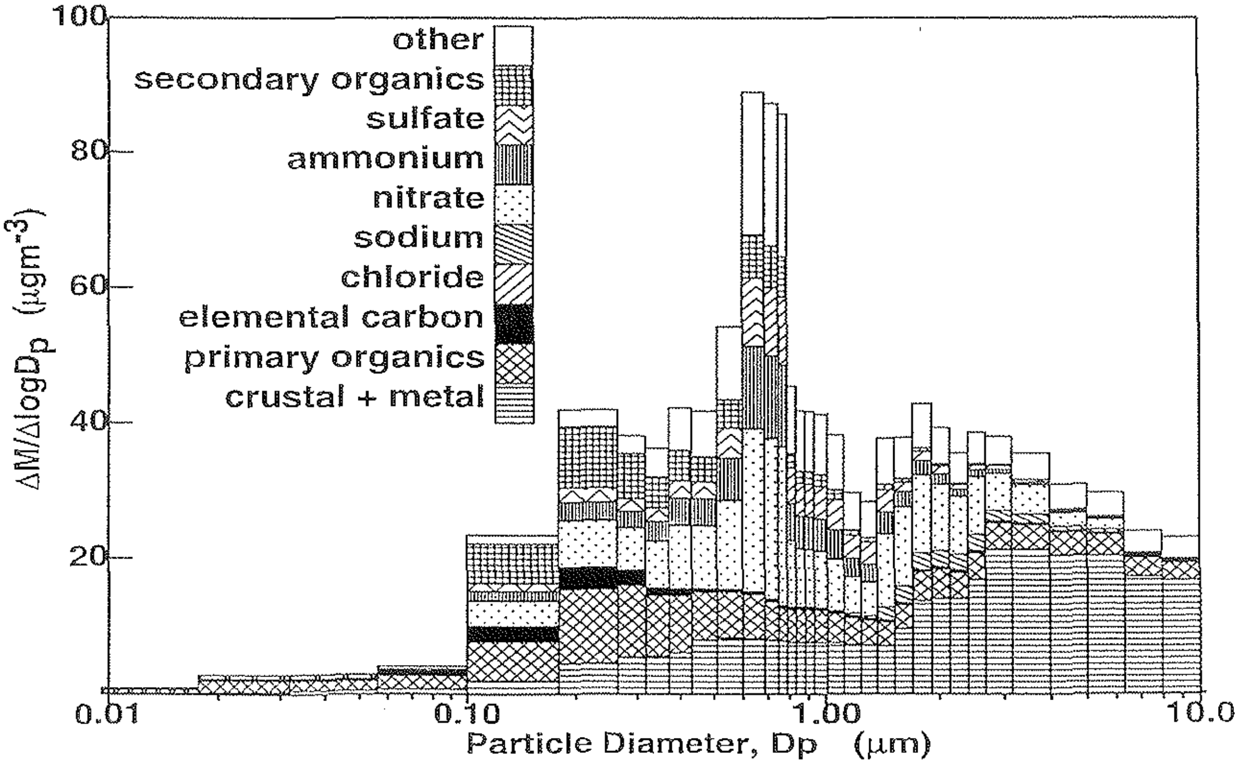

CATALYST-EQUIPPED AUTOMOBILES
PAVED-ROAD DUST
NON-CATALYST AUTOMOBILES
MEAT CHARBROILING
DIESEL ENGINES
DISTILLATE OIL-FIRED BOILERS
fraction of TSP mass
0.0
0.2
0.4
0.6
0.8
1.0
diameter (µm)
0.01
0.1
1.0
10

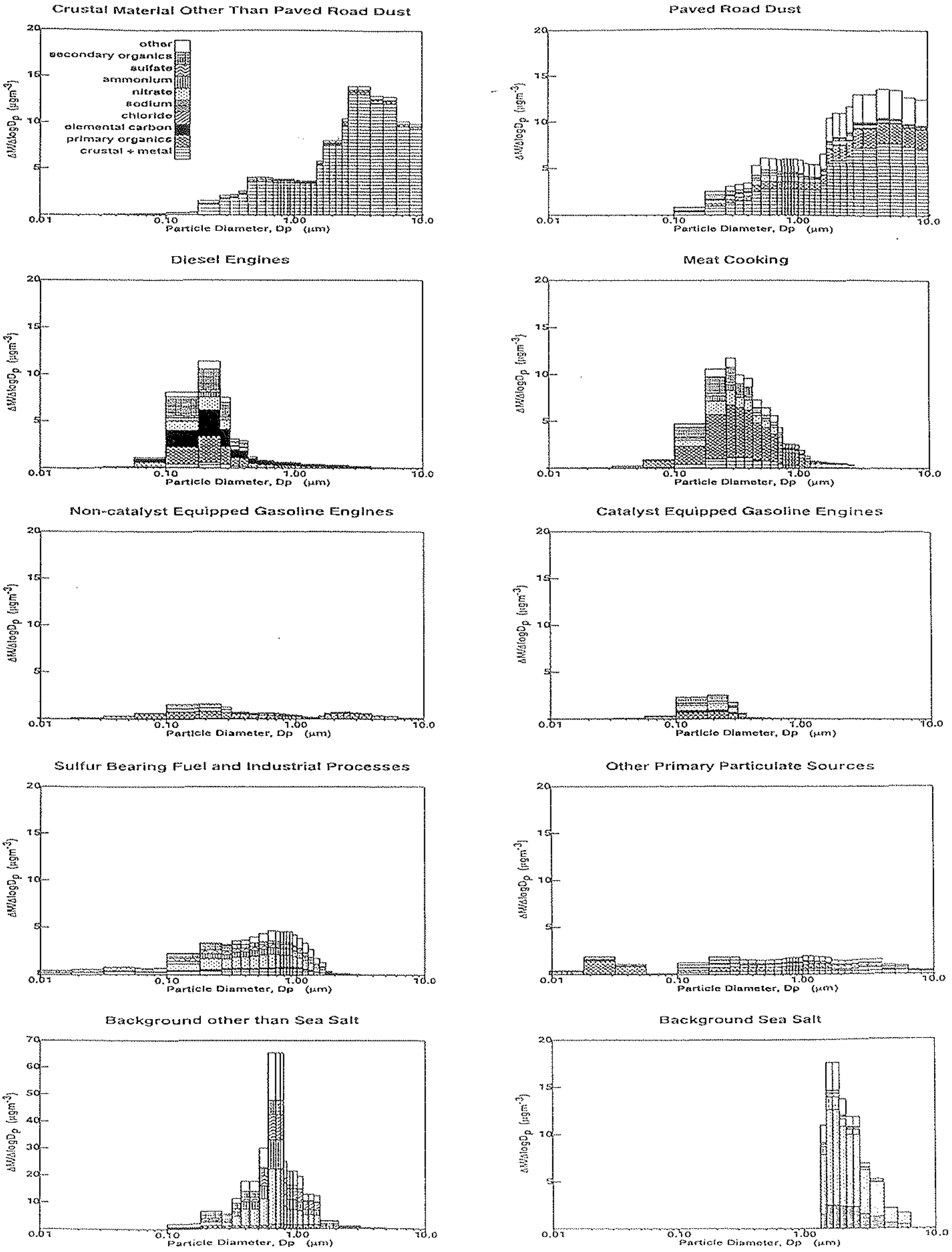
Aerosol Formation and Transport

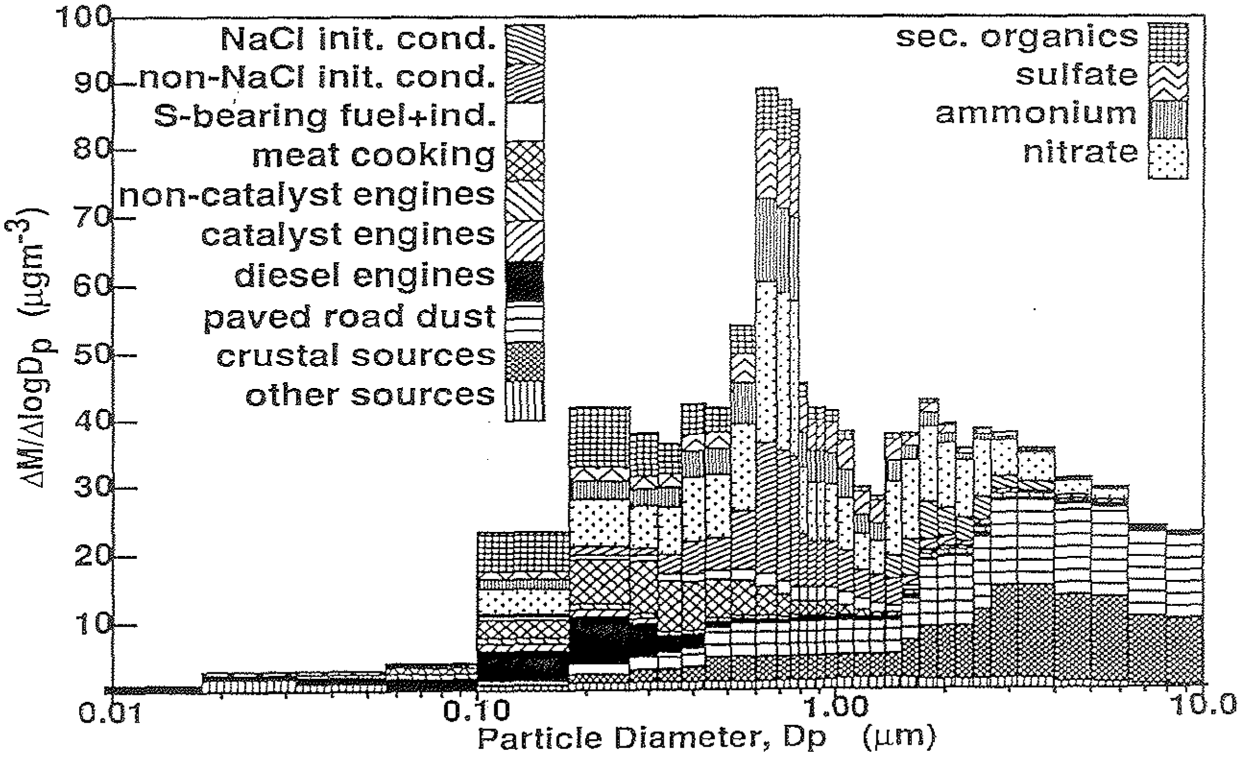
- photochemical trajectory model - Russell *et al.*
- 15 discrete particle sizes that grow by condensation or incorporation into fog
- gas phase chemistry - Carter, 1990
- aerosol thermodynamics and diffusion of condensable secondary species to and from particles - Wexler *et al.*
- fog module
- model tracks reactive gases, primary aerosol, sea salt, secondary sulfates, nitrates, and organics




observations
external mixture model
100
80
60
40
20
$\Delta V/\Delta \log D_p$ ($\mu m^3 cm^{-3}$)
0.01
0.10
1.00
10.0
Particle Diameter, Dp (μm)


other
secondary organics
sulfate
ammonium
nitrate
sodium
chloride
elemental carbon
primary organics
crustal + metal
100
80
60
40
20
$\Delta M/\Delta \log D_p$ ($\mu g m^{-3}$)
0.01
0.10
1.00
10.0
Particle Diameter, Dp (μm)


Crustal Material Other Than Paved Road Dust
Paved Road Dust
Diesel Engines
Meat Cooking
Non-catalyst Equipped Gasoline Engines
Catalyst Equipped Gasoline Engines
Sulfur Bearing Fuel and Industrial Processes
Other Primary Particulate Sources
Background other than Sea Salt
Background Sea Salt
other
secondary organics
sulfate
ammonium
nitrate
sodium
chloride
elemental carbon
primary organics
crustal + metal
ΔM/ΔlogDp (μgm-3)
Particle Diameter, Dp (μm)
0.01
0.10
1.00
10.0
0
5
10
15
20
30
40
50
60
70


NaCl init. cond.
non-NaCl init. cond.
S-bearing fuel+ind.
meat cooking
non-catalyst engines
catalyst engines
diesel engines
paved road dust
crustal sources
other sources
sec. organics
sulfate
ammonium
nitrate
ΔM/ΔlogDp (μgm-3)
100
90
80
70
60
50
40
30
20
10
0.01
0.10
1.00
10.0
Particle Diameter, Dp (μm)

MOLECULAR TRACER TECHNIQUES

- SEEK SINGLE COMPOUNDS OR COMPOUND GROUPS THAT ARE
  - CHARACTERISTIC OF A SOURCE
  - RELATIVELY STABLE

PROCEDURE:

- Characterize ambient pollutant concentrations
  - Air monitoring network
  - Bulk elemental analysis
  - Organic chemical composition by GC/MS

- Characterize major emissions sources
  - Use dilution source sampler
  - Measure 15 most important particle source types
  - Organic chemical analysis by GC/MS

- Compute linear combination of source effluents needed to reproduce distribution of particulate organic compounds in the atmosphere

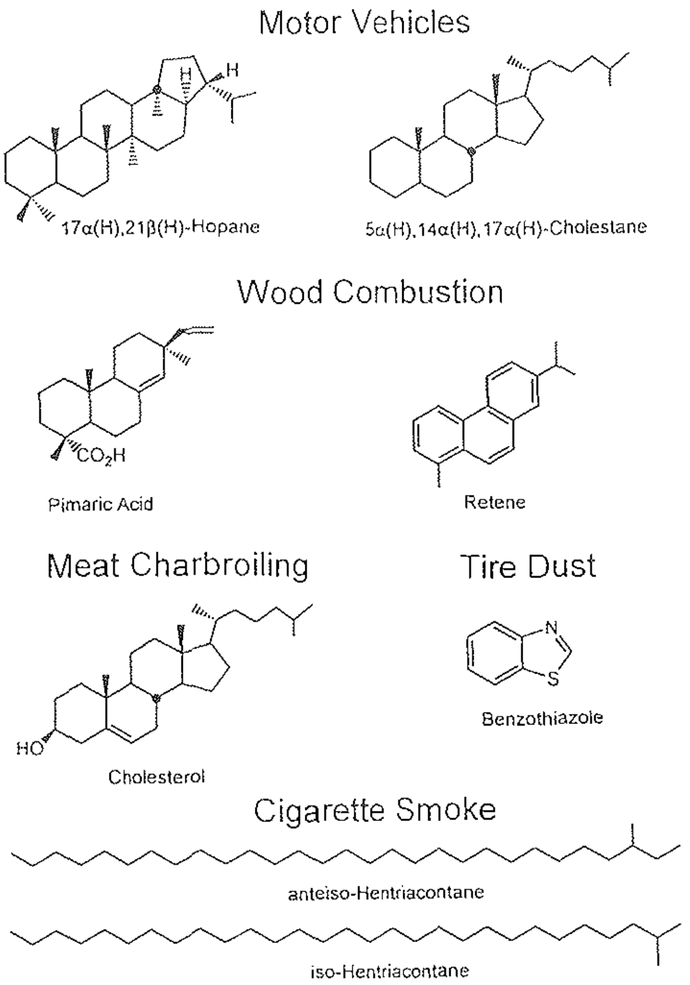
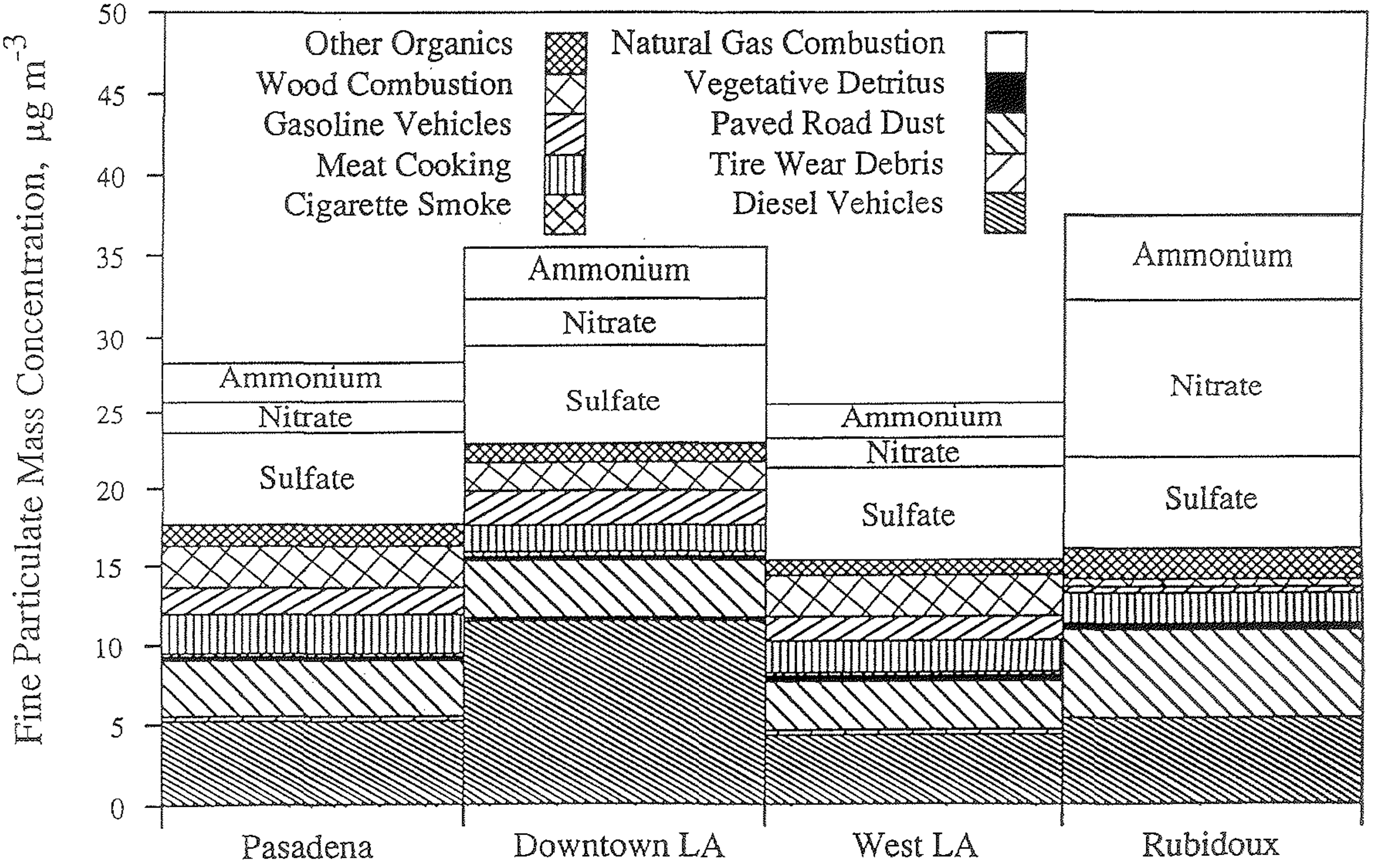
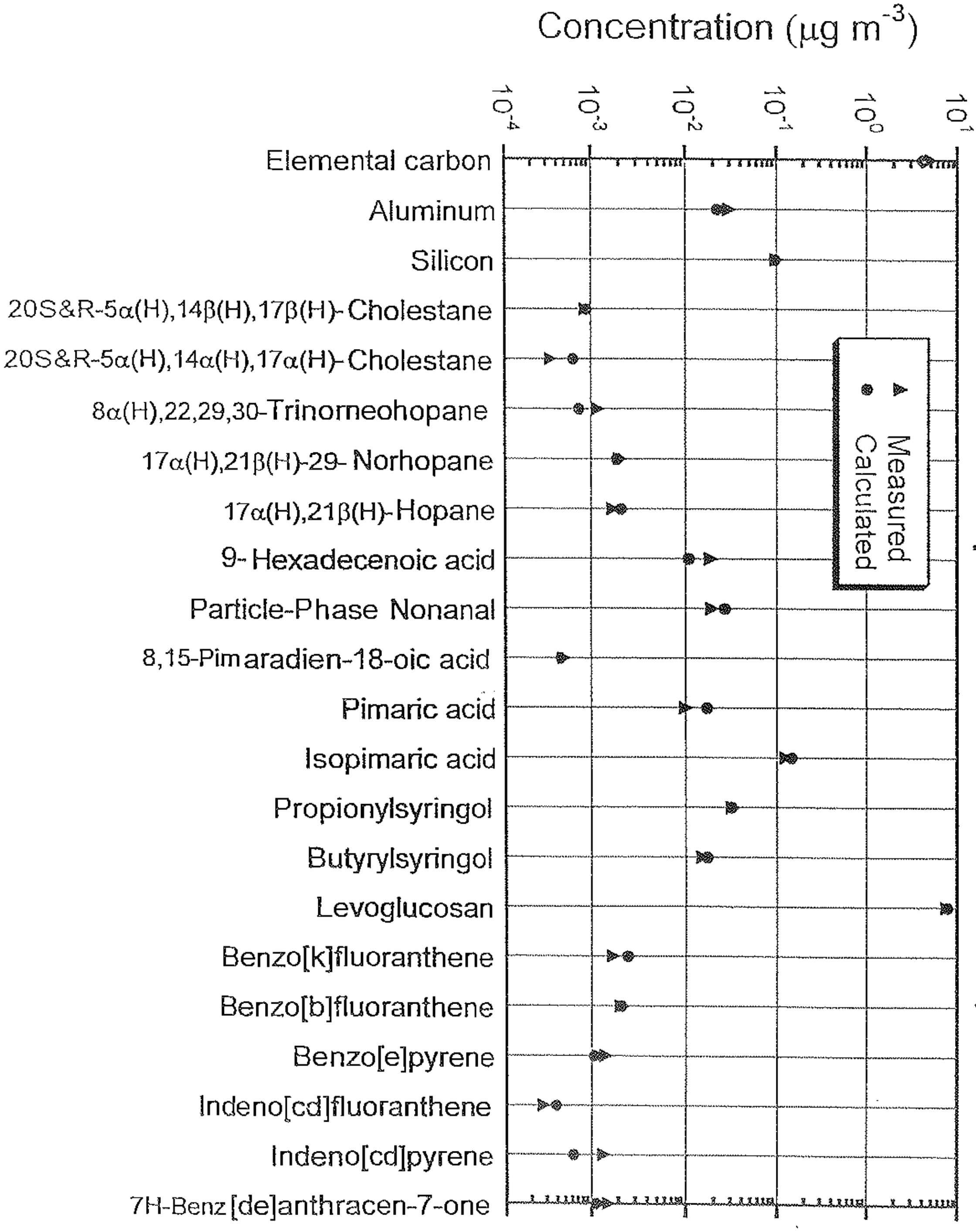


Fig. 1. The chemical structures of some useful organic molecular tracers for particulate air pollution sources.

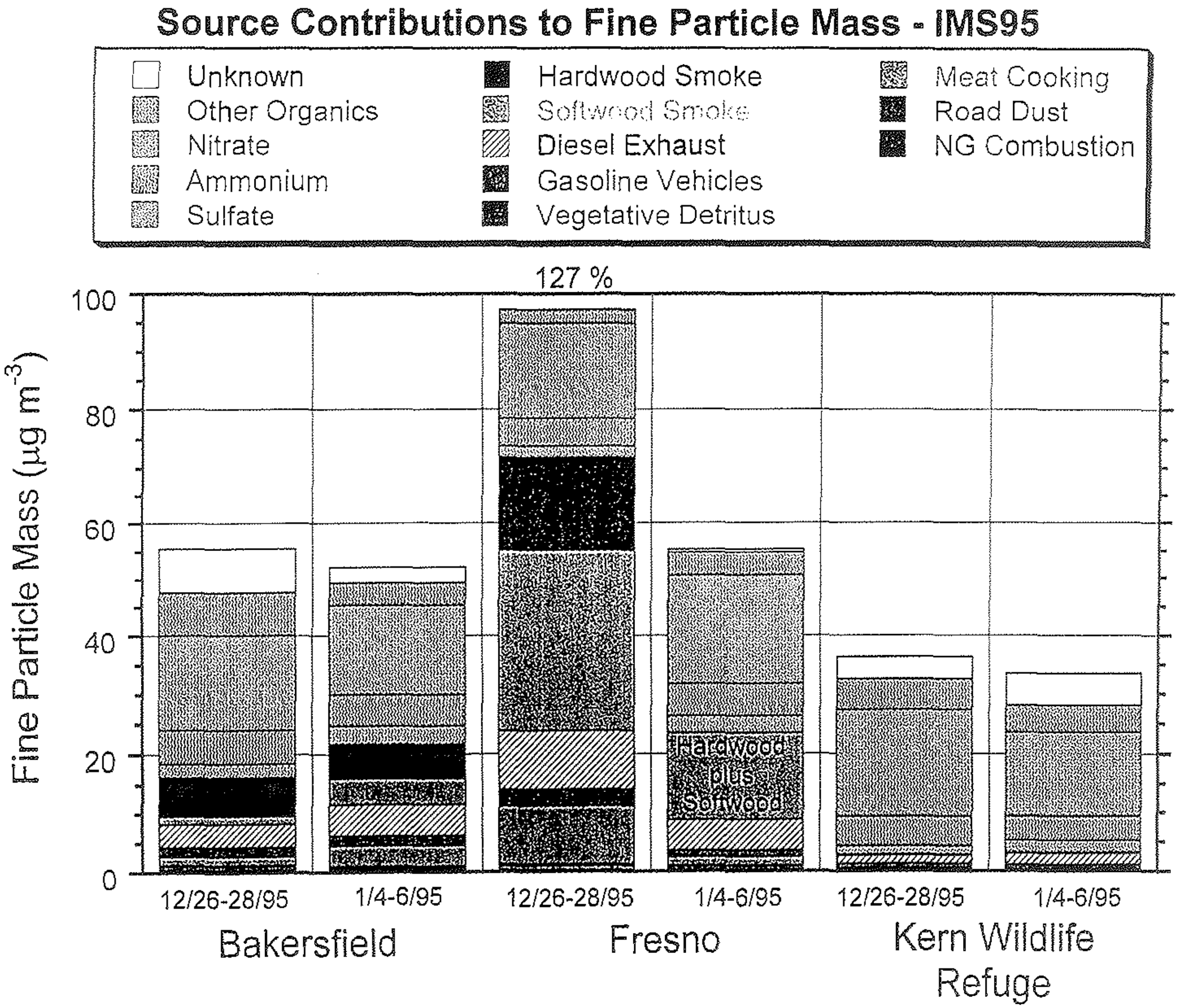
Airborne particulate matter



Source apportionment of fine mass concentrations — annual average.



Comparison of Calculated and Measured Concentrations of Mass Balance Species - Fresno Dec 26-28, 1995


Source Contributions to Fine Particle Mass - IMS95
Unknown
Other Organics
Nitrate
Ammonium
Sulfate
Hardwood Smoke
Softwood Smoke
Diesel Exhaust
Gasoline Vehicles
Vegetative Detritus
Meat Cooking
Road Dust
NG Combustion
127 %
Hardwood plus Softwood
Fine Particle Mass (µg m-3)
100
80
60
40
20
0
12/26-28/95
1/4-6/95
12/26-28/95
1/4-6/95
12/26-28/95
1/4-6/95
Bakersfield
Fresno
Kern Wildlife Refuge

What Is Known About the Health Effects of PM?

Chair: *Ross Anderson,* St. George's Hospital Medical School,
United Kingdom

BERT BRUNEKREEF

University of Wageningen, The Netherlands

SUMMARY OF THE RESULTS OF EPIDEMIOLOGIC STUDIES OF ACUTE AND LONG-TERM EFFECTS OF PM

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A large number of studies have documented effects of short-term exposures to particulate matter in ambient air on mortality and morbidity. These studies have led regulatory agencies such as the U.S. EPA and the E.U. to promulgate new, tight air quality standards.

Effects of day-to-day variations in PM concentrations on day-to-day variations in mortality have been frequently reported. Effects of respirable particles (PM2.5) seem stronger than effects of the coarse fraction, consisting of particles between 2.5 and 10 µm diameter, of inhalable particles (PM10). Studies from Europe, notably APHEA (Air pollution and health, a European approach) have documented effects of other measures of ambient air pollution as well, notably SO_2, Black Smoke and ozone. These studies have also documented rather strong differences in effects between the warm and the cold season, effects in the warm season being usually larger. A restriction of many European studies is the absence of measured concentrations of PM10, PM2.5 or specific components such as sulfates. A new study from the Netherlands had three years of nationwide data for PM10 in addition to data on sulfate, nitrate and gaseous components. Effects on mortality were found to vary by season, with strongest effects being observed again in the warm season. The clearest effects were found for ozone. In addition, measures of fine particulate matter (sulfate and nitrate) were also associated with mortality, more so than PM10.

Potential confounding by weather variables was investigated in recent years by two different research groups in great detail. In both studies, the effects of air pollution on mortality did not change after more elaborate control for weather variables.

There are fewer studies of effects of long-term exposure. Two cohort studies from the U.S. have suggested that at low concentrations of PM2.5 or sulfate, effects on cardio-respiratory survival are present. A new study from California that came out in the beginning of 1999 supported the findings of the earlier two studies to some extent, but there were also notable differences, for example a stronger effect on lung cancer, and a weaker effect on cardiorespiratory mortality. New cohort studies seem needed to resolve effects of current, long-term exposures to PM on survival.

Epidemiology of acute and long term PM effects on health

Bert Brunekreef PhD

Studying health effects of air pollution

- *Epidemiology*
  - **strength**: studies real world
  - **weakness**: observational mostly
- *Controlled human exposures*
  - **strength**: humans, experimental control
  - **weakness**: limited in effects to be studied
- *Animal and in vitro experiments*
  - **strength**: full experimental control
  - **weakness**: removed from real-world situation

Causal inference from epidemiology

- Hill AB. *The environment and disease: association or causation?* Proc Royal Soc Med 1965; 295-300
- 'Criteria':
  - temporality (cause precedes effect)
  - consistency (finding is repeatable)
  - coherence (with animal experiments etc.)
  - biological gradient (dose-response relationship)

Epidemiologic studies

- *Time series*: day-to-day variation of air pollution and health indicators: acute effects
- *Cohort studies*: incidence of chronic disease and death in well-defined populations with different air pollution exposures
- *Cross-sectional studies*: concurrent measurement of health and pollution exposures

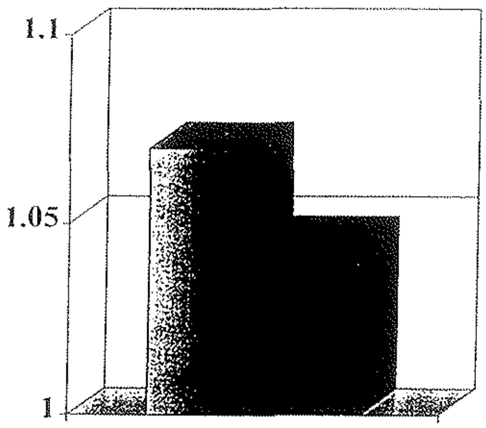
Basic Epidemiology......

	Disease	No disease
Exposed	A	B
Unexposed	C	D

Relative Risk: $\dfrac{A / (A + B)}{C / (C + D)}$

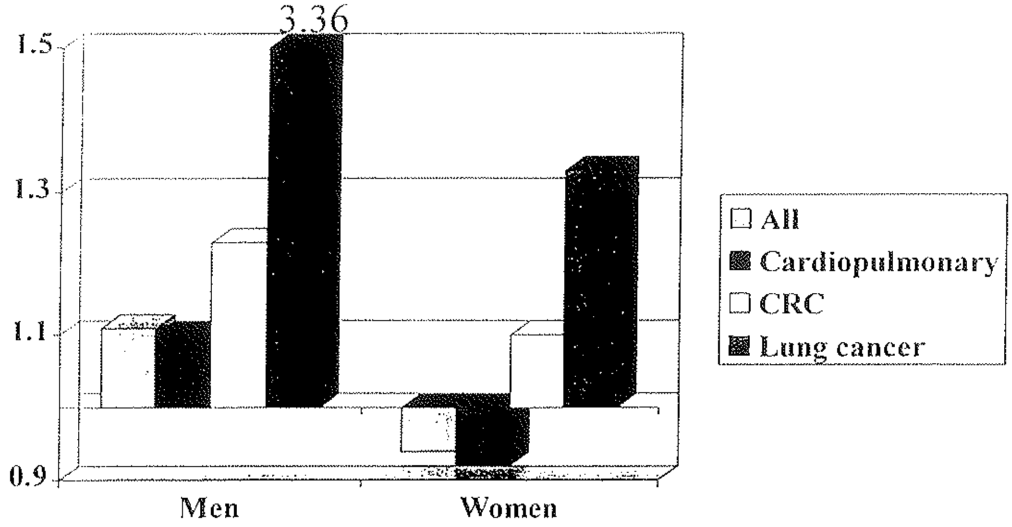
Presentation of Relative Risks (RRs):

- Bar graphs
- RRs per Interquartile Range (IQR), or typical range, or 5-95 percentiles
- Shading in bars indicates statistical significance

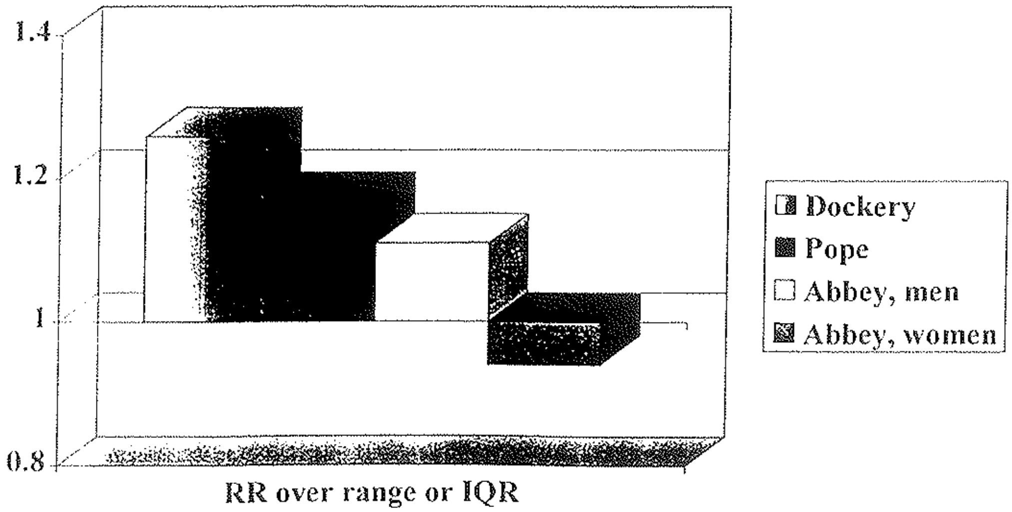


Abbey et al., AJRCCM 1999

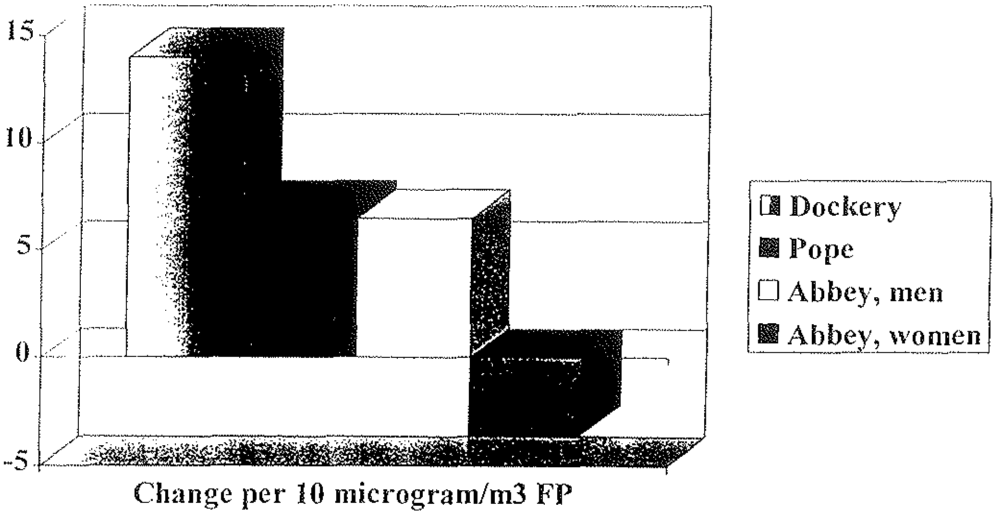
Relative mortality Risks per 24 $\mu g/m^3$ PM10 (IQR)



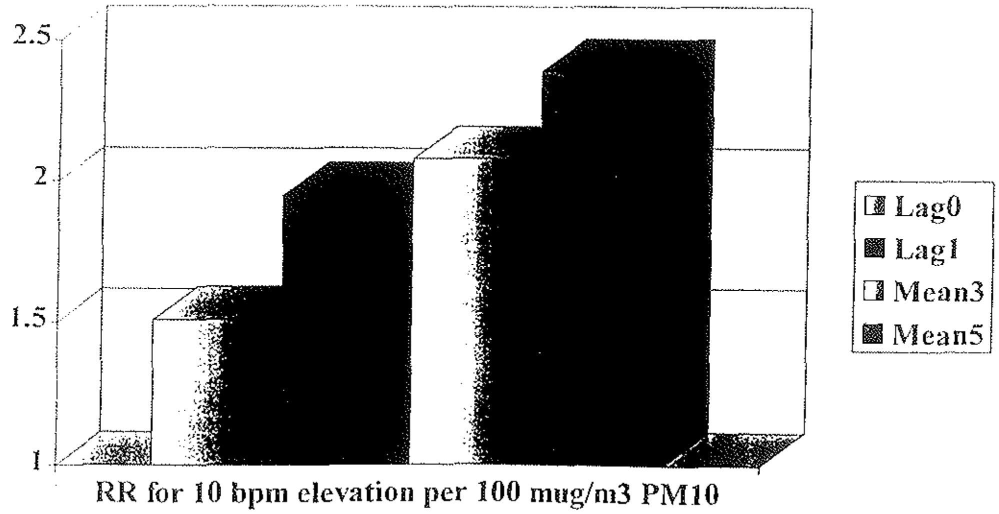
US Cohort studies PM and all-cause mortality



US Cohort studies
PM and all-cause mortality



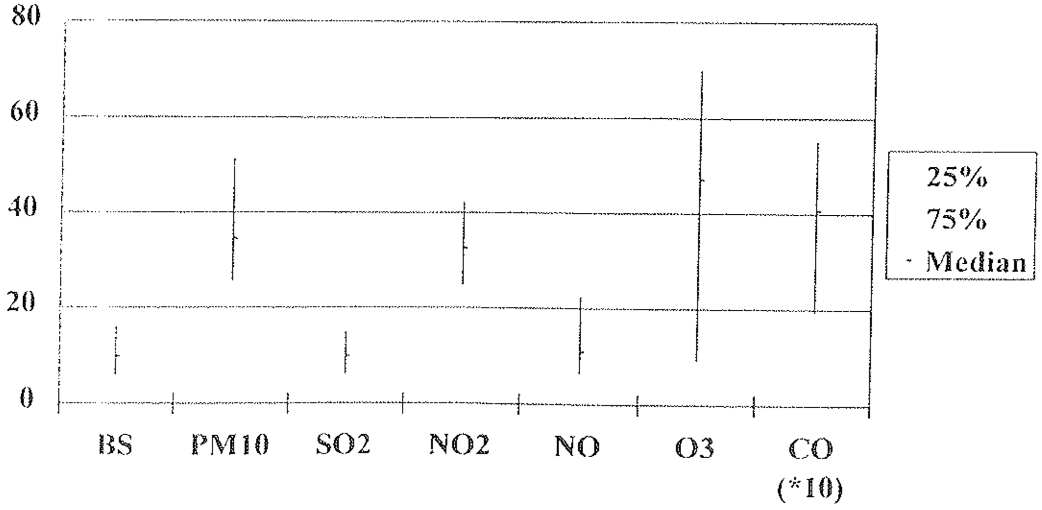
Pope et al., AJRCCM 1999
PM10, oxygen saturation and pulse rate



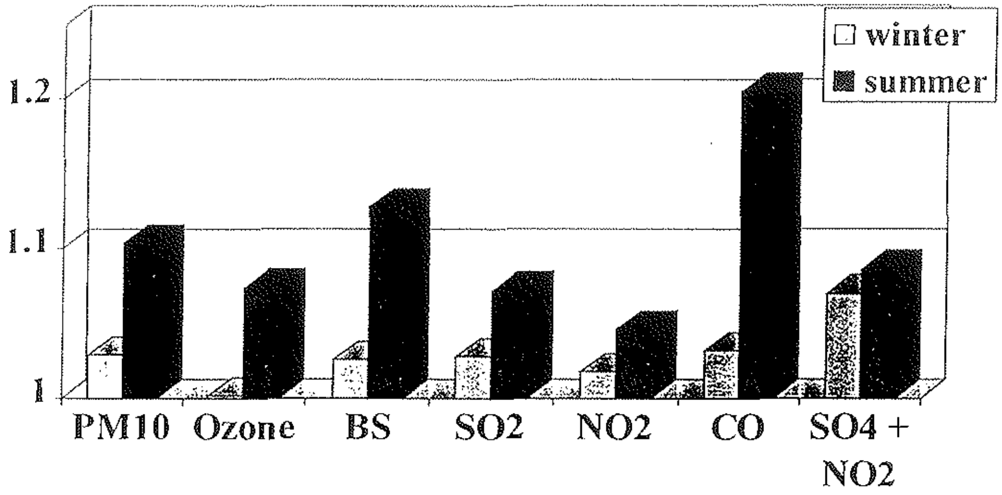
Dutch short-term mortality study

- Period 1986-1994
- Approximately 1,090,000 deaths
- Data on BS, SO_2, NO_2, NO, CO, O_3 (all years), PM10, sulfate, nitrate (1992-1994)
- Adjustment for influenza, pollen counts
- OR's per 25 μg/m³ (sulfate/nitrate), 50 (SO_2, NO_2, BS), 100 (PM10), 150 (O_3)

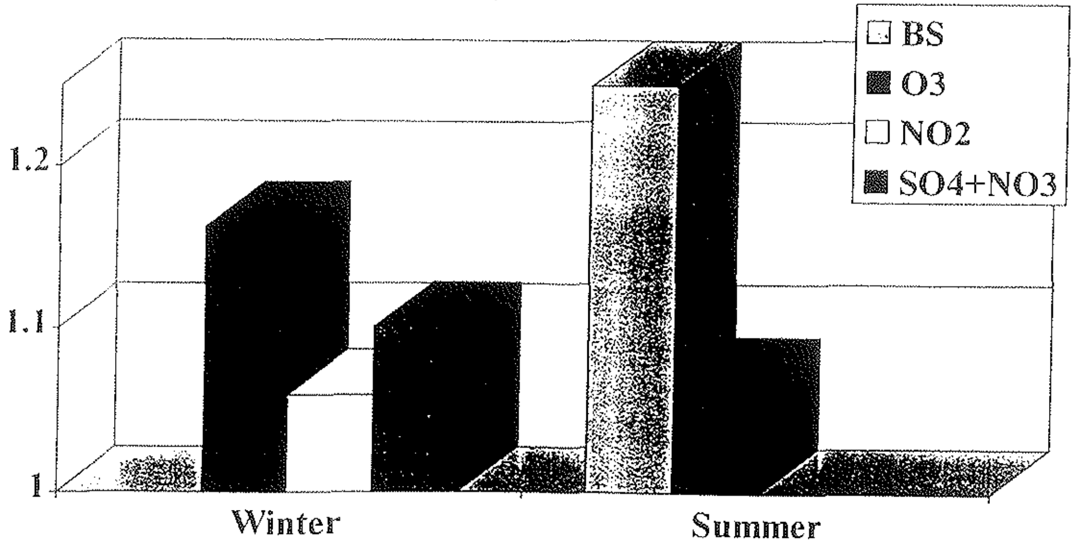
Pollution levels, Dutch mortality study in μg/m³



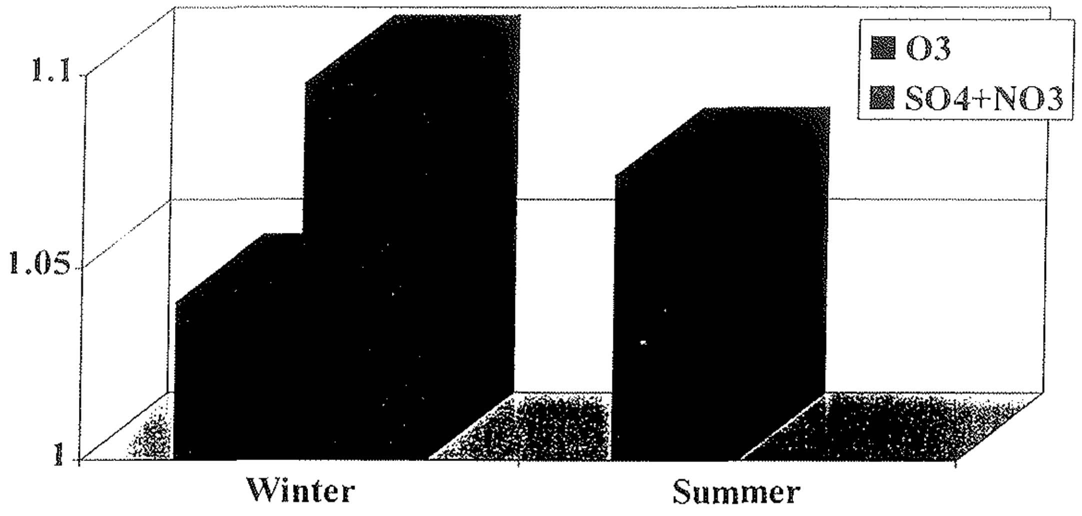
Dutch mortality study - RRs by season (lag1 for O_3, SO_4+NO_3, mean7 for others)



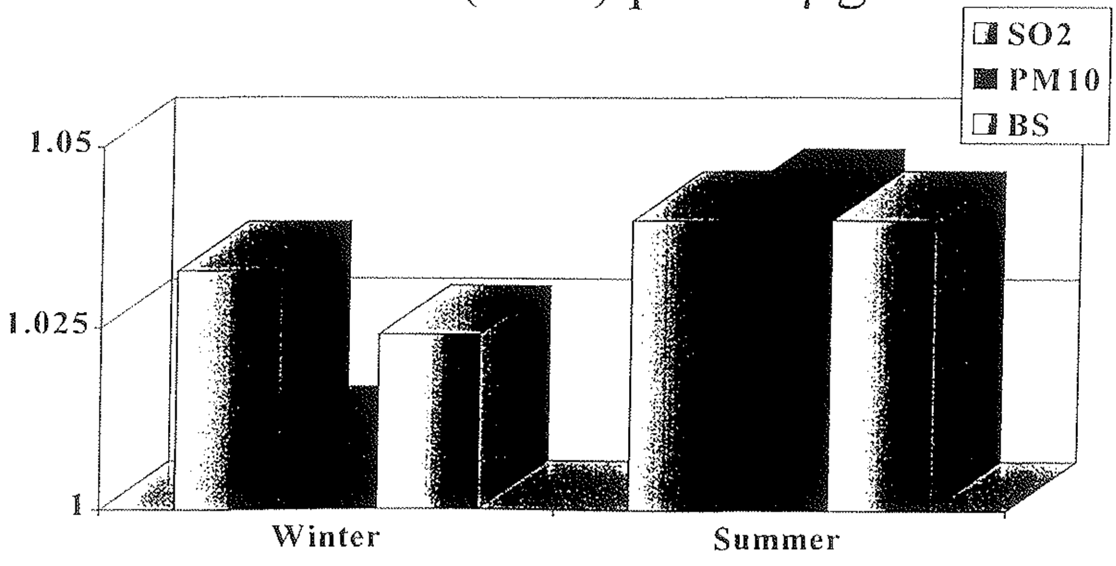
Dutch mortality study - multipollutant RRs by season



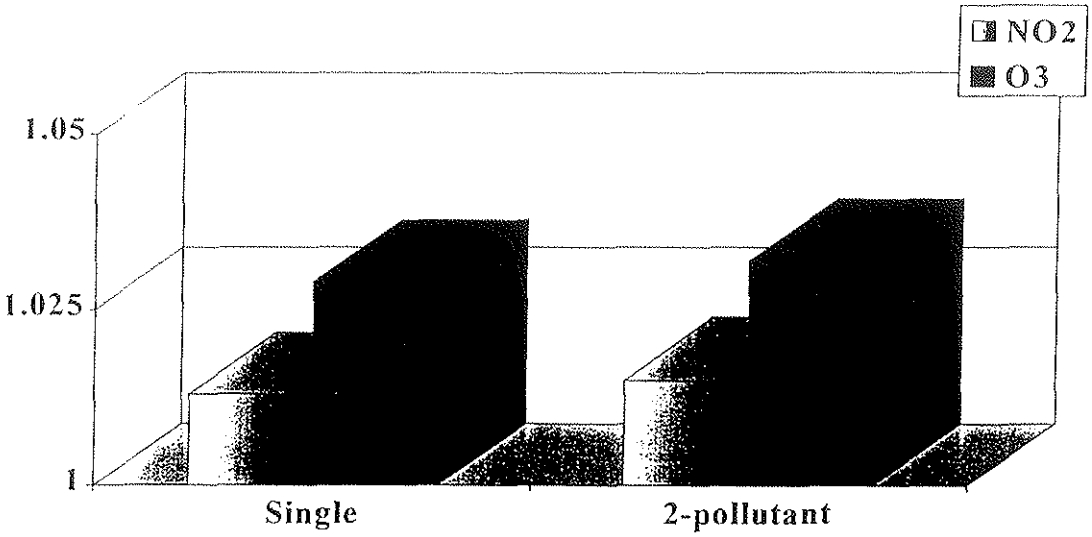
Dutch mortality study - multipollutant RRs by season



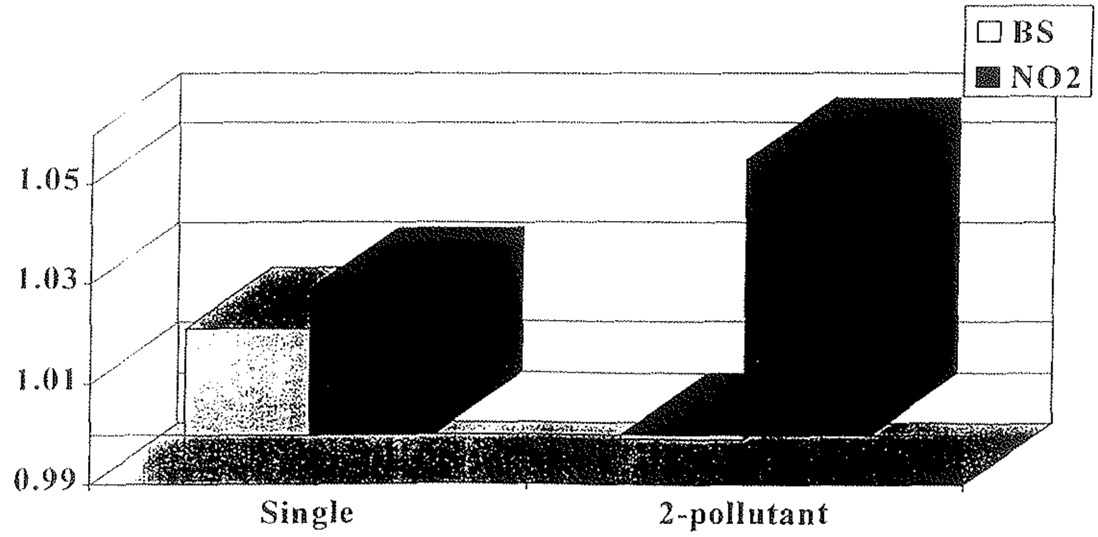
APHEA mortality study (BMJ 1997) relative risks (west) per 50 μg/m^3



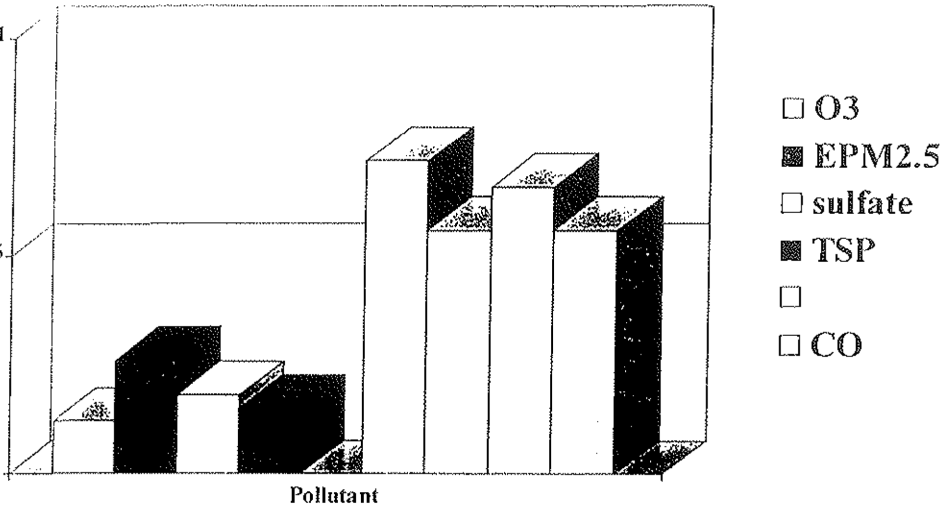
APHEA mortality study (AJE 1997) relative risks per 50 $\mu g/m^3$



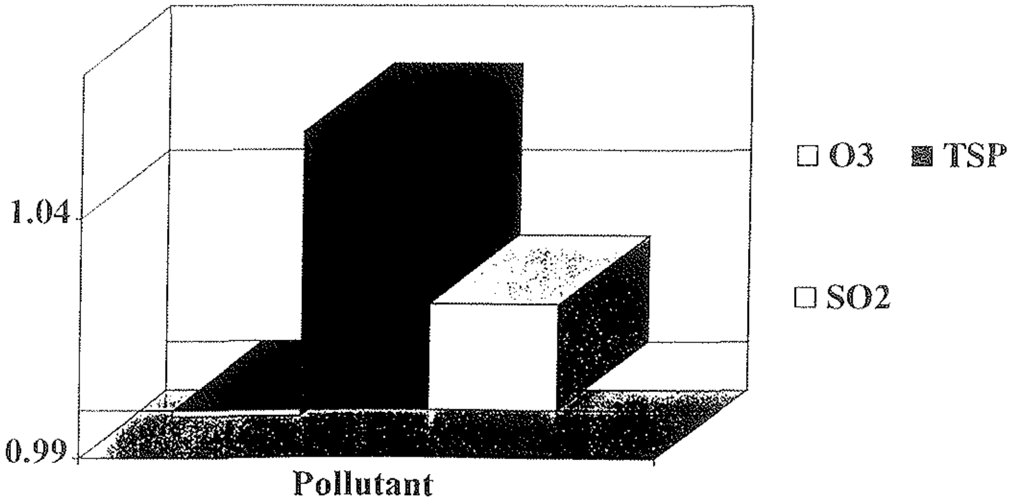
APHEA asthma admissions (Thorax 1997) relative risks per 50 $\mu g/m^3$



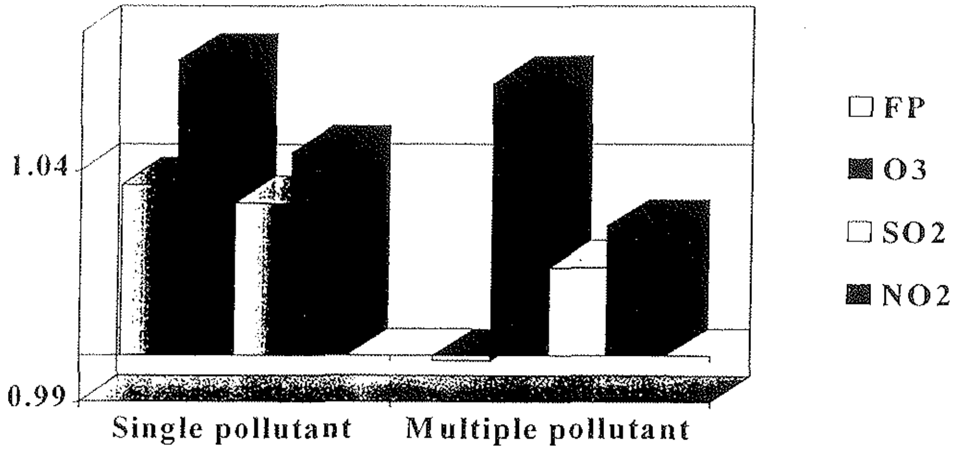
Burnett, JAWMA 1998
Mortality RRs for CO, O_3, PM in Toronto



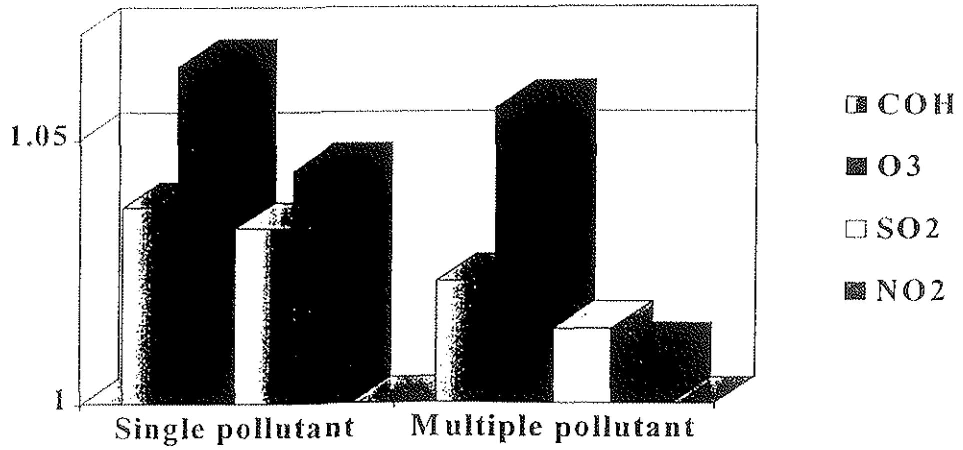
Borja-Aburto, AJE 1998
Mortality RRs for O_3, PM and SO_2 in Mexico City



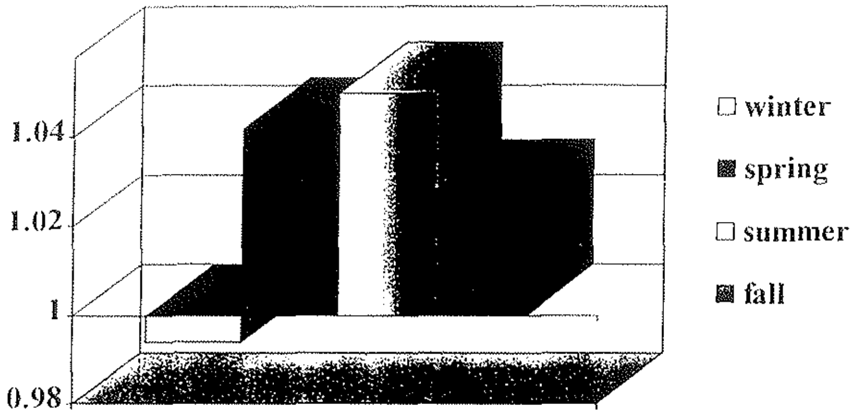
Burnett, EHP 1997
Summer Respiratory Admission RRs for O_3, NO_2, SO_2, PM in Toronto



Burnett, EHP 1997
Summer Respiratory Admission RRs for O_3, NO_2, SO_2, PM in Toronto



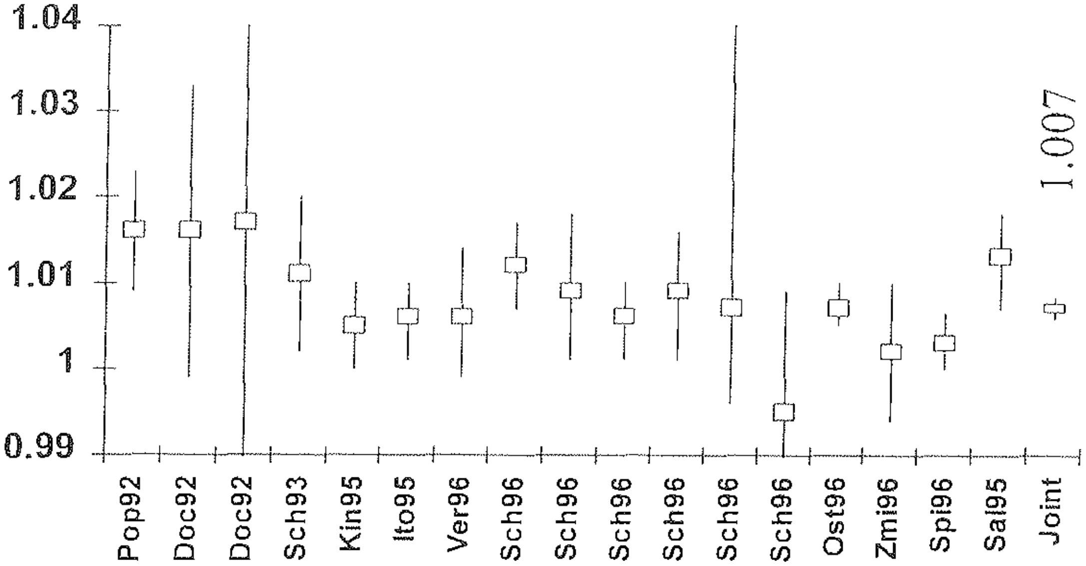
Burnett, Environ Res 1997 Respiratory Admission RRs for O_3 by season in 16 Canadian cities



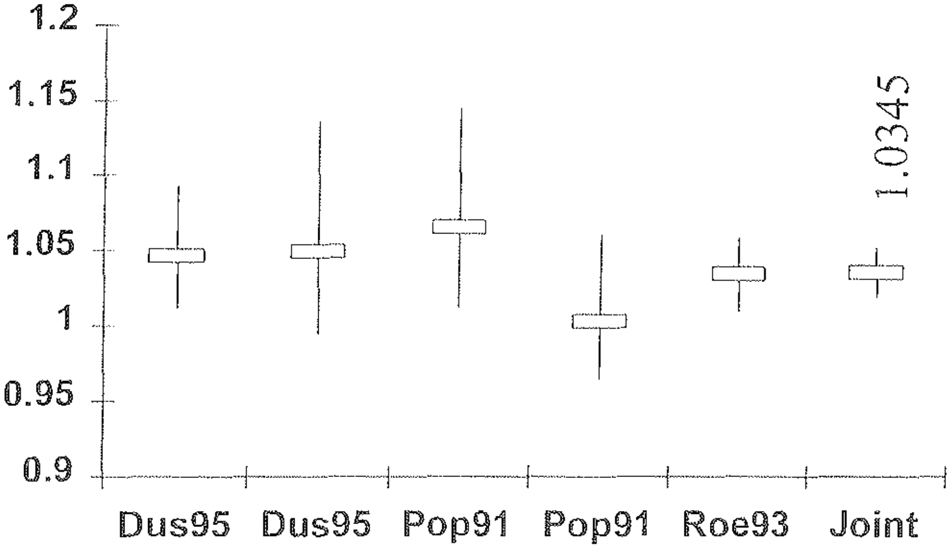
Pope & Kalkstein, EHP 1996 Samet, Environ Res 1998

Alternative weather specifications do not confound or modify associations between particulate air pollution and mortality in Utah Valley or Philadelphia

Relative risk of Mortality per 10 $\mu g/m^3$ PM10



Relative risk of Lower Respiratory Symptoms per 10 $\mu g/m^3$ PM10



Some conclusions

- New studies continue to document effects of PM on mortality and morbidity
- Role of gaseous co-pollutants cannot be ignored
- More information available now on role o PM attributes (number concentration, met content, diesel exhaust, aerosol acidity etc

Some recommendations

- More studies needed that incorporate PM attributes
- More studies needed on PM effects on life expectancy
- More studies needed on toxic potency of real-world air pollution mixtures including gaseous components

MICHAL KRZYZANOWSKI

World Health Organization, The Netherlands

GAPS AND UNCERTAINTIES OF EPIDEMIOLOGY STUDIES

Michal Krzyzanowski
WHO European Centre for Environment and Health, Bilthoven Division

Epidemiology studies conducted in the recent decade have greatly contributed to the identification and quantification of health risk due to exposure to air pollution with respirable particulate matter. The accumulated results provide good scientific basis for the revised WHO Air Quality Guidelines, for the newly proposed Daughter Directive of the EC and for the US national air quality standards. However it is well understood that the available epidemiological studies have significant gaps in the data. That leads to uncertainties in interpretation of the accumulated results and in determination of causality of the observed associations. It also restricts ability of the research to provide more accurate recommendations regarding risk management. The purpose of this presentation is to summarize the main, well-recognized problems of the available studies.

Gaps in exposure assessment
Assessment of exposure in majority of the published studies is based on the routinely collected air quality monitoring data. The most relevant data on respirable fraction of particulate matter, PM10, was available for a limited number of studies conducted in early 1990s, and especially for those done in Europe. The data on PM2.5, or other indicators of fine fraction of the particulate pollution, could have been used in even smaller number of studies. Due to the limited feasibility of the personal monitoring of exposure, the most commonly used exposure indicators for the study subjects are based on data collected in a limited number of fixed monitoring locations. This inevitably leads to the errors in exposure assessment. In most of the studies, a non-differential exposure misclassification can be expected to occur. In the result of such error, the observed association between the level of exposure and health effects (such as relative risk) is smaller than the true one. The extent of the underestimation of the magnitude of association is related to the correlation between the observed and true exposure levels. In some cities (e.g. Prague) the correlation between daily mean ambient concentration of PM10 measured in various locations ranges from 0.6 to 0.7. It may be higher in days with higher wind velocity, and is often better for a fine fraction of particulates, as indicated by a study conducted in Erfurt, Germany (1). Also the correlation between personal exposure and ambient measurements of PM was found to be better for PM2.5 than for PM10 (2). Therefore it can be assumed that the estimates of the effects of fine particles, assessed using the data from a central monitoring location, may be less biased than the estimates of effects of PM10.

Confounders
Factors associated both with the exposure levels and the health outcomes may distort the association between the exposure and health. Epidemiology puts a lot of effort in the identification and elimination of this potential bias from the studies. Typically applied methods consider the confounding in the study design or at the analysis phase. In the studies of the effects of short-term exposures (e.g. time-series studies) the main potential confounding factors include seasonality and other cyclical patterns, long term trends,

calendar effects, weather variables and other temporal events affecting health (e.g. influenza epidemics). Statistical methods developed in the recent decade efficiently adjust the time series data and significantly reduce the risk of the bias by the temporal confounders. In the few available studies of long-term exposures, the confounding by age, gender, tobacco smoking, occupational exposures or residence history is adjusted in the analysis. The effectiveness of this adjustment may depend on the precision of the available information on the confounders and on the model specification used in the analysis. In some of the studies (e.g. in the Swiss SAPALDIA study), intensive sensitivity analysis has been performed to check the possibility of bias introduced by one of many additional potential confounders (3). One of the important tasks of the on-going re-analysis of the two American cohort studies is to test the effectiveness of the applied control of confounding.

Other pollutants
In most studies, the measured indicators of particulate pollution are highly correlated with the levels of gaseous pollutants, such as SO2 or NO2. This results in a limited ability of the present studies to separate the effects of PM from those of the gases. In some studies, the conclusions on the respective roles of individual pollutants are indicated by a graphical analysis, in some other analysis stratified by the levels of co-pollutants is conducted, and in a few studies - multi-pollutant models are possible. The limited data from the European studies suggests that, besides the association of various health outcomes with the PM levels, independent impact of gaseous pollutants takes place (4). Also the existence of the, independent form PM, impact of ozone is well supported by the available studies.

Uncertainties in public health significance of epidemiological observations
Increase of mortality associated with the long-term exposure to particulate matter, as indicated by the cohort studies, reduces life expectancy significantly (by years of life) even in moderate exposure levels (5). However it is still not certain to what extent the effects seen on a population level can be translated to an individual level, and to what extent the effect of pollution reduces healthy life. Less clear is public health significance of the variability of mortality associated with short-term changes in PM concentration, due to a possible harvesting effect (6). However, the reversible changes in health, such as the incidence of symptoms or hospitalization, are believed to represent avoidable excess of morbidity due to the increases in pollution levels.

Existence of a "safe" pollution level
The present studies do not indicate a pollution level below which no effects are found. However it is not certain to what extent such conclusion is related to the real linearity of the association, and to what extent to the limited ability of epidemiological studies to detect such hypothetical safe exposure level. With the diminishing magnitude of the effects it may be not feasible to explore the shape of association at the lower end of the exposure distribution using epidemiology. The necessary size of the study and the precision of measurement of health and exposure limit the feasibility of such research.

Applicability of the present data to various populations

The understanding of the mechanisms involved in the production of the health effects observed in epidemiological studies, knowledge on the specific properties of the particles and on relevant characteristics of the susceptible individuals is still very limited. Therefore the ability to generalize the results of the study depends on the consistency of the studies performed in various populations. There are a large number of studies on the effects of short-term variability of particulates in ambient air, conducted in a variety of populations, climatic and social conditions, as well with various air pollution mixes. This gives a more solid basis to an agreement that similar impacts of air pollution should be experienced also in populations not covered by the completed epidemiological studies. However the applicability of the few studies on the effects of long-term exposures, and the magnitude of the, potentially very serious, impacts on the mortality, is less certain and needs to be supported by further studies performed in various populations.

Conclusions

Progress of epidemiology in the recent decades has enabled detection of associations not seen when simple methods are used. Improved control for confounding, sophisticated statistical models and computing capability orders of magnitude greater than two decades ago contribute to the epidemiological analysis. However numerous uncertainties need still to be resolved by epidemiology. The most important area for progress is the improvement of exposure assessment, reduction of errors in determination of personal exposure and inclusion of a wider set of, possibly more relevant for health, indicators of air pollution mix. Further studies of long-term exposures are also urgently needed to confirm the risk estimates of the presently available studies, to better characterize the pollution characteristics responsible for the effects and population sub-groups at the highest risk of the impacts. To reduce the present vast uncertainties, epidemiology must be guided by a better understanding of mechanisms of the pollution based on clinical and toxicological research.

References

1. Cyrys J, Heinrich J, Brauer M, Wichmann HE. Spatial variability of acidic aerosols, sulfate and PM10 in Erfurt, Eastern Germany. J Exp Anal Environ Epidemiol 1998; 8: 447-463
2. Janssen NAH, Hoek G, Harssema H, Brunekreef B. Personal exposure to fine particles in children correlates closely with ambient fine particles. Arch Env Health 1999 (in press)
3. Ackermann-Liebrich U, Leuenberger P, Schwartz J et al. Lung function and long term exposure to air pollutants in Switzerland. Am J Respir Crit Care Med 1997; 155: 122-129
4. Katsouyanni K, Touloumi G, Spix C et al. Short term effects of ambient sulphur dioxide and particulate matter on mortality in 12 European cities: results from time series data from the APHEA project. Brit Med J 1997; 314: 1658-63
5. Brunekreef B. Air pollution and life expectancy: is there a relation. Occup Environ Med 1997; 54: 781-784

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GAPS AND UNCERTAINTIES IN EPIDEMIOLOGY STUDIES

M. Krzyzanowski
WHO ECEH Bilthoven Division

GAPS IN DATA

- Exposure assessment
- Confounding factors
- Other pollutants

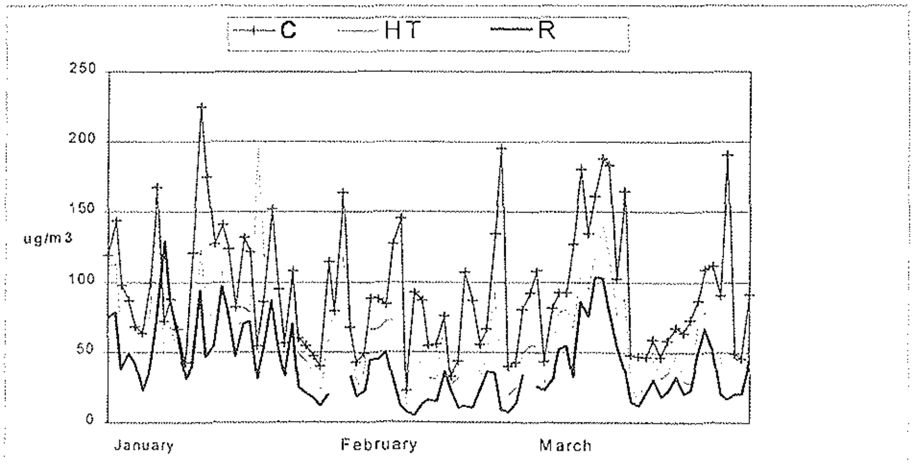
UNCERTAINTIES OF INTERPRETATION

- Public health impacts (loss of health / life years)
- Reference ("threshold") exposure level
- Applicability outside study population

GAPS IN EXPOSURE ASSESSMENT

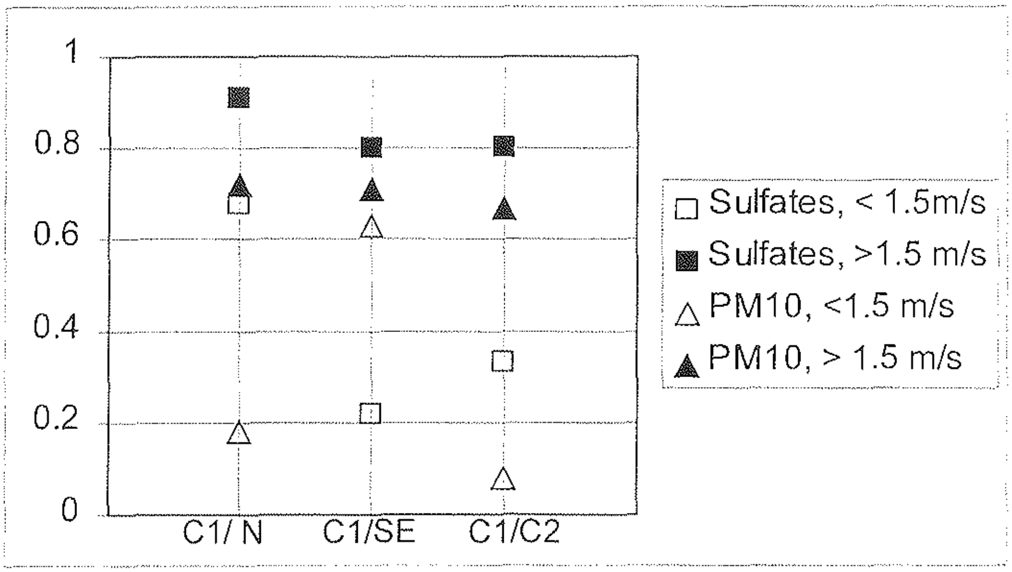
- Limited availability of PM10 or PM2.5 data
- Few studies with other indicators of respirable PM
- Ambient monitoring in a central location
- Personal exposure vs.. monitoring data

Daily average PM10 concentrations, Prague, 1 January - 31 March 1997



Correlation: 0.56 (Centre / Resid.) – 0.68 (Traffic – Resid.)

Correlation of sulfate and PM10 levels by monitoring sites in Erfurt, GER, stratified by wind speed



Cyrys et al., JEAEE 1998;8: 447-64

EFFECTS OF NON-DIFFERENTIAL EXPOSURE MEASUREMENT ERROR

$$B(obs.) = B(true) * r^2(obs., true)$$

$$OR(obs.) = OR(true) * \exp[r^2(obs., true)]$$

$$0 < r^2(obs., true) < 1$$

PERSONAL vs. AMBIENT LEVELS OF PM

(individual correlation coefficients in a group of Dutch children not exposed to ETS)

	Median	Range
PM2.5	0.92	0.63 - 0.97
PM10	0.73	0.07 - 0.99

Source: Janssen et al., Arch Env Health 19

CONFOUNDING FACTORS

Studies of effects of exposure:	Confounders:
short-term	*time - dependent*
long-term	*study group - dependent*

CONFOUNDING FACTORS CONTROLLED IN TIME-SERIES STUDY (*APHEA*)

- Seasonality and other cyclical pattern
- Ambient temperature and humidity
- Long term trends
- Calendar effects
- Influenza epidemics

- LAG TIMES?

Source: Katsouyanni et al, J Epidemiol Comm Health 1995; 50(Suppl. 1), S12-18

CONFOUNDING FACTORS CONTROLLED IN STUDIES ON EFFECTS OF LONG-TERM EXPOSURE

- Age, gender
- Smoking status (present, past, amount, duration)
- Pre-existing respiratory symptoms
- (Occupational exposures)
- (Duration of residence in the study area)

RELATION OF FVC TO PM10 IN HEALTHY NEVER-SMOKERS: SENSITIVITY ANALYSIS TO CONTROL FOR ADDITIONAL COVARIATES *(Ackermann-Liebrich et al, AJRCCM 1997: 155:122-29)*

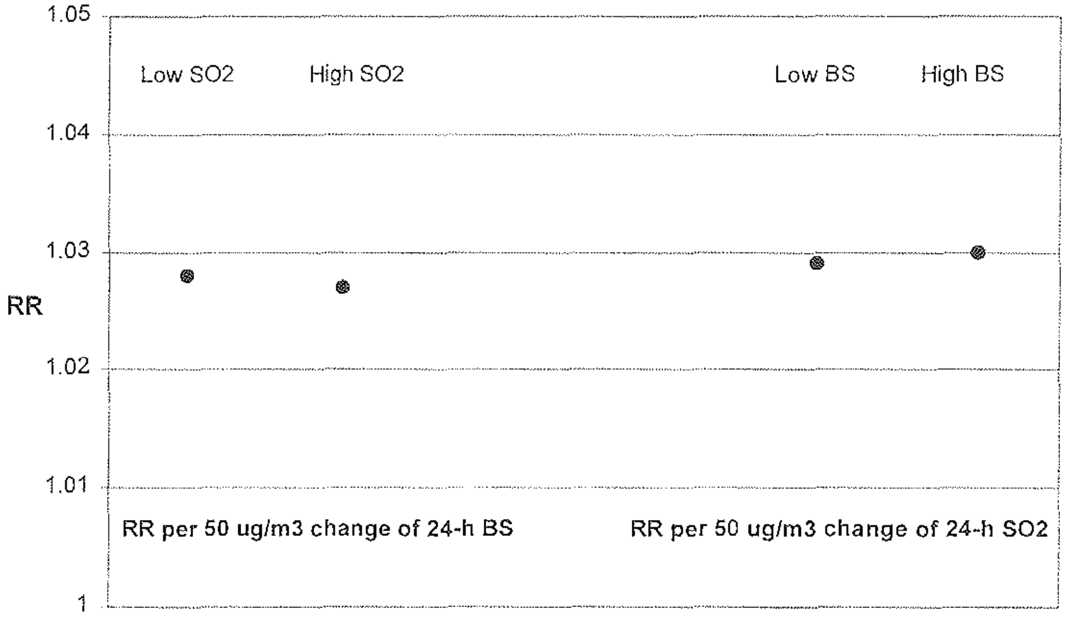
Variable	Estimated decrement in FVC (L) per 10 µg/m3 PM10
Basic model	**0.0345**
Low education	0.0349
Work exposure	0.0347
CO<7.7	0.0362
Foreign citizenship	0.0360
Respir. infection, age < 5 years	0.0362
Sibling asthma	0.0362
Maternal smoking	0.0360
Gas stove	0.0366
Mountain area (> 1000 m a.s.l.)	0.0336
Random effect model	0.0360

CORRELATION COEFFICIENTS OF ANNUAL MEAN PM10 AND OTHER AIR POLLUTANTS IN 8 STUDY AREAS IN SWITZERLAND

(Ackermann-Liebrich et al, AJRCCM 1997: 155:122-29)

Pollutant coefficient	Correlation
SO2	0.93
NO2	0.91
O3	-0.55
Summer daytime O3	0.31
O3, excess concentr. over 120 µg/m3	0.67

Estimated RR (and 95%C.I.) of deaths from all causes in Western European cities *(Katsouyanni et al, BMJ 1997; 314: 1658-63)*



Estimated health impacts of air pollution with particulate matter in Europe in mid-1990s

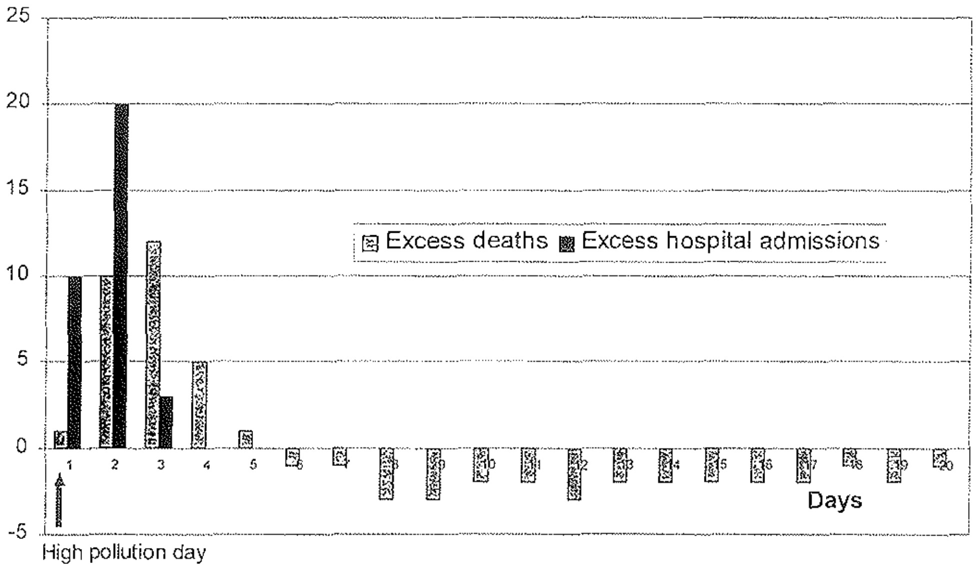
Exposure	Health indicator, population at risk	Attributable proportion	Estimated number of cases per year (x1000)
Long term	Mortality[1], urban, age >35	4% - 13%	102 - 368
Short term variability	Daily number of deaths[1], urban	1.4% - 3.2%	41 - 89
	Hospital admissions, for respiratory disease, urban	1.5% – 3.4%	7 - 16

1) All deaths except accidents

PROBLEMS IN ESTIMATION OF IMPACTS ON PUBLIC HEALTH

- Short-term exposures - harvesting effect (?)
- Long term exposure - transformation of RR from a cohort to increase in mortality in a population

HYPOTHETICAL EFFECT OF ONE-DAY POLLUTION EPISODE



CONCLUSION

Substantial reduction of uncertainties related to epidemiology studies on health effects of PM can be achieved through:

- Improvement of exposure assessment in epidemiology studies

- Expansion of the range of measured PM indicators and other compounds of the pollution mix

- Implementation of further studies on effects of long-term exposure

MARK UTELL

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CURRENT STATUS OF KNOWLEDGE OF ABOUT HOW PARTICLES MIGHT CAUSE HEALTH EFFECTS

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During the past decade, there has been a remarkable expansion of the epidemiologic literature on the health effects of air pollution in general and of particulate air pollution in particular. Despite the expanding epidemiologic database, the biologic mechanisms by which particulate matter induce health effects at low mass concentrations remains unclear. Furthermore, the particle size which accounts for the epidemiologic associations is uncertain. The urban ambient particles are present as a trimodal distribution consisting of a coarse mode with a median size of about 5 µm, an accumulation mode with a median size of about 0.2 µm, a nucleation mode with a median particle size of about 20 nm (ultrafine particles). Hypotheses proposed to explain which particles are responsible have focused on issues related to particle acidity, particle content of transition metals, bioaerosols, and ultrafine particles. We have suggested that ambient ultrafines are important with regard to respiratory health effects, for several reasons: 1) ultrafine are biologically more reactive than larger-size particles, and elicit effects at low concentrations; 2) ultrafines at the same mass concentration in the air have a much higher number concentration and surface area than larger particles; 3) inhaled singlet ultrafines have a very high deposition efficiency in the pulmonary region; and 4) ultrafines have a propensity to penetrate the epithelium and reach interstitial sites.

Populations at Risk

Several lines of evidence suggest that persons at risk from inhaled particles are those with severe heart and lung diseases and perhaps the elderly as well as children. Persons with coronary artery disease have arthrosclerotic narrowing of the coronary arteries, which deliver blood to the heart. Cardiovascular disease accounts for nearly 45% of US mortality and much more morbidity. Its cost to the U.S. economy is by far the largest for any diagnostic group, amounting to an estimated 144 billion dollars in 1988. Coronary artery disease causes 800,000 new myocardial infarctions per year. The incidence in women lags behind that in men by 10 years for total coronary heart disease and by 20 years for myocardial infarction and sudden death. Although the incidence and severity of coronary heart disease increases with age in both sexes, it is not a disease limited to the elderly.

Persons with chronic obstructive pulmonary disease (COPD) have physiologically significant impairment of lung function, most often from underlying emphysema and airways narrowing caused by smoking. The term COPD encompasses various pathophysiological states (emphysema and chronic bronchitis) associated with obstruction to air flow. The obstruction is relatively fixed, differentiating this condition from asthma, in which reversibility or variability in air flow obstruction is a cardinal feature. In the U.S., COPD accounts for 13% of hospitalizations and is the fifth leading cause of death and climbing.

In contrast with COPD, asthma is often a disease of the young and otherwise healthy; the incidence is highest in the first 10 years of life. It is a very common condition, affecting up to 10% of the U.S. population. Moreover, not only are the incidence and prevalence increasing, but based on the increase in asthma hospitalizations, the severity and acuity of the disease seems to have increased. The hallmark features of asthma are reversible airway obstruction, hyperresponsiveness, and inflammation. A growing body of evidence implicates an allergic sensitization in the etiology of asthma.

Potential Mechanisms of Cardiopulmonary Responses

There is no established mechanism to explain the relationship between pollutant exposure and excess cardiovascular mortality. Recent studies in healthy and compromised animals have suggested that inhalation of particulate matter may induce changes in cardiac rhythm or repolarization. Although there is little evidence to suggest that exposure to particulate air pollution has direct cardiac effects, penetration of very small particles (e.g., ultrafines) or their reaction products into the systemic circulation could induce inflammatory cytokine expression in the myocardium, resulting in myocarditis or epicarditis, or progression of coronary artery disease. Furthermore, an acute inflammatory responses in the airway may be accompanied by an acute phase response, with increases in plasma viscosity and blood coagulation factors, such as fibrinogen, Factor VII, and plasminogen activator inhibitor. Such events could cause the observed increases in heart rate, changes in heart rate variability, contribute to congestive heart failure and arrhythmias, or precipitate coronary events in individuals with coronary artery disease.

Similarly, the mechanisms by which particles cause adverse effects in patients with asthma and COPD have not been determined. Particle exposure could contribute to progression of disease by enhancing inflammation. A mechanistic model for particle-induced lung inflammation involves injury to the epithelial cells by reactive oxygen species, possibly enhanced in the presence of metals via Haber-Weiss and Fenton chemistry, accompanied by activation of nuclear regulatory factors, leading to elaboration of proinflammatory cytokines, including IL-8 and IL-6, and increased expression of nitric oxide synthase with increased nitric oxide in exhaled air. This in principle could also result in activation of vascular endothelium and circulating leukocytes. Alternatively, particles could increase susceptibility to infectious complications by impairing mucociliary clearance, by increasing adhesion of bacteria to epithelial cells, by impairing alveolar macrophage function, or by impairing specific or non-specific functions of the immune system.

The research effort to understand the mechanisms of particulate toxicity has been intensive and has fostered meaningful collaborations between epidemiologists, toxicologists and clinical investigators.

CURRENT STATE OF KNOWLEDGE ABOUT HOW PARTICLES MIGHT CAUSE HEALTH EFFECTS

Mark J. Utell, M.D.

University of Rochester Medical Center
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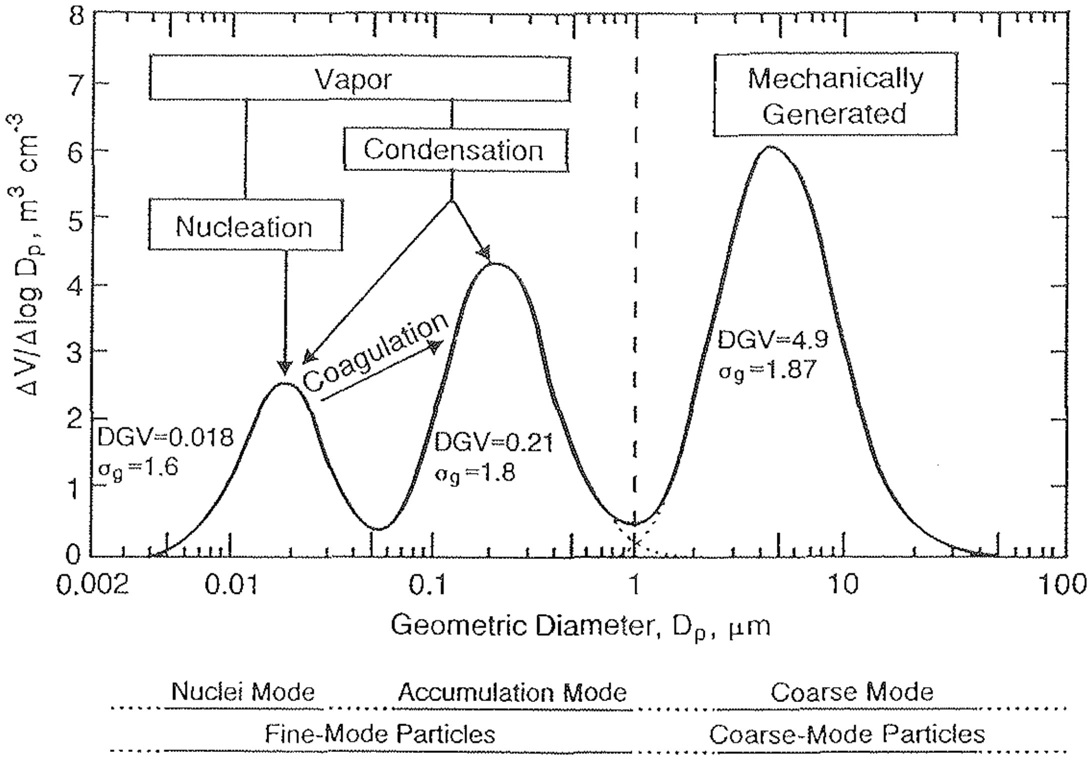
FOUNDATION FOR NATIONAL AMBIENT AIR QUALITY STANDARDS (NAAQS)

1. Epidemiological Investigations
2. Toxicologic Studies
    - Whole Animal
    - Molecular and Cellular Studies
3. Human Controlled Exposure Studies



HYPOTHESES FOR PM HEALTH EFFECTS

1. **Acidic Properties of PM**
2. **Ultrafine Distribution of PM Fraction**
3. **Altered Intrapulmonary Distribution of the Lung PM Dose**
4. **Organic Fraction of PM**
5. **Presence of Biological Materials in PM**
6. **Transition Metal Constituents of PM**

Populations considered susceptible to air pollution

Population	Potential mechanism	Consequences
Asthmatics	Increased airways responsiveness	Increased risk for exacerbation of respiratory symptoms
Cigarette smokers	Impaired defense and clearance, lung injury	Increased damage through synergism
Elderly	Impaired respiratory defenses, reduced functional reserve	Increased risk for respiratory infection, increased risk for clinically significant effects on function
Infants	Immature defense mechanisms of the lung	Increased risk for respiratory infection
Persons with CHD	Impaired myocardial oxygenation	Increased risk for myocardial ischemia
Persons with COPD	Reduced level of lung function	Increased risk for clinically significant effects on function

CHD, coronary heart disease.

CORONARY HEART DISEASE

FACTS:

- Cardiovascular disease accounts for 44% of U.S. mortality
- Cost = $144 billion/year (1991)
- 5-year medical cost of MI = $51,000
- CAD causes 800,000 new MIs/year

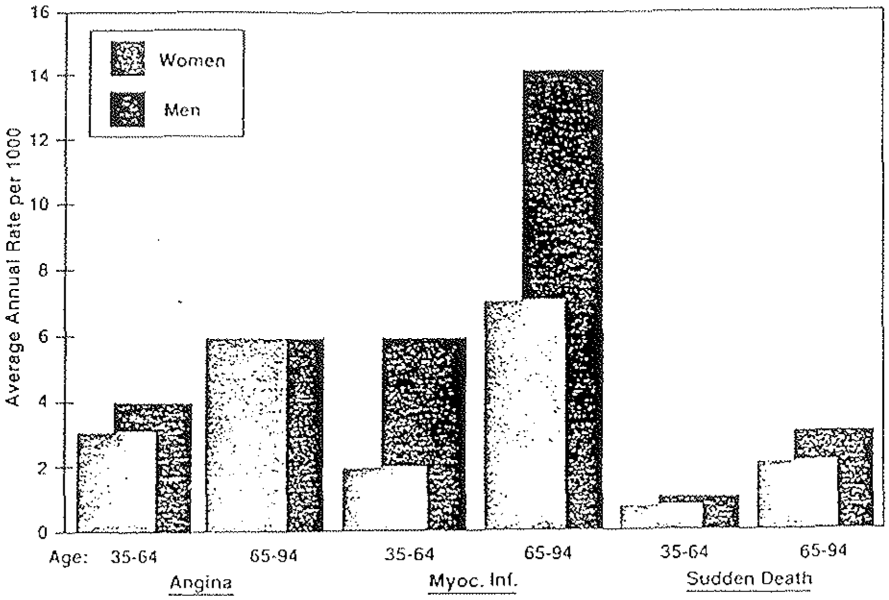
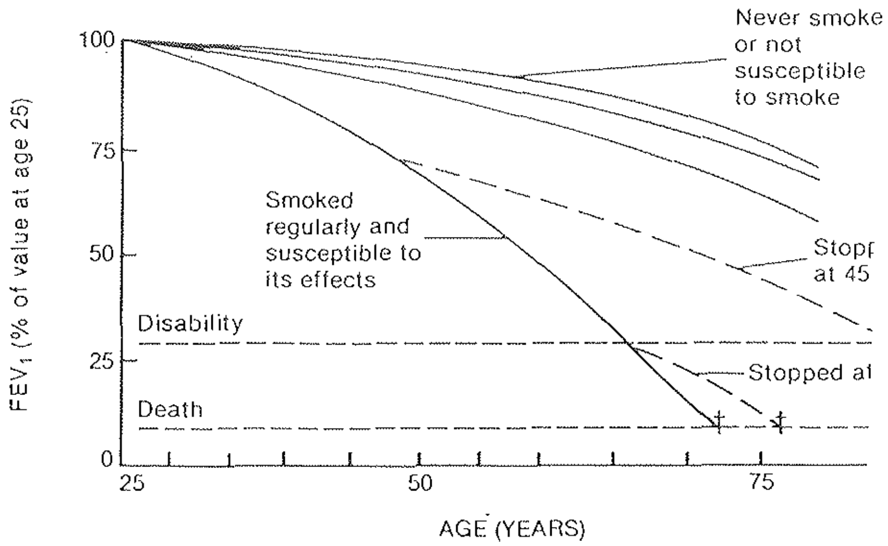


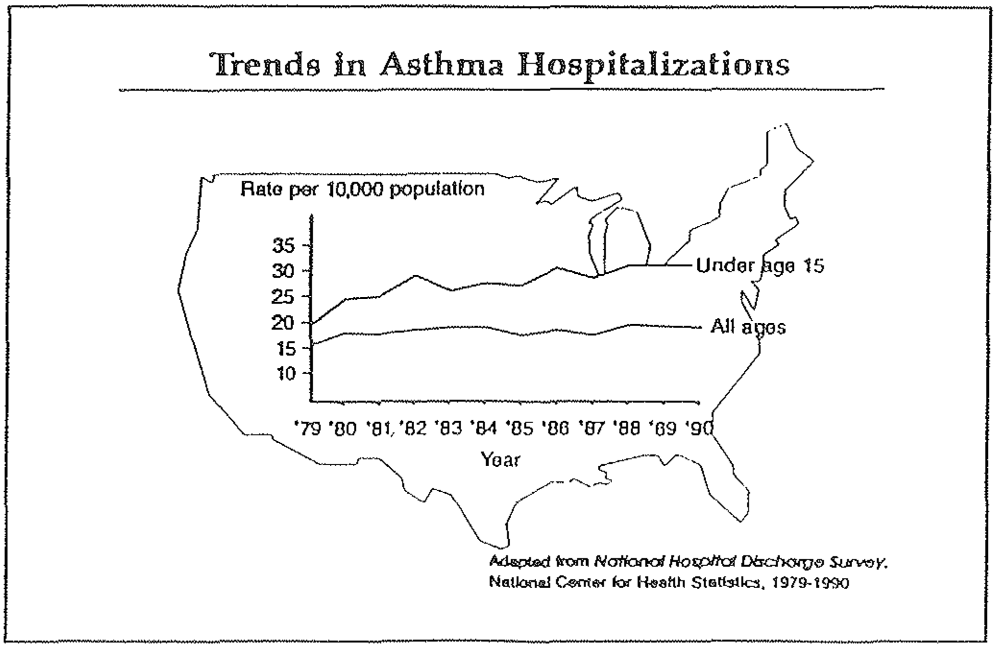
FIG. 4. Incidence of clinical manifestation of coronary heart disease by age and sex.

CHRONIC OBSTRUCTIVE LUNG DISEASE (COPD)

1. COPD
   - Chronic Bronchitis
   - Emphysema

2. Severity and Significance
   - 5th leading cause of death and climbing
   - Accounts for 13% of hospitalizations





Adapted from *National Hospital Discharge Survey*. National Center for Health Statistics, 1979-1990

HYPOTHESIS

(Seaton et al., The Lancet, 1995)

Airway Inflammation →

Systemic Acute Phase Response →

Blood Hypercoagulability →

Coronary Event

LOCAL INFLAMMATORY RESPONSE

Ozone Exposure:

→ ↑ IL-6 → APR
→ ↑ IL-8 → PMNs

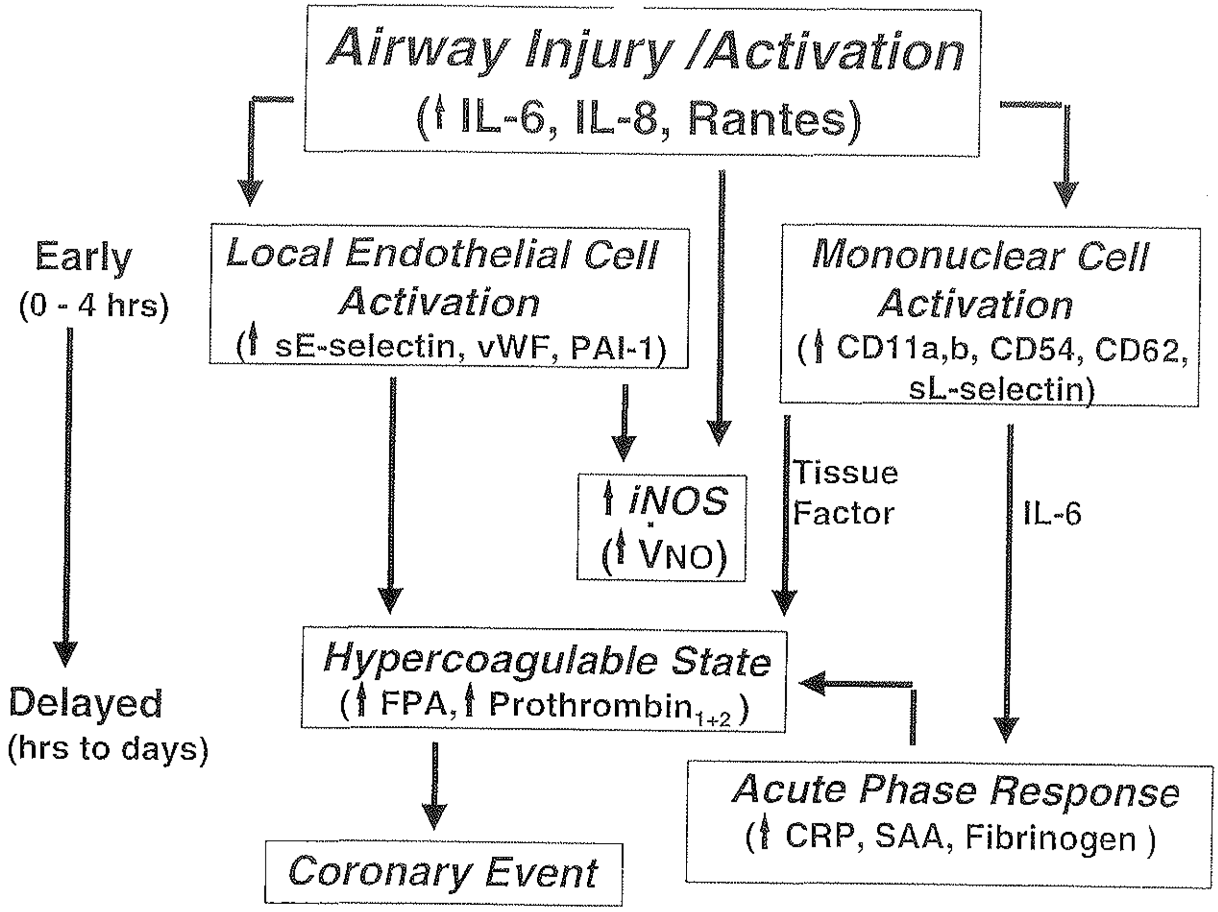
Exacerbation of Airways Diseases

SYSTEMIC ACUTE PHASE RESPONSE (APR)

- Initiation of APR → IL-6
- Stimulates liver to make acute phase proteins
  (CRP, Fibrinogen, etc.)
    → Hypercoagulable State

CARDIAC RESPONSES

- AUTONOMIC NERVOUS SYSTEM
  airway inflammation → alteration in HR variability →
    in severe CAD → EVENT
- DIRECT PARTICLE EFFECT
  cytokines in cardiac muscle →
    - vasoconstriction
    - plaque rupture
    - occlusion 2° increased inflammation
- INDIRECT EFFECT
  mononuclear cell activation
    → activated in lung → circulation →
      settle in coronary plaque
  (smoking role in CAD)



Airway Injury /Activation

(↑ IL-6, IL-8, Rantes)

Early
(0 - 4 hrs)

Local Endothelial Cell Activation

(↑ sE-selectin, vWF, PAI-1)

Mononuclear Cell Activation

(↑ CD11a,b, CD54, CD62, sL-selectin)

↑ *iNOS*

(↑ $\dot{V}_{NO}$)

Tissue Factor

IL-6

Hypercoagulable State

(↑ FPA, ↑ Prothrombin$_{1+2}$)

Delayed
(hrs to days)

Acute Phase Response

(↑ CRP, SAA, Fibrinogen)

Coronary Event

CONCLUSIONS: A CLINICAL PERSPECTIVE

- The Epidemiologic Observations Have Consistently Shown an Association Between Low Level Particle Concentrations and Mortality.
- From a Clinical Perspective, the Mechanism of Cardiopulmonary Mortality and Morbidity Is Not Yet Explained.
- To Date, Clinical Inhalation Studies Do Not Demonstrate Marked Sensitivity of Individuals With Chronic Respiratory Diseases to Particulate Matter.

What New Research Results Are Emerging?

Chairs: *Robert Maynard,* Department of Health, United Kingdom; and *Bernd Seifert,* Umweltbundesamt, Germany

- Which Groups of the General Population May Be at Increased Risk of Exposure to PM?
- Which Are the Characteristics of PM that Are Important to Human Health?
- The Role of Multicenter Studies in Air Pollution Research

BERT BRUNEKREEF

University of Wageningen, The Netherlands

IDENTIFICATION OF SUSCEPTIBLE SUBGROUPS FOR PM EFFECTS

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A large number of studies have documented effects of short-term exposures to particulate matter in ambient air on mortality and morbidity. These studies have led regulatory agencies such as the U.S. EPA and the E.U. to promulgate new, tight air quality standards.

An important question is whether some groups in the general population are more vulnerable than others. It has been well established that for effects on mortality, elderly subjects are more vulnerable than the young. It has been shown also that subjects with asthma more readily respond to low levels of air pollution with symptoms and lung function changes than subjects without asthma. Children are sometimes thought to be more sensitive than adults, but few systematic attempts to compare the effects in children and adults have been made. A recent study from the Netherlands compared 7-12 yr. old children with 50-70 yr. old adults, and found evidence that under the same circumstances, effects on lung function and symptoms in children who already had chronic respiratory symptoms were stronger than in similar adults. There was little effect, and hence little difference in effect, between non-symptomatic children and adults. Other characteristics were also investigated in this study, such as atopy and bronchial responsiveness. It was found that children as well as adults who had atopy (increased total IgE) as well as bronchial hyperresponsiveness to methacholine, were clearly more sensitive to PM air pollution than subjects without these characteristics, or with only one of the two.

Another factor that may influence susceptibility is nutrition. In the newest U.S. cohort study, it was shown that subject who had low antioxidant vitamin intake in the beginning of the follow-up were more likely to die in relation to air pollution than subjects with a high antioxidant vitamin intake. A study from The Netherlands showed that acute effects of PM were also modulated by antioxidant vitamin status (based on serum measurements).

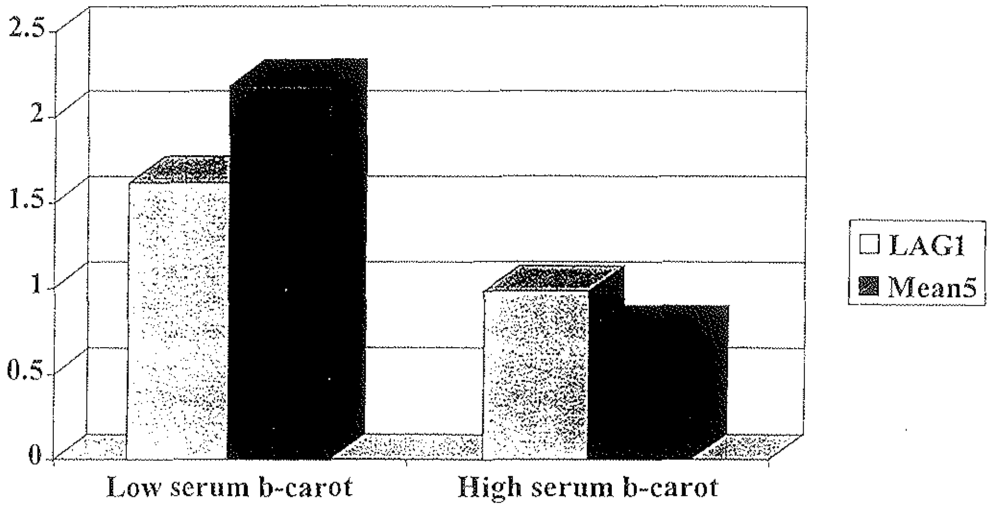
Further studies on subject' characteristics is likely to reveal other susceptible subgroups as well. Whereas these studies are important from a mechanistic point of view, it is less clear what they contribute to environmental policy.

Which groups of the general population may be at increased risk of exposure to PM

Bert Brunekreef PhD

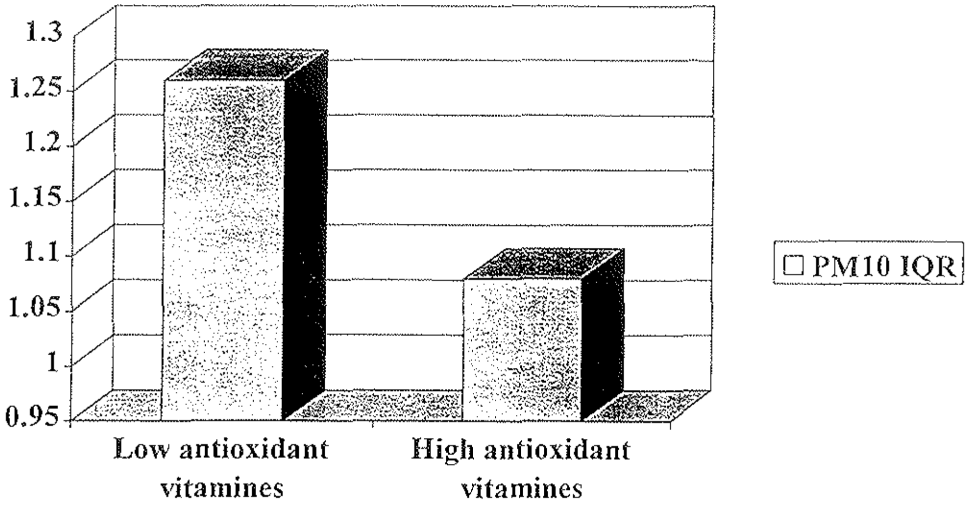
Grievink et al., ERJ 1999

RRs for 10% amPEF decrements per 100 µg/m^3 PM10



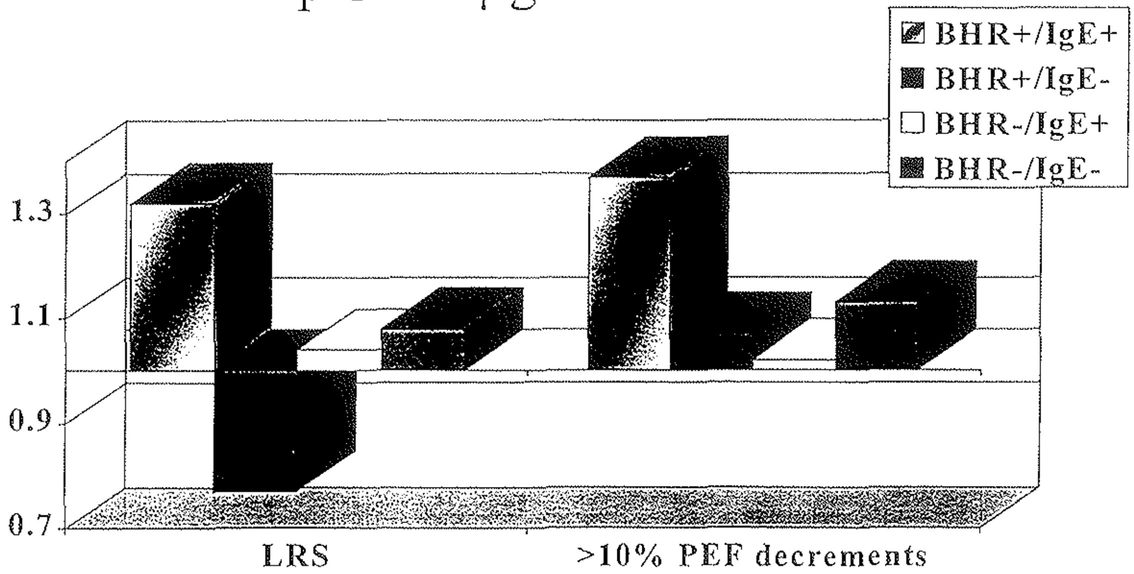
Abbey et al., AJRCCM 1999

RRs for CRC mortality per 24 $\mu g/m^3$ PM10 (IQR)

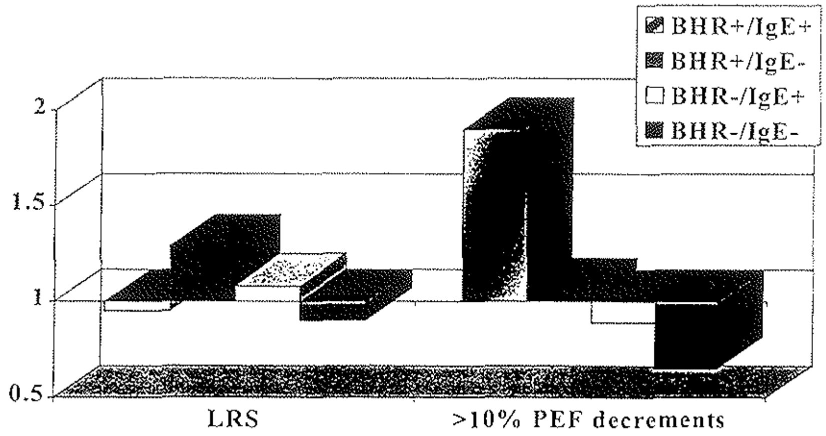


Boezen et al., Lancet (?) 1999

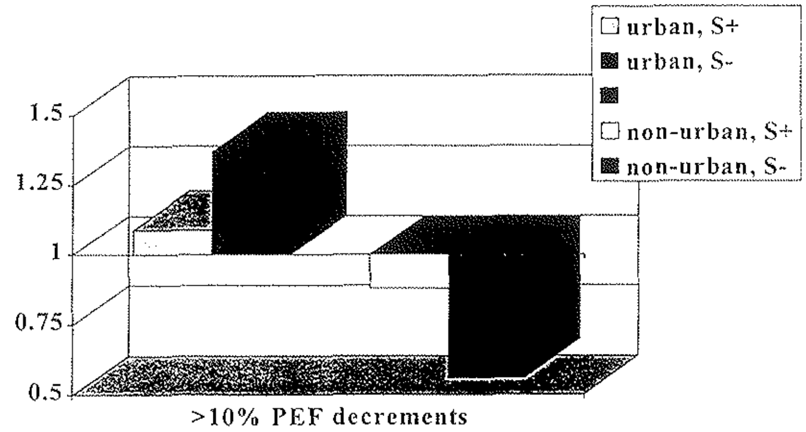
relative risks in sub-groups of children,
per 100 $\mu g/m^3$ PM10



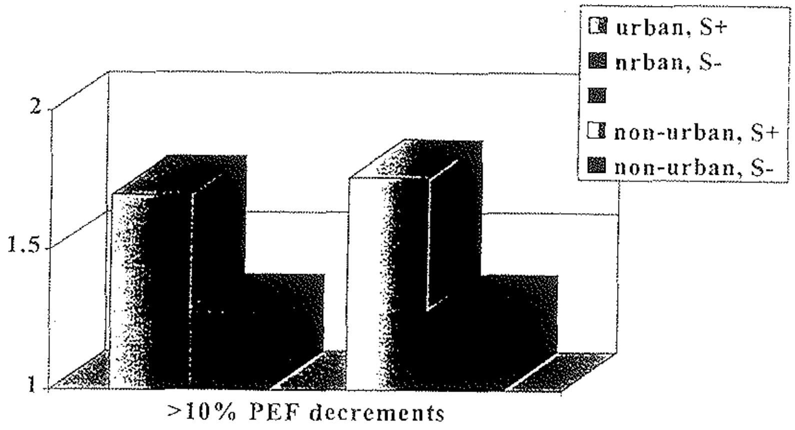
Hoek, 1999
relative risks in sub-groups of adults,
per 100 $\mu g/m^3$ PM10



Van der Zee et al., 1999
relative risks in sub-groups of *adults*, per 100
$\mu g/m^3$ PM10



Van der Zee et al., 1999
relative risks in sub-groups of *children*, per 100
$\mu g/m^3$ PM10



FRANK SPEIZER (for Douglas Dockery)

Harvard School of Public Health, United States

Air Pollution and Incidence of Cardiac Arrhythmia

Annette Peters[1,2], Emerson Liu[3], Richard L. Verrier[3], Joel Schwartz[1,4], Diane R. Gold[1,4], Jeff Baliff[1], Any Oh[1], George Allen[1], Kevin Monahan[3], Douglas W. Dockery[1,4]

[1]Department of Environmental Health, Harvard School of Public Health;[2] Institute of Epidemiology, GSF-National Research Center for Environment and Health, Munich, Germany; [3]Beth Israel-Deaconess Hospital; [4]Channing Laboratory, Harvard Medical School

Abstract

Elevated levels of particulate air pollution have been associated with an increase in hospital admissions and mortality for cardiovascular diseases. This study tests the hypothesis that patients with implanted cardioverter defibrillators (ICD) experience potentially life threatening arrhythmias associated with particulate air pollution episodes.

Tachycardia and ventricular fibrillation were identified from records of 100 patients with ICD devices at the Beth Israel-Deaconess Hospital Cardiac Device Clinic for the years 1995 through1997. During the same period, 24 hour mean concentrations of particulate matter (PM_{10} and $PM_{2.5}$), Black Carbon and gaseous air pollutants were measured in the Boston area.

Air pollution concentrations were moderate during this period. However, an increase of 26 ppb NO_2 was associated with increased tachycardia and ventricular fibrillation two days later, odds ratio of 1.8 (95% confidence interval: 1.1 to 2.9). Patients with repeated events (10 or more events during a three year follow-up) were especially at risk of experiencing arrhythmia in association with $PM_{2.5}$ (odds ratio: 1.6 for an increase of 22 $\mu g/m^3$ (95% confidence interval: 1.0 to 2.6)) and NO_2 (odds ratio: 2.8 for an increase of 26 ppb (95% confidence interval: 1.5 to 5.1)).

These results suggest that elevated levels air pollutants are associated with potentially life-threatening arrhythmia leading to therapeutic interventions by an implanted cardioverter defibrillator.

Introduction

- Episodes of particulate air pollution have been associated with increased hospital admissions for cardiovascular disease and increased cardiovascular mortality in epidemiological studies.
- Studies on hospital admissions and mortality suggest that persons with underlying heart disease are at an increased risk for particulate air pollution health effects.
- Controlled particulate air pollution exposure studies in dogs suggest modifications in heart rate variability and morphological changes of electrocardiograms (EKG) consistent with an increase in sympathetic activity.
- This study tests the hypothesis that patients subject to serious arrhythmia requiring implanted cardioverter defibrillators (ICD) experience potentially life-threatening arrhythmia associated with particulate air pollution episodes.

PATIENT CHARACTERISTICS

- Patient records of Beth Israel-Deaconess Hospital Cardiac Device Clinic, 1995 through 1997.
- History of coronary artery disease and arrhythmia often resulting in syncope.
- Patients followed up at clinic every three months.
- ICD detected arrhythmia and therapeutic interventions are downloaded, printed, and reviewed by the nurse managers.

Patients were included if the

- device was implanted before September 1997
- alive as of December 1997
- more than 30 days of follow-up and
- lived within the zip code areas 01800-02799 (greater Boston and Cape Cod).
- first two months after implantation were excluded

Table 1: Distribution of events in the sample of 100 patients with implanted cardioverter defibrillators

Number of events	Number of persons	Total number of events	Person -days	Mean follow-up	Age	Male patients
0	67	0	40248	601	61.9 (22-85)	75%
1-4	20	39	12459	623	64.9 (30-81)	90%
5-9	7	48	5583	798	59.0 (30-78)	86%
10 and more	6	136	5338	890	60.5 (37-77)	83%
Total	100	223	63628	636	62.2 (22-85)	79%

Air Pollution Measurements

Air pollution measured (Jan 1995 – Dec 1997) in South Boston.

- $PM_{2.5}$ (mass of particles with an aerodynamic diameter below 2.5 μm) by TEOM
- PM_{10} (mass of particles with an aerodynamic diameter below 10 μm) by TEOM
- Elemental carbon by a light absorption method - Aethalometer.
- Ozone (O_3) by UV photometer analyzer (TECO Model 49).
- CO by non-dispersive infrared analyzer (Bendix Model 8501-5CA).
- Relative humidity and temperature measured continuously.
- SO_2 and NO_2 measured in Chelsea by Mass DEP.

Pollution metrics

- 24-hour means (midnight to midnight) when 16+ hourly measurements were available
- Five-day-means calculated whenever at least three 24-hour means were available.

Table 2: Distribution of the air pollutants measured at two sites in Boston between January 1995 and December 1997.

					Percentiles					
	Unit	Time period	N	Mean	5%	25%	50%	75%	95%	Maximum
PM$_{10}$	$[\mu g/m^3]$	1/95 –12/97	1066	19.3	7.8	12.6	17.2	24.2	37.0	62.5
PM$_{25}$	$[\mu g/m^3]$	1/95 –12/97	1052	12.7	4.6	7.6	10.8	16.1	26.6	53.2
BC	$[\mu g/m^3]$	1/95 – 3/97	769	1.38	0.49	0.86	1.21	1.74	2.84	4.92
CO	[ppm]	1/95 – 3/97	789	0.58	0.32	0.43	0.53	0.66	0.97	1.66
O$_3$	[ppm]	1/95 – 3/97	746	0.019	0.004	0.011	0.017	0.025	0.036	0.059
SO$_2$	[ppm]	1/95– 12/97	1072	0.007	0.001	0.003	0.005	0.009	0.019	0.087
NO$_2$	[ppm]	1/95 –12/97	1043	0.023	0.011	0.017	0.022	0.028	0.037	0.065
Minimum Temp	[°C]	1/95 –12/97	1075	7.03	-7.8	0.6	7.2	15.0	20.6	24.4
Relative Humidity	[%]	1/95 –12/97	1068	66.61	38.8	53.8	67.1	80.0	93.0	101.8

Table 3: Day-to-day correlation between air pollution and meteorological measures.

	PM$_{25}$	BC	CO	O$_3$	NO$_2$	SO$_2$	Min. Temp.	Humidity
PM$_{10}$	0.90	0.69	0.51	0.13	0.60	0.35	0.16	-0.01
PM$_{25}$		0.74	0.56	0.06	0.57	0.37	0.13	0.13
BC			0.75	-0.28	0.75	0.37	0.08	0.13
CO				-0.40	0.71	0.41	-0.13	0.22
O$_3$					-0.33	-0.26	0.46	-0.12
NO$_2$						0.51	-0.15	-0.03
SO$_2$							-0.31	-0.05
Min. Temp.								0.28

Table 3: Odds ratios for defibrillator discharges in association with an increase of the air pollutant from the 5th to 95th percentile adjusted for season, trend, and minimum temperature, humidity and day of the week.

		unit change	At least 1 event: 33 patients			At least 10 events: 6 patients		
			OR	CI-lower	CI-upper	OR	CI-lower	CI-upper
$PM_{2.5}$		[μg/m^3]						
	Same day	22	**0.87**	0.55	1.37	**0.82**	0.45	1.49
	2 days ago	22	**1.12**	0.75	1.66	**1.64**	1.03	2.62
	5-day mean	13	**0.88**	0.57	1.35	**1.22**	0.73	2.01
Black carbon		[μg/m^3]						
	Same day	2.4	**0.95**	0.51	1.78	**1.27**	0.57	2.81
	2 days ago	2.4	**1.33**	0.75	2.38	**1.90**	0.91	3.96
	5-day mean	1.4	**1.26**	0.66	2.39	**2.16**	0.96	4.86
Ozone		[ppm]						
	same day	0.032	**0.96**	0.47	1.98	**1.23**	0.53	2.87
	2 days ago	0.032	**1.53**	0.80	2.92	**1.51**	0.71	3.21
	5-day mean	0.014	**0.88**	0.53	1.45	**0.89**	0.49	1.60
Nitrogen dioxide		[ppm]						
	same day	0.026	**1.24**	0.75	2.06	**1.70**	0.91	3.18
	2 days ago	0.026	**1.48**	0.91	2.40	**2.79**	1.53	5.10
	5-day mean	0.016	**1.66**	1.01	2.72	**3.13**	1.76	5.56

Statistical Analyses

- Outcome defibrillator discharge following either tachycardia or ventricular fibrillation
- Logistic regression with fixed effect models with individual intercepts.
- Adjusted for trend, season, meteorological conditions, and day-of-the-week
- 24-hour air pollution concentrations considered for up to three days prior; five-day-mean concentration was used to evaluate cumulative effect of air pollution.
- Linearity of associations assessed by dividing pollutant concentrations into quintiles.
- Odds ratios (95% confidence intervals) presented for increase in air pollution from 5th to 95th percentile.

Discussion

Increased risk of a cardiac arrhythmia associated with elevated air pollution episodes in patients requiring implanted cardioverter defibrillators.

Odds of therapeutic intervention to treat ventricular tachycardia in patients with repeated discharges

- nearly tripled in association with an increase of 26 ppb NO_2
- increased 60% in association with an increase in $PM_{2.5}$ concentrations of 22 $\mu g/m^3$.

Associations are close to linear

Discharges do not occur immediately, but require induction time of several days.

Patients with repeated arrhythmias were most susceptible to air pollution effects.

WHICH ARE THE CHARACTERISTICS OF PARTICULATE MATTER THAT ARE IMPORTANT TO HUMAN HEALTH - PRIMARY VERSUS SECONDARY PARTICLES ?

Leendert van Bree, Flemming R. Cassee, and Peter J.A. Rombout
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Epidemiological studies have shown that short-term and long-term exposure to ambient air fine particulate matter (PM) is associated with substantial adverse health effects in urban and rural populations. The most responsible PM size fraction and composition, as well as their respective sources are unclear. These uncertainties reduce confidence in health risk assessment and standard setting, as well as in application of the most cost-effective emission and risk reduction, because such strategies should be targeted on responsible components related to the most important sources and emissions. Despite these uncertainties, regulatory decisions resulted recently in new and more tight air quality standards and limit values, forcing larger emission reductions than formerly anticipated. Both the scientific and regulatory community have called for the performance of toxicity studies revealing the most relevant causative PM particle size(s) and component(s) as well as plausible mechanisms underlying the adverse health effects.

Ambient PM is a complex and heterogeneous mixture, varying in particle size (coarse, fine, ultrafine) and composition, dependent of geographical location, meteorology, season, and type of source and emission. Many components may be adsorbed to the primary, carbonaceous core like e.g. acids and (partly) neutralized salts (as secondary products), aliphatic and (polycyclic) aromatic organic compounds (sometimes in oxidized form), metals (heavy metals, transition metals), and biological material like allergens, pollen fragments, and endotoxins.

This presentation will include a brief overview of a literature survey on potential mechanisms underlying PM_{10}-associated acute health effects. This survey shows that data from inhalation exposure studies with ambient PM and components performed with near-ambient particle mass and number concentrations are very limited and have not yet resulted in sufficient evidence to favor strongly a particular PM size fraction or composition. A limited number of relatively high dose PM mechanism studies (*in vivo* and *in vitro*) show that PM_{10} is able to induce acute lung injury and inflammation, whereby toxicity appears to correlate with the content of soluble, first-row transitions metals or organic PAHs and this response can blocked by chelation, extraction and radical scavengers. This might prudently point to a slightly more important role of the anthropogenic, carbonaceous (fine) fraction and might suggest that surface chemistry and (oxy)radical reactivity might be more important for biological activity than the generic, physical nature of ultrafine particles, the secondary fine components, or the non-soluble components of coarse particles. These findings are supported by animal toxicity data from our own laboratory on effects of fine and ultrafine carbonaceous (carbon black, diesel exhaust) and secondary (ammonium-bisulfate and -nitrate) aerosols in various animal models. Epidemiological data, however, have not yet shown sufficient evidence to conclude that health effects are preferentially associated with one of the three PM size subfractions or with a specific PM chemical composition. A few animal toxicity studies has shown significant positive interactions between PM and O_3 in inducing pulmonary toxicity, suggesting that the mixture of air pollution (oxidants) might be more important than PM components alone.

What New Research Results Are Emerging ?

Which are the characteristics of PM that are important to human health - primary versus secondary particles ?

Leendert van Bree
RIVM, the Netherlands

January 15, 1999
EC-HEI Joint Meeting, Brussels

The Particulate Matter Dilemma

A targeted standard setting and policy for the most cost-effective emission and risk control for ambient PM should be based on biological plausibility and responsible components related to the most important sources, BUT

the relative importance of these PM components, i.e.

- **PM_{10} , $PM_{2.5}$, or ultrafine PM, and**
- **primary, secondary, or biological components**

is largely unknown !!

Important Features of Epidemiological Data on Particulate Matter

- Associations at relatively low PM levels (< Limit Values, NAAQS)
- Associations with various air quality indicators like TSP, PM_{10}, $PM_{2.5}$, SO_4^{2-}, H^+, SO_2, NO_2, CO, or O_3, and under different air quality situations (wood smoke, wind blown dust, metals, acid aerosol, neutralized aerosol)
- Statistics and confounding still in discussion
- Susceptible groups

rivm

Impression of Relative Particle Sizes (Coarse, Fine, Ultrafine) of Ambient Particulate Matter

(Stone and Donaldson, 1998)

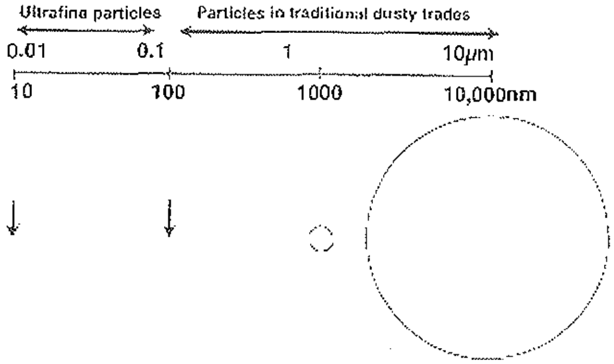


Figure 1. The relative size of particles which occur in various dusty trades compared with ultrafine particles such as those produced by the combustion of diesel oil.

rivm

Recommended PM Research Issues

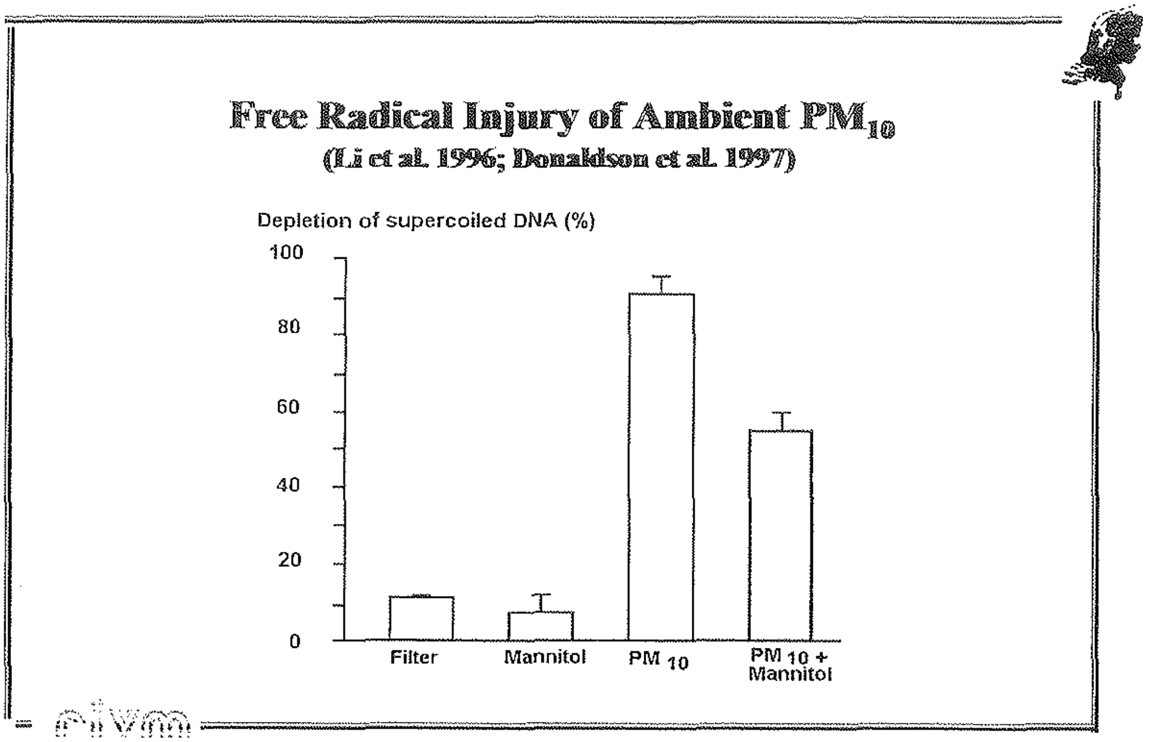
(e.g. by ESF, HEI, US-EPA, and US-NAS)

Apply studies to identify the physico-chemical properties of ambient PM and the air quality characteristics that produce or determine the PM-associated health effects:

- *air quality and source apportionment data*
- *epidemiological data (health effect - air quality contrast analyses)*
- *toxicological data (causality, plausibility, PM particle size and composition; combined effects with gases)*

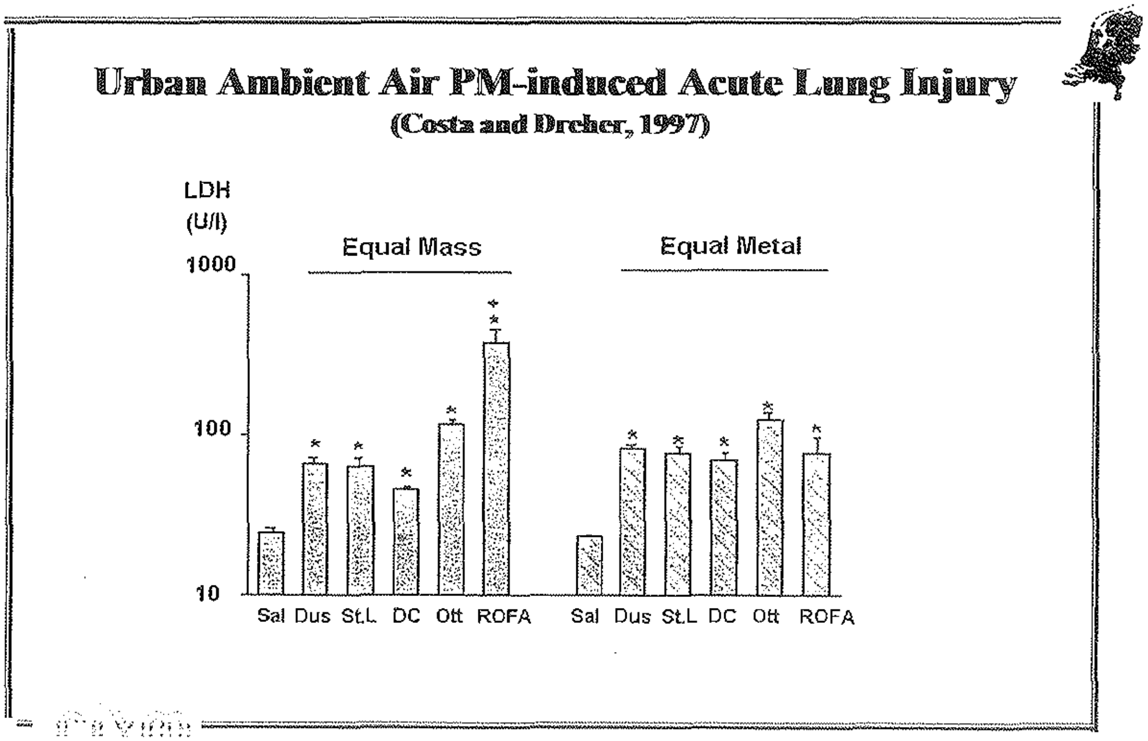
Possibly Important Components to Cause PM-Associated Health Effects

- Acids, or acidic or partly neutralized salts (coarse, fine)
  e.g. H_2SO_4, HSO_4^-, HNO_3, *and organics*
- Soluble, reactive components (coarse, fine, ultrafine)
  e.g. transition metals and oxy-PAHs
- Insoluble ultrafine particles
- Particles with adsorbed biological material
- Combined effects of particles with gases



Free Radical Injury of Ambient PM_{10}

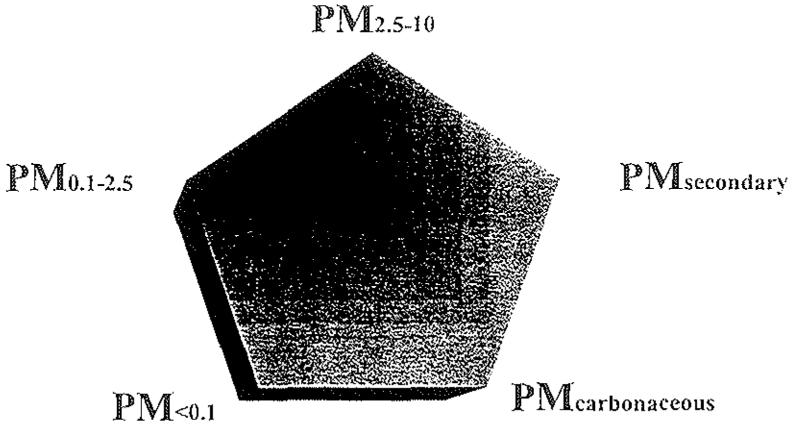
(Li et al. 1996; Donaldson et al. 1997)



Urban Ambient Air PM-induced Acute Lung Injury

(Costa and Dreher, 1997)

The RIVM PM “Pentagon” Approach



$PM_{2.5\text{-}10}$

$PM_{0.1\text{-}2.5}$

$PM_{secondary}$

$PM_{<0.1}$

$PM_{carbonaceous}$

RIVM
PM_{10} Toxicity Research Programme

- Review and Evaluation of Potential Mechanisms Underlying PM_{10}-Associated Health Effects (report in preparation)
- Development and use of a mobile particle concentrator for ambient PM_{fine} <u>inhalation</u> studies in animal models (causality and relationship with specific emissions)
- Animal toxicity studies with <u>inhalation</u> of fine and ultrafine model particles representing primary and secondary ambient PM fractions
- Toxicity testing of air sampled PM_{10}, $PM_{2.5}$, and $PM_{0.1}$
- Dosimetry models to predict dose and to extrapolate from animals to man

A Mobile Fine Particle Concentrator to Study the Direct Effects of Ambient PM_{fine} and the Relationship with PM Composition and with Specific Emissions and Sources

Study Design for Inhalation Toxicity of Fine and Ultrafine Model Particles Representing the Primary and Secondary PM

- Primary particles (carbon black, diesel exhaust)
- Secondary particles (ammonium-bisulfate and -nitrate)
- Freshly generated aerosols in 2 particle sizes (and with mass levels < 2000 µg/m³): fine ~ 0.3 - 0.7µm
  ultrafine ~ 0.04 - 0.09µm
- Exposures during 4 hr/day on 3 consecutive days in asthmatic mice, pulm. hypertensive rats, and controls
- Post-exposure analysis of airway inflammation, function, and morphology

Inhalation Toxicity of Model Particles

(Cassee *et al.* 1999)

Compound	Fine		Ultrafine	
	Asthma	PH	Asthma	PH
Bisulfate	-	LDH/PMN ↑	-	-
Nitrate	NAG/PMN ↑	-	NAG ↑	
Carbon Black	PMN ↑	-	-	-
Carbon + Nitrate	*N.D.*	-	*N.D.*	-

Secondary fine/ultrafine : <0.35 mg/m^3

Fine CB < 2 mg/m^3, Ultrafine CB < 0.015 mg/m^3

Summary of Toxicological Evidence for a Role of PM Subfractions in PM-associated Health Effects

- Current realistic inhalation data do not strongly favor a particular PM size fraction or composition
- Secondary PM is not or hardly able to induce general effects and inflammation at near-ambient levels (e.g. only marginal effects of H_2SO_4 exposure of mild asthmatics)
- Primary (fine) PM might play a slightly more important role because animal toxicity data on high doses suggest:
  - ⇒ inflammation correlates with levels of soluble, adsorbed components (transition metals, reactive PAHs)
  - ⇒ effects can be blocked by chelation, extraction, radical scavenging
  - ⇒ few data on ambient TSP, PM_{10}, and emission source-PM (diesel exhaust, oil fly ash)

H.- ERICH WICHMANN

GSF - Forschungszentrum für Umwelt and Gesundheit, Germany

SHORT TERM EFFECTS OF FINE AND ULTRAFINE AIR POLLUTION ON MORTALITY AN ONGOING STUDY

C. Spix[1a,2], T. Tuch[1a,2], J. Heyder[1b], C. Roth[1b], H.E. Wichmann[1a,2]

1) GSF National Research Center for Environment and Health, Neuherberg, Germany a) Institute of Epidemiology, b) Institute of Inhalation Biology
2) Ludwig-Maximilians-University, München, Germany.

Study design: Relating daily mortality counts in Erfurt, Germany, to daily measurements of particulate size classes from 0.01 µm to 2.5 µm, standard PM2.5 and PM10 measurements, and gaseous components. Measurements have begun in summer 1995. In the first study year we observed 1919 deaths from natural causes and 1891 in the second, that is 5.2 deaths per day. Mortality data is based on individual death certificates and allows subsetting classes of cause, age and place of death.

Data: We have shown previously, that using all the information from the death certificates gives a better classification of causes than relying on the officially recorded underlying cause of death. Based on the recoding of a random sample we could show, that coding is accurate in broad classes.

Analysis: The analysis is performed using Poisson regression techniques controlling for season, epidemics, calendar patterns and weather. Nonparametric smoothing techniques are used alongside parametric models.

Results: We will present results for particulates in the size classes 0.01-0.1 µm (ultrafine), 0.1-0.5 µm (fine) and 0.5-2.5 µm („coarse") for total deaths and by cause of death for the 1st and 2nd study year. The average mass concentrations are 0.7 µg/m^3, 22.3 µg/m^3 and 5.1 µg/m^3 and the average numbers per m^3 are 15960, 2254 and 20 for the ultrafine, fine and coarse fraction. The ultrafine fraction contains most of the particle number, while the fine fraction contains most of the mass. For the first year we saw some effects in the smaller size fractions which were most pronounced in cases with cardiovascular and respiratory diseases combined. The analysis of the second year is in progress.

Conclusions: No health effect conclusions are available as yet since the study is still ongoing. However the results indicate that effects on daily mortality exist although the ambient concentrations are not very high.

Epidemiological studies on health effects of fine and ultrafine particles in Germany

H.- Erich Wichmann and Annette Peters

Institute of Epidemiology, GSF-National Research Center
for Environment and Health, Neuherberg

Presentation at the Institute Meeting of the EU and HEI:
'Health Effects of Fine Particles: Key Questions and the
2003 Review' Brussles, Belgium 14./15. January 1999

Contents

- Introduction
- available knowledge
- ongoing research
- future needs
- summary

Introduction

- sources of fine and ultrafine particles
- number vs surface vs mass

Epidemiological Studies

- 91 - 92 Asthma Study Erfurt (GSF - Pilot)
- 95 - 99 Mortality Study Erfurt (HEI)
- 96 - 99 Cardiovascular Disease Study Erfurt, Amsterdam, Helsinki (EU - ULTRA)
- 99 - 01 Myocardial Infarction Study Augsburg (HEI)

91 - 92 Asthma Study Erfurt (GSF - Pilot)

Peters, Tuch, Heinrich, Heyder, Wichmann

- panel study with adult asthmatics
- fine and ultrafine particles, PM_{10}
- Symptom diary and peak expiratory flow
- Regression analyses with adjustment for trend, meterology and weekend.

g Institute of Epidemiology

91 - 92 Asthma Study Erfurt (GSF - Pilot)

Results

- significant associations between exacerbation of asthma and 5-day means of fine and ultrafine particles
- increase in symptoms, decreases in lung function
- the strongest effect was observed for paricle number concentrations (ultrafine particles)

Peters et al., 1997 Am J. Resp. Crit. Care Med.

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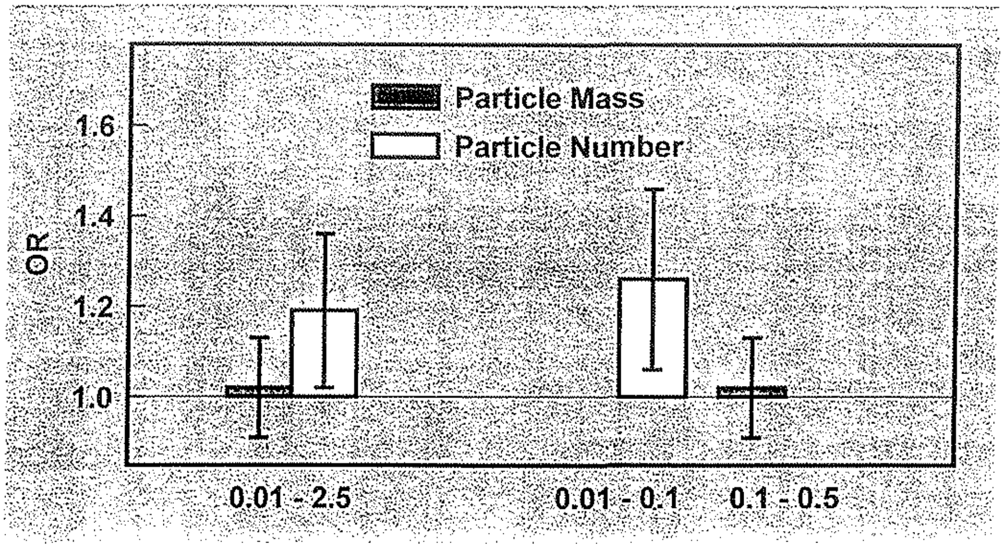
Number concentration of particles was associated with an increase in cough



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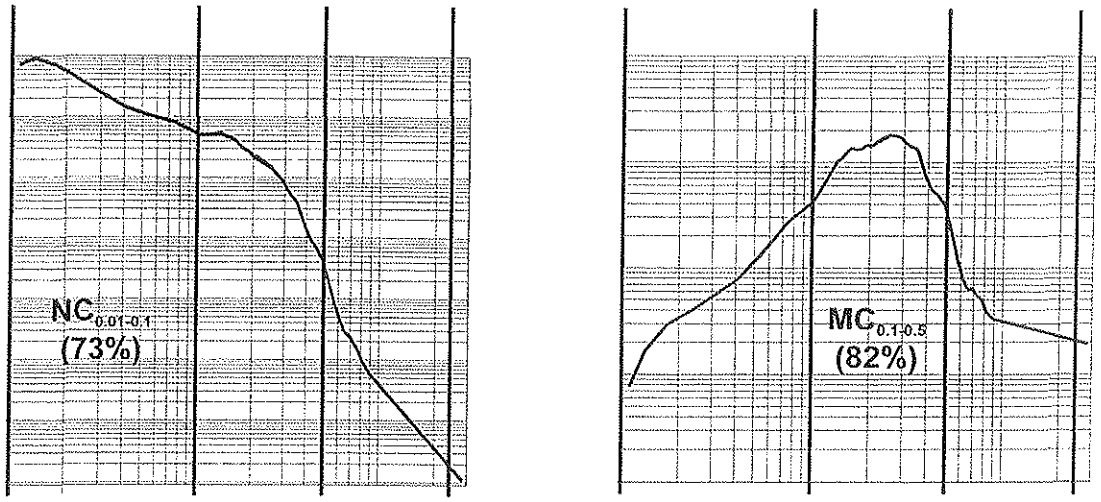
Mean number and mass distribution of ambient fine particles (Erfurt 91/92)



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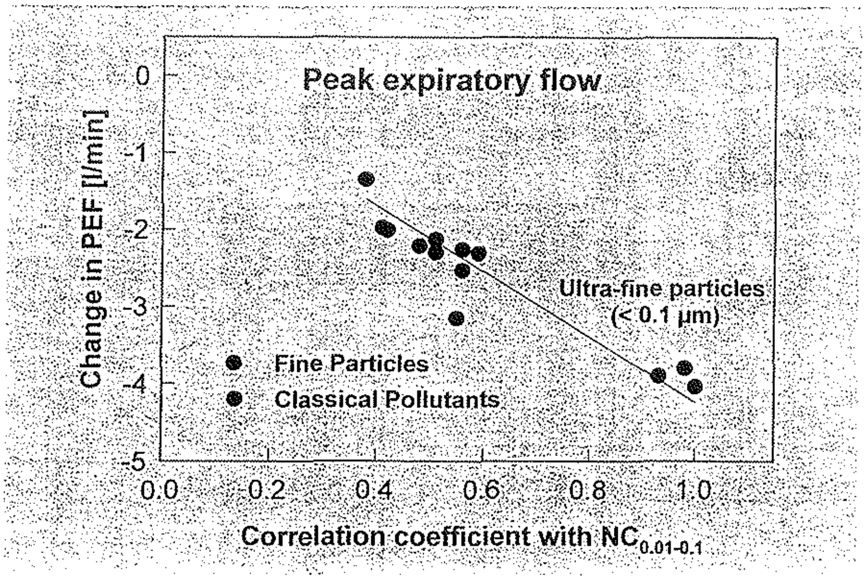
Associations between air pollutants and decreases in PEF for adults



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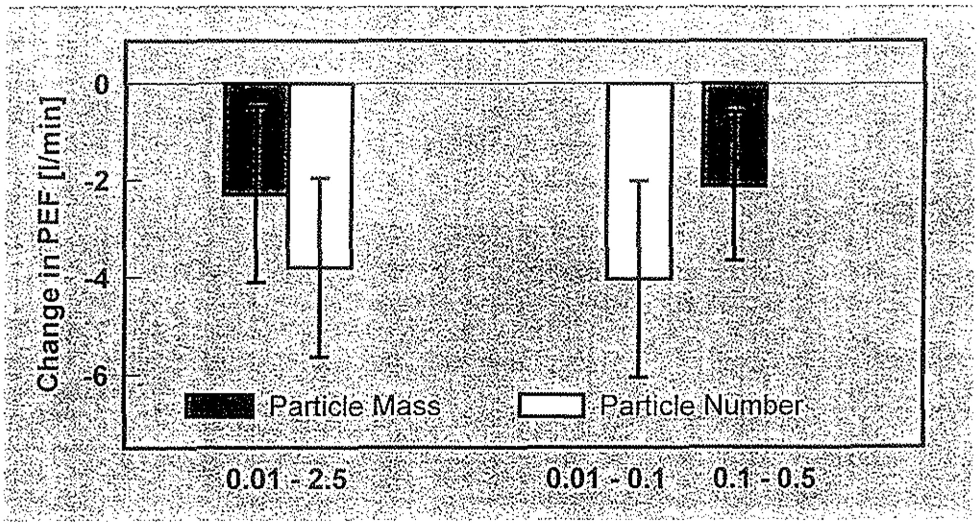
Number concentration of particles was associated with decreases in peak expiratory flow



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95 - 99 Mortality analyses Erfurt (HEI)

Spix, Tuch, Kreyling, Wittmaack, Wichmann

- daily mortality (total and cause-specific)
- fine and ultrafine particles, PM_{10}, $PM_{2,5}$, elemental composition (PIXE)
- Poission regression analyses with adjustment for trend, season, meterology and influenza epidemics

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95 - 99 Mortality Erfurt (HEI)

Results

- decreases in particle mass (fine particles), increases in particle number (ultrafine particles) over time
- interim analyses (2 years of data): Borderline significance of increases in mortality in assocaition with fine and ultrafine particles

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96 - 99 Cardiovascular disease study (EU - ULTRA)

Heinrich, Peters, Tuch, Ibald, Roth, Heyder, Kreyling, Wichmann; Brunekreef et al.; Pekkanen (PI) et al.

- comparison of fine and ultrafine particles in 3 cities
- 150 elderly with cardiovascular diseases (panel study)
- daily symptoms, biweekly EKG, lung function.

96 - 99 Cardiovascular disease study (EU - ULTRA)

Results

- good agreement between the aerosol spectrometers, sepecially for ultrafine particles
- concentrations of ultrafine particles comparable throughout the locations
- epidemiological studies are ongoing

99 - 01 Myocardial Infarction Study

Peters, Löwel, Tuch, Heinrich

- case-crossover study based on the Coronary event registry Augsburg
- cases: Survivors of an acute myocardial infarction
- measurements of fine particle mass and total number concentrations on an hourly basis

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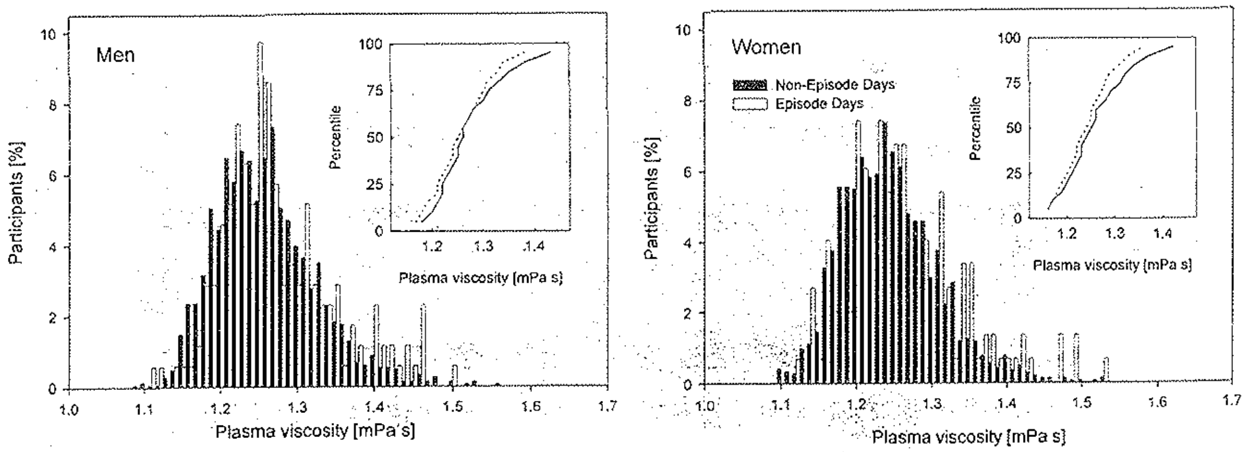
Elevated levles of plasma viscosity during the 1985 air pollution episode



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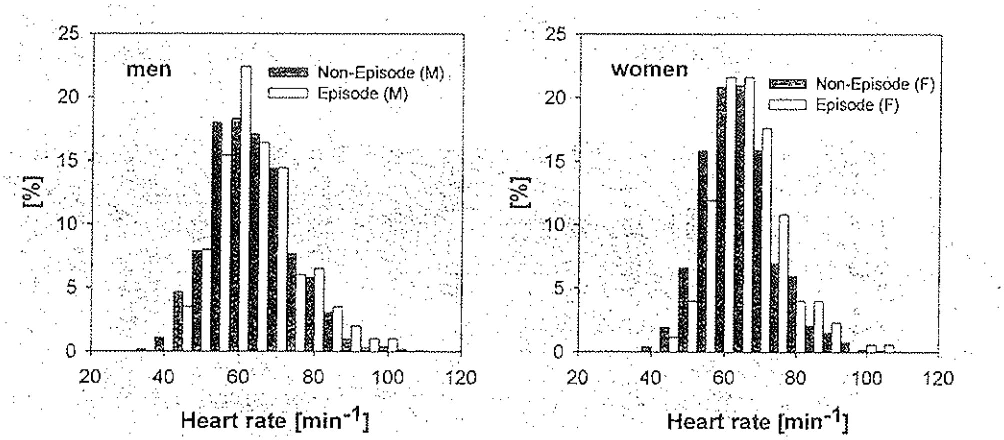
Heart rate increases during the episode



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Relevance for regulatory purposes

- reduction of mass does not result in a reduction of the number of particles
- modern (diesel) motors produce less fine but more ultrafine particles than old motors

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Summary

- asthmatics: effects of ultrafine particles stronger than effects of fine particles
- mortality: clear until summer 1999
- cardiovascular effects: clear until 2000/01
- myocaridal infarction: clear until 2001

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Research needs

- chemical characterisation of fine and ultrafine particles
- Is the number or the surface more relevant than the mass?
- Are particles from specific sources more toxic?

g Institute of Epidemiology

KLEA KATSOUYANNI

University of Athens, Greece

SHORT-TERM EFFECTS OF AIR POLLUTION ON HEALTH
A EUROPEAN APPROACH TO
METHODOLOGY, DOSE-RESPONSE ASSESSMENT
AND EVALUATION OF PUBLIC HEALTH SIGNIFICANCE
(THE APHEA2 PROJECT)

Klea Katsouyanni, Department of Hygiene and Epidemiology, University of Athens Medical School, Greece

The APHEA2 project started on February 1st, 1998. It is sponsored by the European Commission DGXII, Contract number ENV4-CT97-0534, under the ENVIRONMENT AND CLIMATE 1994-1998 Programme. Twenty research groups, several of which represent research networks, from 19 different European countries, are participating. The objectives of the project are: 1. To investigate dose-response relationships between air pollutants and daily mortality and hospital admissions, 2. To test hypotheses about mortality displacement caused by the short-term effects of air pollution, 3. To investigate regional differences and effect modifiers and 4. To conduct a second stage analysis with data from 32 cities on mortality and 7 cities on hospital admissions and, possibly, a combined analysis with the U.S., H.E.I. sponsored NMMAPS project.

Air pollution data include measurements of ambient particles (either as PM_{10} or black smoke), nitrogen dioxide (NO_2), ozone (O_3), sulphur dioxide (SO_2) and carbon monoxide (CO). Most air pollution measurements will come from operating fixed site monitors. To make exposure data as comparable as possible criteria are applied concerning placement of monitors and measurement completeness. Mortality data will be available from 34 cities, 9 of which are in Central-Eastern European countries. Hospital admissions data from 7 cities will be analysed. Analysis of mortality time series will be done by cause of death and age. Respiratory and cardiac hospital admission will be analysed separately. Currently, the individual city analysis is under way.

The APHEA2 project is a continuation of the older APHEA (Air Pollution and Health: a European Approach) project, sponsored also by the E.C. under ENVIRONMENT 1991-1994. The APHEA project was the first large scale multi-center project addressing short-term effects of air pollution on health and included 11 research groups and data from 15 cities. When the project started (in 1993) there were few available results from Europe analysing current air pollution data. The objectives of this older project were to provide estimates of air pollution short-term health effects in Europe, to standardise the methodology of analysing epidemiological time series data and applying a meta-analysis procedure. The project finished in 1995 and the results have been published and had a considerable impact in policy making at the national and E.U. level. All pollutants studied, with a partial exception of NO_2, were found to have small acute effects on daily total, cardiovascular and respiratory mortality. Particulate matter and ozone levels were consistently associated with respiratory and COPD admissions. NO_2 levels were associated with asthma admissions. The estimated effects for particulate matter were consistent with those observed in the U.S. studies but they were on the lower side of the U.S. range of estimates. The above effects were observed in locations were, on most days, air pollution levels were well below the set standards.

Several open questions remained. Are there regional differences in air pollution health effects? What is the independent effect of each pollutant? Is there synergy? What is the extent of mortality displacement caused by the short-term effects of air pollution? The APHEA 2 project aims at addressing the above questions.

There are advantages and disadvantages in such a large scale multicenter project. Among the advantages, the use of a larger data base and a stardardised protocol, the varying pollutant mixes, the different environmental and socioeconomic conditions, the transfer of know-how and the joining of scientific forces and different perspectives should be mentioned. The major disadvantage is the difficulty in managing and coordinating all the research groups.

Short-term effects of Air Pollution on Health
a European Approach
using epidemiological time-series data

THE APHEA PROJECT

Environment 1991-94 Programme

CONTRACT NUMBER EV5V - CT92 -0202

PUBLICATIONS AND IMPACT

ATHENS, MARCH 1998

SHORT-TERM EFFECTS OF AIR POLLUTION ON HEALTH
A EUROPEAN APPROACH USING EPIDEMIOLOGIC TIME-SERIES DATA

THE APHEA PROJECT

JANUARY 1, 1993 - OCTOBER 31, 1995
34 MONTHS

APHEA PROJECT OBJECTIVES:

- TO PROVIDE QUANTITATIVE ESTIMATES OF SHORT-TERM HEALTH EFFECTS OF CURRENT LEVELS OF AIR POLLUTION IN EUROPE
- TO STANDARDISE THE METHODOLOGY OF ANALYSING EPIDEMIOLOGICAL TIME SERIES DATA
- TO DEVELOP AND APPLY A WAY TO COMBINE THE RESULTS (META-ANALYSIS)

APHEA PARTICIPATION

11 RESEARCH GROUPS FROM
10 DIFFERENT EUROPEAN COUNTRIES

DATA FROM 15 CITIES WITH
MORE THAN 25,000,000 INHABITANTS

APHEA PROJECT

ADVANTAGE OF THE EUROPEAN DIMENSION:

- USE OF A MUCH LARGER DATA BASE
- STANDARDISED PROTOCOL
- A PRIORI PLANNED SECOND-STAGE ANALYSIS (META-ANALYSIS)
- VARYING POLLUTANT MIXES
- DIFFERENT ENVIRONMENTAL AND SOCIOECONOMIC SITUATIONS.
- TRANSFER OF KNOW-HOW
- JOINING SCIENTIFIC FORCES AND DIFFERENT PERSPECTIVES

APHEA project: Summary effects of pollutants on the daily total number of deaths in Western European cities*.

Pollutant	No of cities	Relative Risk per 50μg/m^3 increase	95%CI
Black smoke (24-h)	4	1.029	1.021, 1.037
PM_{10} (24-h)	5	1.021	1.012, 1.030
SO_2 (24-h)	7	1.035	1.020, 1.050
NO_2 (1-h)	5	1.021	1.007, 1.034
Ozone (1-h)	4	1.029	1.010, 1.049

*From: Katsouyanni et al, BMJ 1997; 314: 1658-63
Touloumi et al, Am J Epidemiol 1997; 146: 177-85

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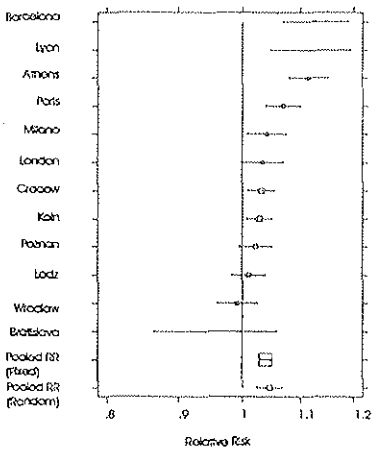
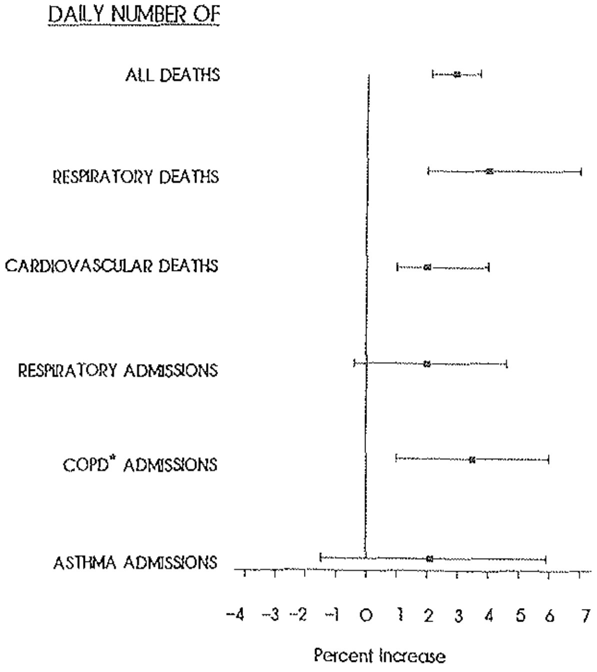
* In Central-Eastern European cities considerably lower effects were observed for particulate matter and SO_2 (the only pollutants which were studied), although in some instances they were statistically significant.

Estimated individual and pooled relative risks and 95% CI
The relative risks are for a 100 μg/m³ increase in SO_2-24h



APHEA PROJECT: MAIN RESULTS

Percent increase in daily health outcomes when black smoke levels rise by 50 μg/m³ in Western European cities



* COPD: CHRONIC OBSTRUCTIVE PULMONARY DISEASE

APHEA PROJECT
MAIN RESULTS

The individual city estimates were in some instances heterogeneous.

Several characteristics of the cities and their populations were examined to see if this heterogeneity could be explained.

The only clear difference was between Western and Eastern European cities: The effects seen for a given increase in a pollutant, were smaller in the Eastern cities.

This issue needs to be further investigated.
The difference may be due to:
- the method of analysis;
- to differences in the susceptible population;
- to different pollution mixes ;
- to the measurement locations & methods etc.

CONCLUSIONS FROM
THE APHEA PROJECT SO FAR:

1. All pollutants studied (ambient suspended particles, sulphur dioxide, ozone and nitrogen dioxide),
   with a partial exception of NO_2,
   have small acute effects on the daily total, cardiovascular and respiratory mortality.

2. Particulate matter and ozone levels were consistently associated with respiratory and COPD admissions.

CONCLUSIONS FROM THE APHEA PROJECT SO FAR:

3. Nitrogen dioxide levels were associated with asthma admissions.

4. The estimated effects for particulate matter were consistent with those observed in the U.S. studies, but on the lower side of the U.S. range.

5. The above effects were observed in locations where, in the majority of days, air pollutant levels are well below the set (W.H.O., E.C., U.S.E.P.A.) standards.

THE APHEA PROJECT PUBLICATIONS AND IMPACT

- The results described have been published in:
  - 22 papers (in 3 languages) in scientific journals
  - 5 papers in conference proceedings
  - 23 abstracts from conference presentations

- They have received press publicity in several European countries.

- They have been used by:
  - The World Health Organisation in their new revised air quality guidelines, which are in press
  - The European Commission in their proposal (8.10.97) for Council Directive relating to limit values for sulphur dioxide, oxides of nitrogen, particulate matter and lead in the ambient air.

- APHEA participants have been invited to present the project in several workshops, meetings and conferences and considerable debate has been initiated.

APHEA PROJECT: THE FUTURE

- SEVERAL OPEN QUESTIONS REMAIN:
  - ARE THERE REGIONAL DIFFERENCES IN AIR POLLUTANTS HEALTH EFFECTS? WHY?
  - WHAT IS THE EFFECT OF EACH POLLUTANT INDEPENDENT OF THE LEVELS OF THE OTHER POLLUTANTS?
  - IS THERE SYNERGY (COMBINED EFFECT OF TWO OR MORE POLLUTANTS)?
  - HOW MUCH PREMATURE ARE THE DEATHS CAUSED BY AIR POLLUTION? (ONE DAY? SEVERAL DAYS? ONE OR MORE MONTHS? ONE OR MORE YEARS?)
- TO ADDRESS THESE, THE APHEA2 PROJECT HAS BEEN INITIATED.

APHEA2 PROJECT

SHORT-TERM EFFECTS
OF AIR POLLUTION ON HEALTH:
A EUROPEAN APPROACH
TO METHODOLOGY, DOSE-RESPONSE
ASSESSMENT
AND EVALUATION OF PUBLIC HEALTH
SIGNIFICANCE

E.C. ENVIRONMENT AND CLIMATE PROGRAMME
CONTRACT NUMBER ENV-CT97-0534

STARTING DATE: FEBRUARY 1, 1998
DURATION: 36 MONTHS

APHEA participation

20 Research Groups (Several represent research networks

19 Different European Countries (5 are Central -Eastern European)

34 cities for Mortality Analysis

7 cities for Hospital Admissions Analysis

[illegible]

1. To investigate dose-response relationships between air pollutants (including ambient particles, carbon monoxide, sulphur dioxide, nitrogen dioxide and ozone) and total and cause-specific daily mortality and hospital respiratory admissions.

2. To test hypotheses about mortality displacement caused by the short- term effects of air pollution (i.e. the time hours, days, months etc. by which deaths are brought forward or, in other words, how "premature" they are) which determines to a large extent the public health significance of these effects.

APHEA 2: OBJECTIVES (2)

3. To investigate the reasons for the observed differences in the effect parameters between European regions. This will also lead to better understanding and quantifying the independent role of each pollutant in the air pollution mix.

4. To conduct a second-stage analysis with data from 32 cities on mortality and 7 cities on hospital admissions, in order to evaluate determinants of the effect size.

The results will also be seen in the context of APHEA 2 - NMMAPS collaboration. NMMAPS (The National Mortality, Morbidity and Air Pollution Study) is a large U.S. project with similar objectives and design (Investigators: Drs J. Samet (PI) and S. Zeger from Johns Hopkins School of Public Health and Drs D.Dockery and J. Schwartz from the Harvard School of Public Health)

APHEA 2
PROJECT MANAGEMENT SCHEME

1. Steering Committee
2. Statistical Advisory Group
3. Data compilations: All Groups
4. Mortality Analysis: dose - response
   Implementation: Greek Group
   Mortality Analysis: Harvesting
   Implementation: German Group
   Hospital Admissions Analysis
   Implementation: London & Paris Group
   Meta-analysis (second - stage analysis)
   Statistical Advisory Group
5. One Workshop was organised in November 1998 and one will be organised in spring 1999 to address the time-series analysis issues in APHEA 2

DATA (1)

Air Pollution

Recent time-series (in most instances up to 1995) for each pollutant (i.e. NO_2, O_3, SO_2, CO and particles) will be analysed.

- Most air pollution measurements will come from operating fixed site monitors.
- Criteria for standardizing the data will be applied concerning placement of monitors and measurement completeness.
- All cites will provide some gravimetric measurements of particles (i.e. PM_{10}), in addition to Black Smoke (reflectometry).
- If the time series have missing values, these will be filled-in using a standard procedure.
- The air pollutant indices to be used (time averaging etc) have been chosen to be compatible with W.H.O. Air Quality Guidelines (1998 in press).

DATA (2)

MORTALITY

- Mortality data sets will be available from 34 cities or areas: Athens, Erfurt and Suhl, London and Birmingham, Paris, Marseilles and Lyon, Barcelona, Madrid, Valencia and Bilbao, Milano, Torino, Roma, The Netherlands, Helsinki, Dublin, Tel Aviv and Haifa, Istambul, Basel, Geneva and Zurich, Cracow, Lodz, Poznan and Wroclaw, Prague, Ljubljana, Budapest and Bucharest, Zagreb, Stockholm.

- Mortality data will be analysed by cause of death, sex and age group

Age-group: 14-64 years; 65-74 years; 75+years

Cause of death: Total mortality (excluding deaths from external causes, *i.e.* ICD9 ≥ 800)

Respiratory deaths (ICD9: 450-519)

Cardiovascular deaths (ICD9: 390-459)

HOSPITAL ADMISSIONS

→ Hospital admissions data will be analysed from Paris, London and Birmingham, Milano and Rome, The Netherlands and Barcelona.

→ Hospital admissions data will be analysed by diagnosis sex and age group

Respiratory admissions (ICD9: 460-519)

age-groups:0-14 yrs; 15-64 yrs; 65-74 yrs; 75+ yrs; all ages

COPD (ICD9: 490-496 excluding 493)

age-groups: All ages

Asthma (ICD9: 493)

age-groups: 0-14 yrs; 15-64 yrs; all ages

Cardiac admissions (ICD9: 390-429)

age groups: 15-64 yrs; 75+ yrs; all ages

Isheamic heart disease (ICD9: 410-414)

age groups: 15-64 yrs; 75+ yrs; all ages

JONATHAN SAMET

Johns Hopkins University, United States

NMMAPS: NATIONAL MORTALITY, MORBIDITY AND AIR POLLUTION STUDY.

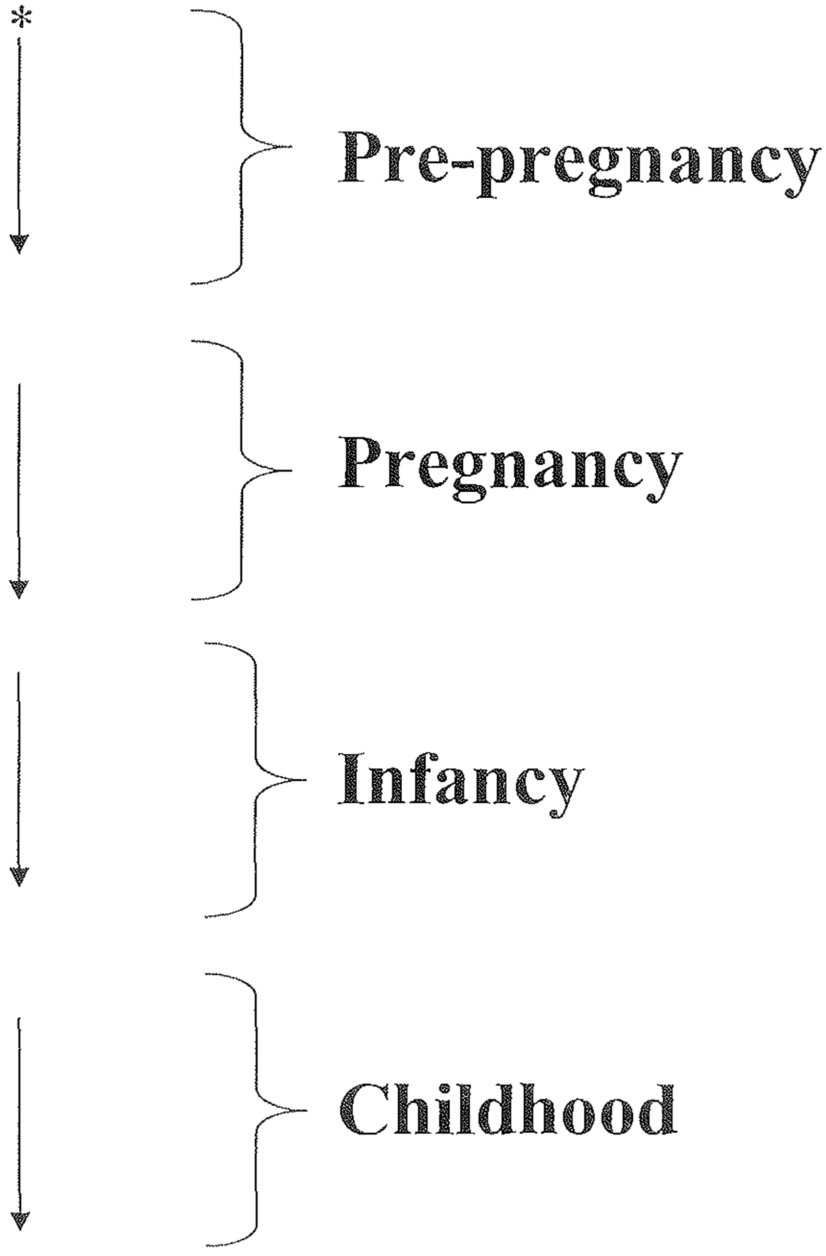
J. M. SAMET, S. Zeger, F. Dominici, J. Xu, D. Dockery, and J. Schwartz. Johns Hopkins University School of Hygiene and Public Health, Baltimore, Maryland, and Harvard School of Public Health, Boston, Massachusetts.

Time-series studies have shown associations between air pollution concentrations and morbidity and mortality. These studies have largely been conducted within single cities with varying methods. Key questions remain unaddressed concerning the findings, including 1) the extent of the heterogeneity of air pollution effects across locations and its sources; 2) the effect of measurement error on the estimated effect of air pollution; and 3) the public health significance of the short-term associations. NMMAPS comprises the development of methods to address these questions and their application to national data sets on mortality and hospitalization among persons 65 years of age and older, as an index of morbidity. For analyzing mortality data from multiple locations and summarizing the findings, a Bayesian hierarchical regression method has been developed and applied to data from 8 cities. Frequency domain regression methods have been used to assess mortality displacement, with the initial finding that air pollution effects can be assessed in time-response bands that are relatively resistant to short-term mortality displacement. The consequences of measurement error are being comprehensively exposured using available data sets. Data bases have been assembled on mortality of the 100 largest U.S. cities and for morbidity. In the next phase of NMMAPS, the morbidity and mortality analyses will be completed and the combined analyses of morbidity and mortality will be undertaken. The methods of NMMAPS should prove useful for future surveillance for the health effects of air pollution.

The NMMAPS/APHEA Collaboration

- Initiated summer, 1998
- Opportunity to
  - address shared methodologic concerns
  - share analytic methods
- Potential for combined analysis

Windows of Exposure



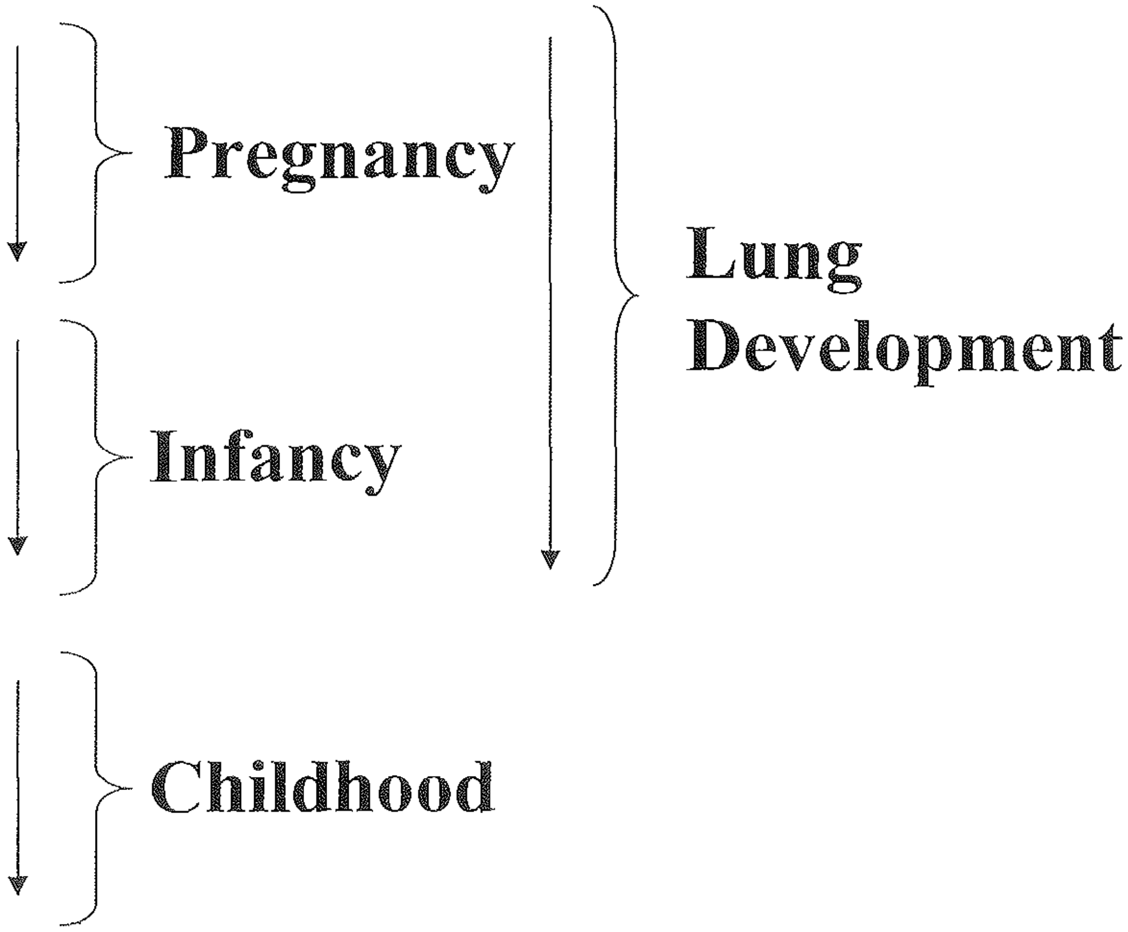
Pre-pregnancy

Pregnancy

Infancy

Childhood

Windows of Susceptibility



Pregnancy

Infancy

Childhood

Lung Development

NMMAPS

National Morbidity, Mortality and Air Pollution Study

⇒ **OBJECTIVE: To develop a national approach to assessing the health effects of air pollution**

- ☐ **Multi-city analysis of air pollution and mortality**
- ☐ **Multi-city analysis of air pollution and hospitalization of the elderly**
- ☐ **Combined analyses of air pollution and morbidity and mortality**
- ☐ **Key methodologic issues**

MATTI JANTUNEN

National Public Health Institute, Finland

THE ROLE OF MULTICENTER STUDIES IN AIR POLLUTION RESEARCH
THE *EXPOLIS* EXPERIENCE

Matti Jantunen, KTL, Environmental Hygiene, Kuopio, FINLAND

ABSTRACT

EXPOLIS goals

Epidemiological literature of the 1990's has revealed surprisingly large public health impacts associated with present common air pollution levels in North American and European cities. Any causal explanation of the health effects of air pollutants must go through exposure, yet, prior to *EXPOLIS* no large, population based air pollution exposure studies have been conducted in Europe, and consequently no European database of air pollution exposures of urban populations has existed until now. *EXPOLIS* is a European multicentre study for measurement of air pollution exposures of working age urban populations during workdays. The selected urban areas are Athens, Basel, Grenoble, Helsinki, Milan and Prague. The main objectives of *EXPOLIS* are:

* To assess the exposures of European urban populations to major air pollutants.

* To analyse the personal and environmental determinants and interrelationships to these exposures.

* To develop an European database for simulation of air pollution exposures.

These objectives were pursued by measuring the personal exposures, home indoor and outdoor and workplace levels of $PM_{2.5}$, VOCs and CO of approximately 500 subjects representing the adult populations of the selected cities.

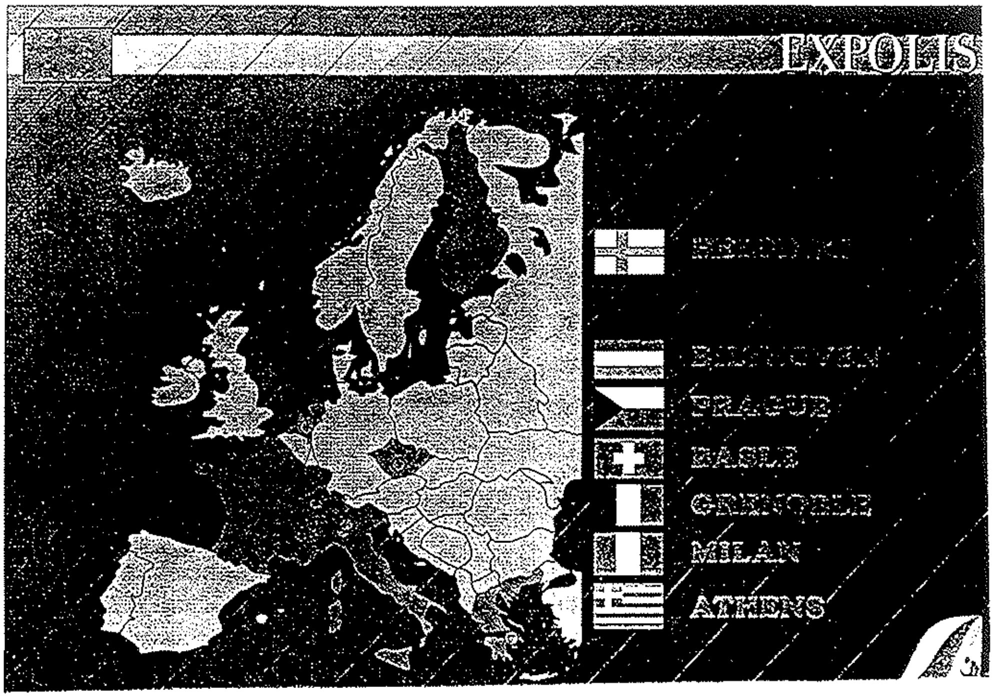
Field work

continued from fall of 1996 to winter of 1997-98. To assure comparability of the data from the 6 cities in 6 countries, a strict QA/QC protocol was established, standard operating procedures were prepared for all subject, laboratory and field procedures, and identical sampling equipment, operating procedures, time-microenvironment-activity diaries, questionnaires, database and data entry tools were used in each centre. VOC laboratory analyses were intercalibrated by the European Commission / Joint Research Centre (EC/JRC) Environment Institute in Ispra. Other techniques were intercalibrated between the teams.

Reasons and consequences of multiple centres

The multicentre nature of the *EXPOLIS* study is based on the need to acquire exposure data for modelling purpose from a wide variety of qualitatively and quantitatively different populations, cultures, climate zones and urban centres, bu also on the need to involve the best available expertise from exposure monitoring, modelling and exposure data application points of view.

A multicultural and -lingual study like *EXPOLIS*, where multiple compounds are monitored in multiple microenvironments, needs a great deal of practical everyday problem-solving and other communication to ensure on the one hand a common practice and comparable study results, and on the other hand minimum data losses. The junior researchers were trained at the different phases of the study together in *EXPOLIS*-Workshops in Prague (Apr. 1996), Helsinki (Sept. 1996), Grenoble (Mar. 1997), and Bilthoven (Feb 1998). In each center one team member was assigned to one or more of the following contact groups: *Equipment*, *Database* and *VOCs*. *QA/QC* and *Privacy Protection* responsibilities lay within the principal investigators. Communication occurred mostly via E-mail, but each team member was also assigned a GSM telephone with contact numbers of all other team members to ensure fast access when and where problems were encountered in the field or laboratory.



PM exposure assessment alternatives

- **Fixed site ambient air monitoring data**
  - routine monitoring; widely available data
  - which location criteria?
  - how good an estimate for personal exposure?
  - certainly ignores individual differences
  - but d(PM exposure) vs d(ambient PM)?

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PM exposure assessment alternatives

- Microenvironmental monitoring
  - measure most important micro-environments, e.g. home, workplace, indoor/outdoor, transport, street, public spaces
  - 'important'= microenvironments with high time allocation or high concentration

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PM exposure assessment alternatives

- Personal monitoring
  - exposure monitor carried through all the time and microenvironments 24 h...1 wk
  - time-microenvironment-activity monitoring by paper or electronic diary
  - very labor intensive, subject invasive and expensive technique

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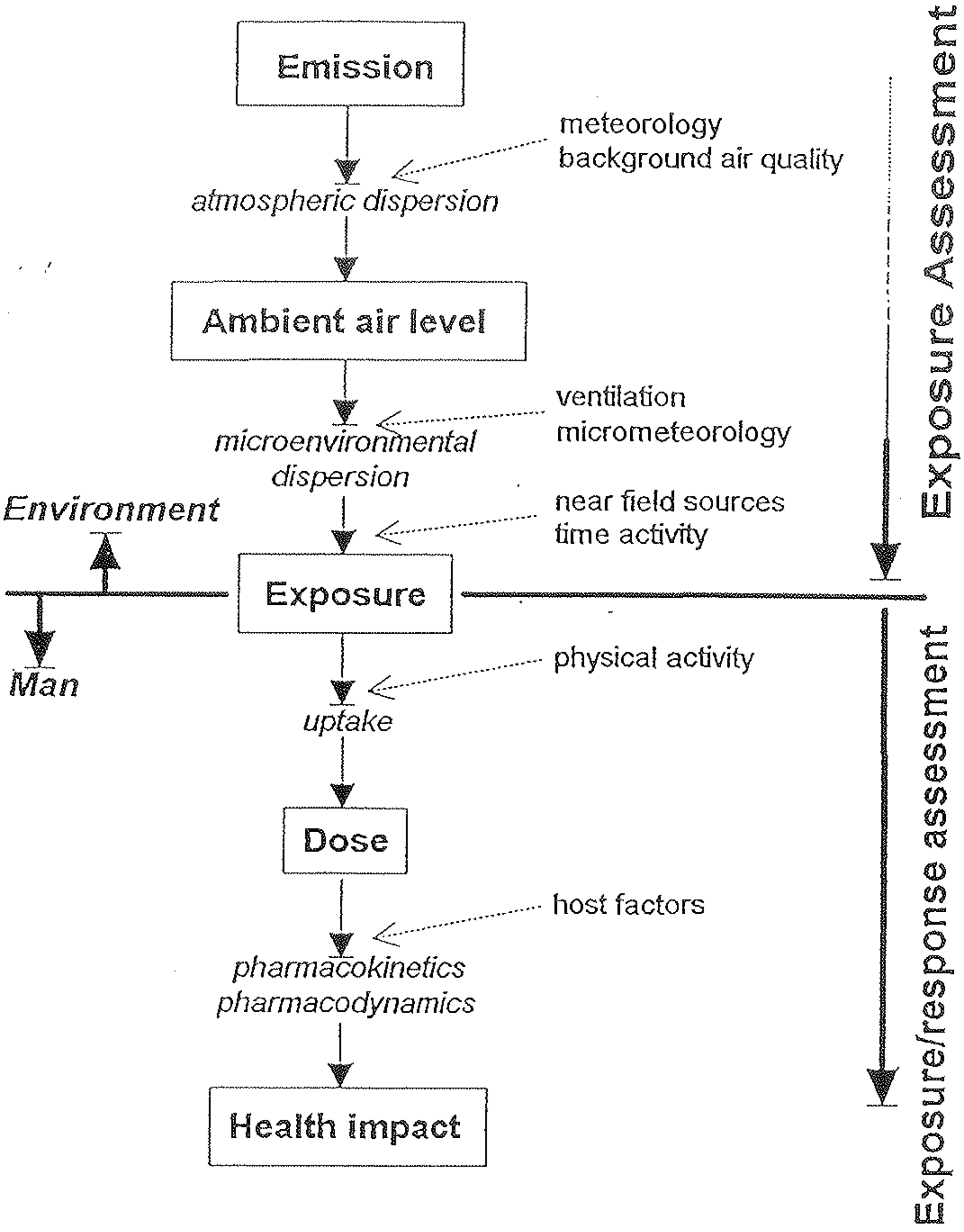
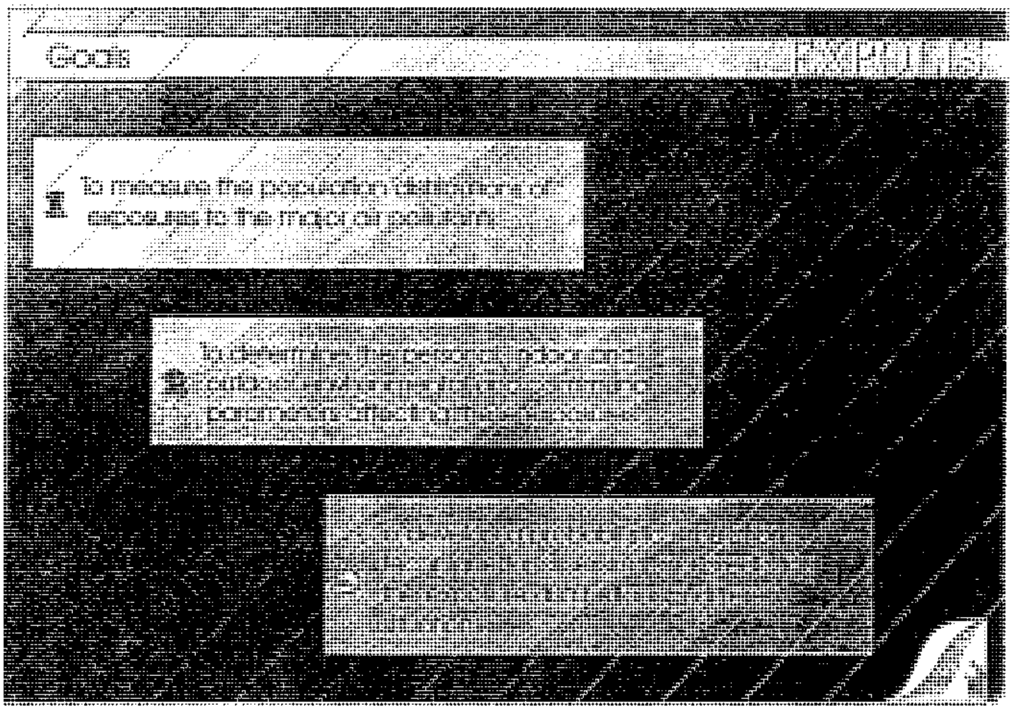


Figure 10:3. The flowchart from emission to health impact showing Exposure as the interface between the human body and the environment, and presenting the roles of Exposure (Dose) Assessment, and Exposure (Dose)/Response Assessment, in Risk Assessment.



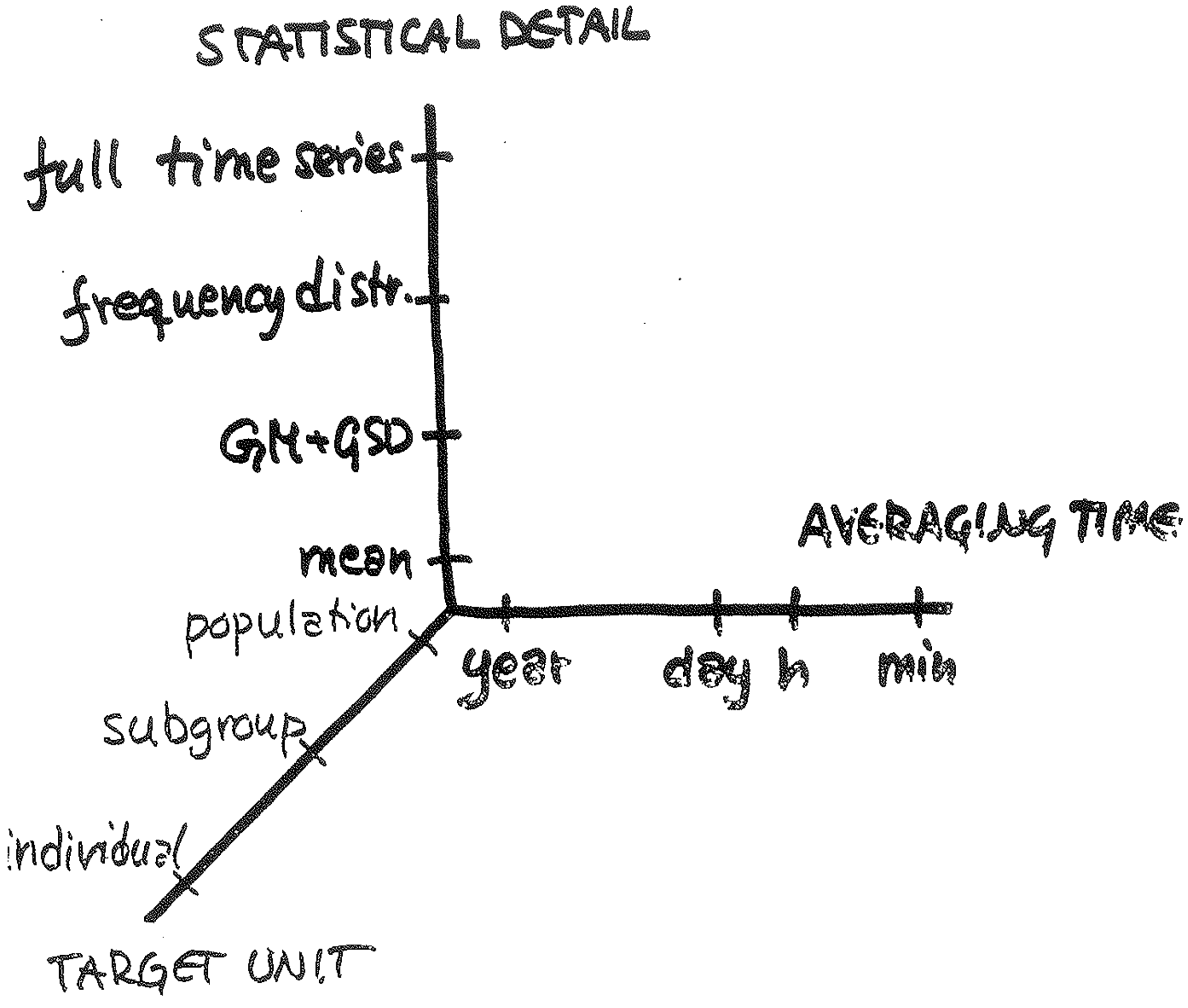
Goals

EXPOLIS

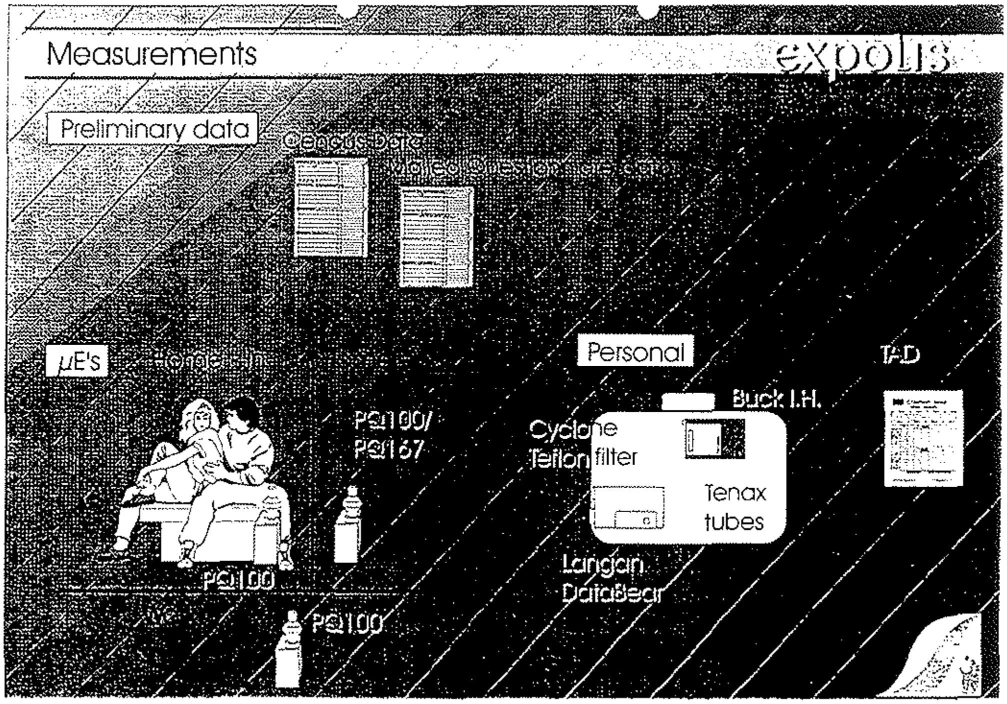
1. To measure the population distributions of exposures to the major air pollutants

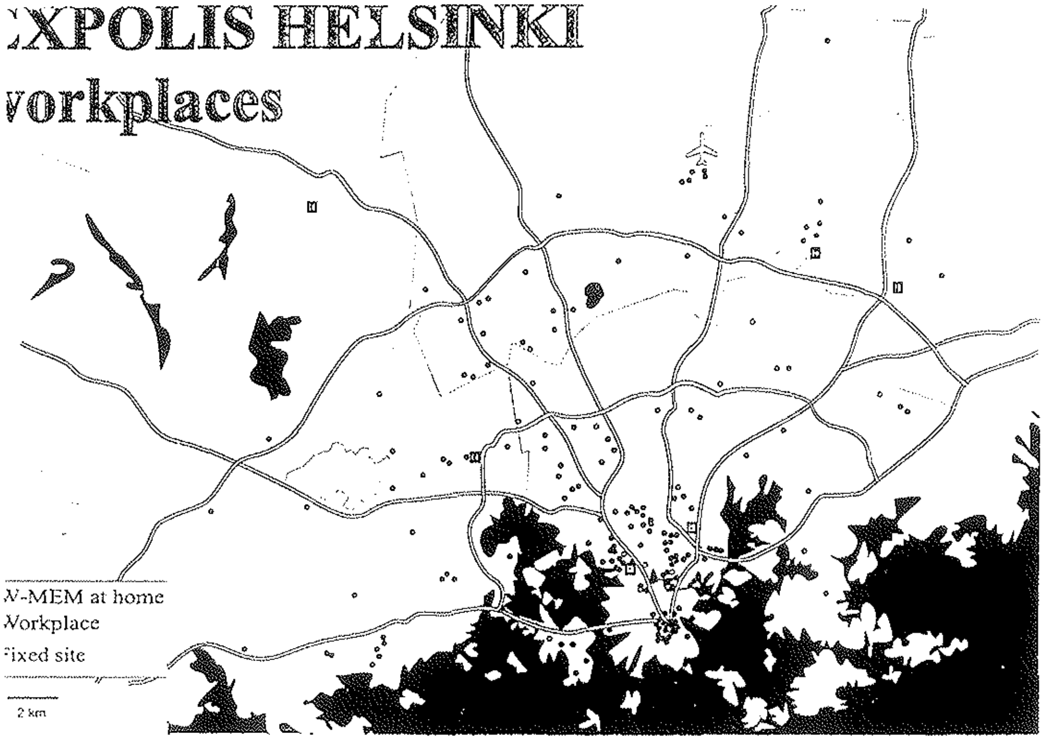
2. To determine the [illegible]

3. [illegible]



Dimensions of exposure


Measurements
expolis
Preliminary data
Census Data
Mailed Questionnaire data
μE's
Home/Work
PQ100/
PQ167
PQ100
PQ100
Personal
Buck I.H.
Cyclone
Teflon filter
Tenax
tubes
Langan
DataBear
TAD


XPOLIS HELSINKI
vorkplaces
V-MEM at home
Vorkplace
ixed site
2 km

PM Exposure Results

- EXPOLIS $PM_{2.5}$ (Helsinki)
  - outdoor vs fixed correlation 91 %
  - indoor vs fixed 22 %
  - PEM night vs fixed 15 %
  - PEM night vs outdoor 35 %
  - indoor vs outdoor 36 %

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PM Exposure Results

- EXPOLIS $PM_{2.5}$ concentration ratios (Helsinki)
  - outdoor / fixed 0.98
  - indoor/ fixed 1.3
  - indoor/ outdoor 1.2
  - personal night/ fixed 1.5

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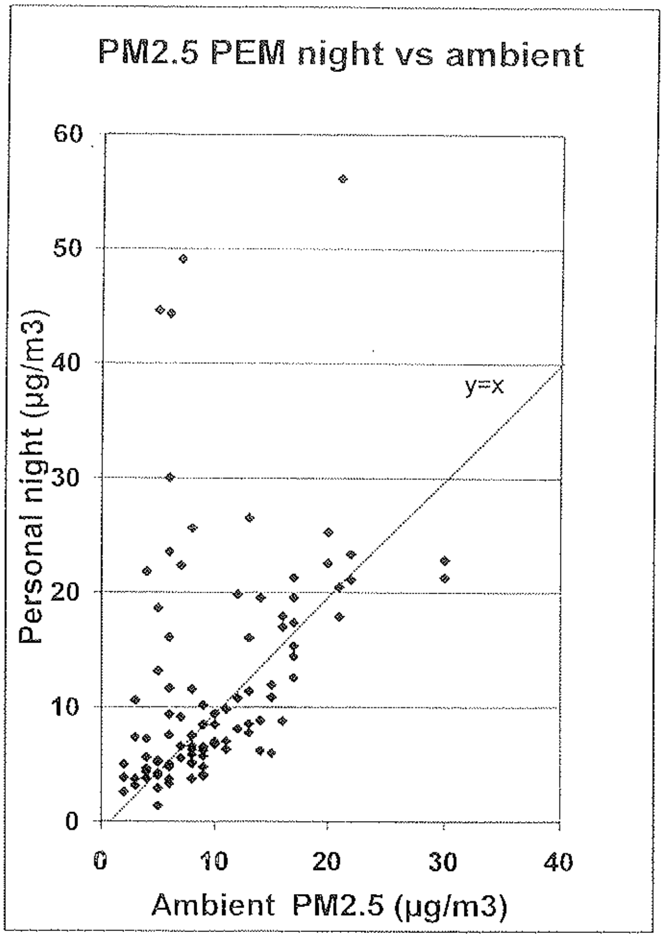

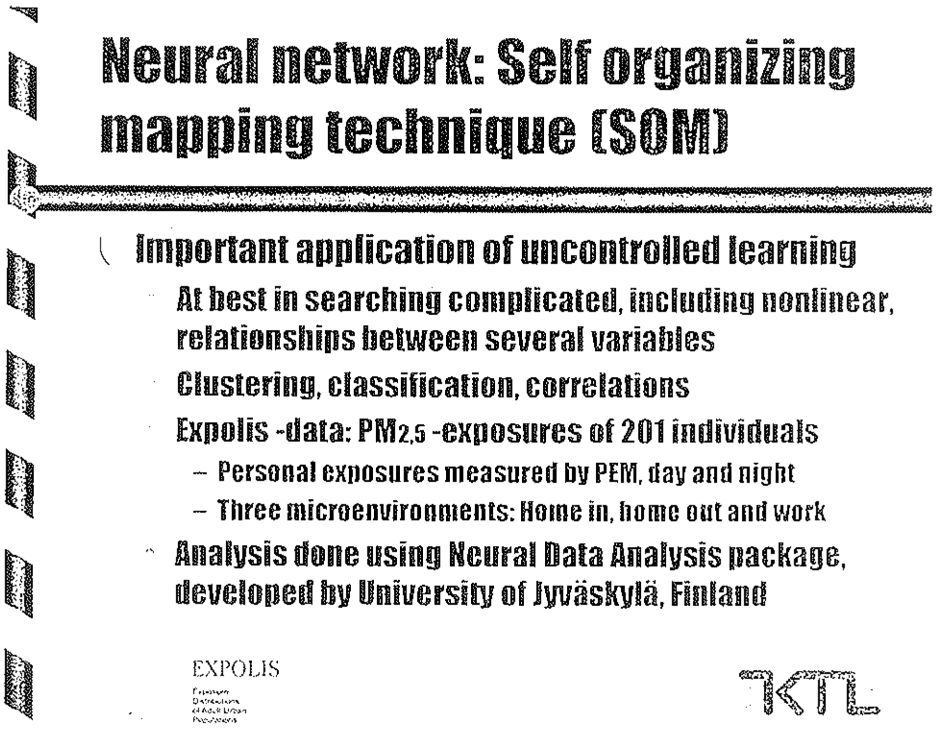

Neural network: Self organizing mapping technique (SOM)

- Important application of uncontrolled learning
  - At best in searching complicated, including nonlinear, relationships between several variables
  - Clustering, classification, correlations
  - Expolis -data: $PM_{2.5}$ -exposures of 201 individuals
    - Personal exposures measured by PEM, day and night
    - Three microenvironments: Home in, home out and work
  - Analysis done using Neural Data Analysis package, developed by University of Jyväskylä, Finland

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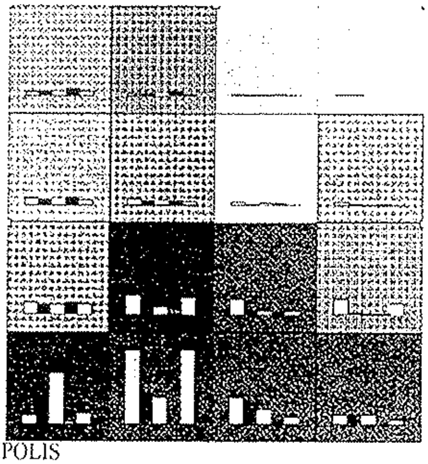
Averages of five $PM_{2.5}$-variables: Personal day, personal night, home in, home out and work. Gray shade indicates number of hits. Lighter shade means increasing number of hits.

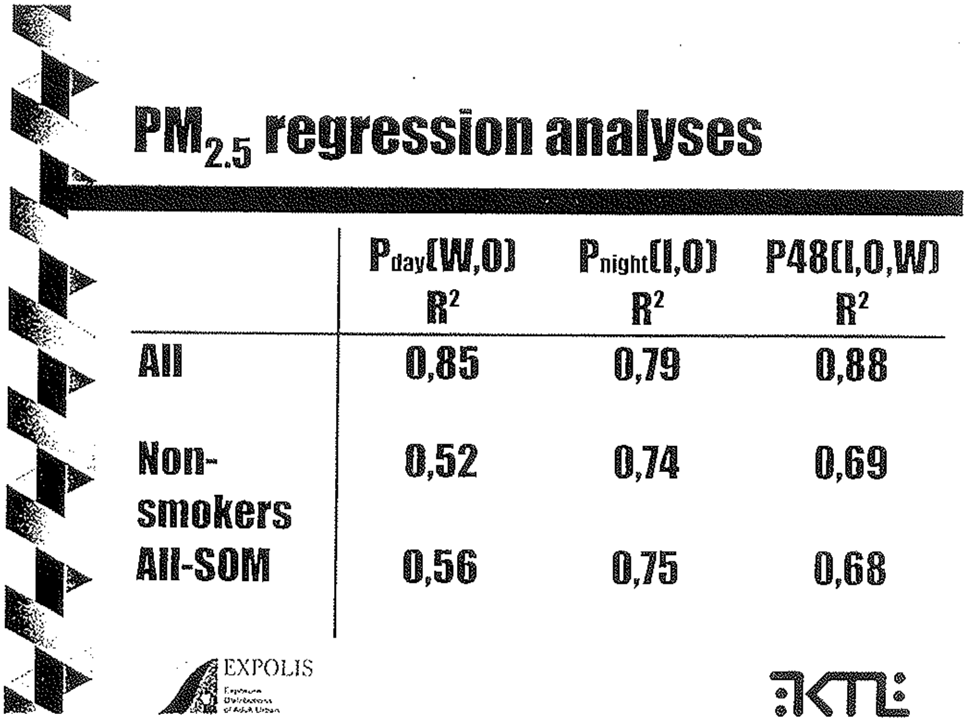


EXPOLIS

Determinants of $PM_{2.5}$ exposures in groups formed by SOM [%].

	"Typical"	"High indoor"	"High day"
Percentage of subjects	94	2	4
Smokers	22	75	100
Home downtown	21	75	25
Workers	19	25	43
Working in traffic	3	0	13
Heavy traffic near home	19	50	25

EXPOLIS



$PM_{2.5}$ regression analyses

	P_{day}(W,O) R^2	P_{night}(I,O) R^2	P48(I,O,W) R^2
All	0,85	0,79	0,88
Non-smokers	0,52	0,74	0,69
All-SOM	0,56	0,75	0,68

EXPOLIS

Outstanding Questions and Gaps for 2003 and Beyond

Chairs: *Daniel Greenbaum,* Health Effects Institute, United States; and *Rolaf van Leeuwen,* World Health Organization, Bilthoven, The Netherlands

NATIONAL ACADEMY OF SCIENCES (US) PM RESEARCH RECOMMENDATIONS

Jonathan M. Samet, M.D., M.D.

Chair, Committee on Research Priorities for Airborne Particulate Matter
And
Professor and Chair, Department of Epidemiology
School of Hygiene and Public Health
Johns Hopkins University
615 N. Wolfe St., Suite 6041
Baltimore, M.D. 21205, U.S.A.

Following the extensive discussion concerning the 1997 promulgation of new National Ambient Air Quality Standards (NAAQS) for particulate matter in the United States, the Congress directed the administrator of the Environmental Protection Agency to arrange for an independent study by the National Research Council on the most important research priorities relevant o setting particulate matter standards. A multidisciplinary committee was convened in early 1998 and its first report, *Research Priorities for Airborne Particulate Matter. I. Immediate Priorities and a Long-Range Research Portfolio*, was published in March, 1998. The first report, in response to the Congressional charge, offered the most-critical research needs in relation to key policy-related scientific uncertainties, and described the long-term timing and estimated costs of the research in an integrated research "portfolio". Recognizing that a long-term commitment to research funding would be needed, the committee set out a 13-year agenda with a mix of components blended over time in a phased sequence. The committee used a toxicological paradigm that begins with sources of particulate matter and ends with human health responses. Ten key research recommendations were based around this paradigm. Over its five-year existence, the committee is to produce three more reports. The second, to be released in early 1999, sets out the committee's strategy for monitoring research progress. Later reports will evaluate progress. The committee's first report has already served as the basis for reorienting the research agenda of the Environmental Protection Agency. The committee process of the National Research Council has proved to be an effective approach for developing a long-term policy relevant research agenda.

Research Priorities for Airborne Particulate Matter

U.S. National Research Council

Jonathan M. Samet, M.D., M.S.

Committee Chair

Committee Expertise

- Epidemiology and biostatistics
- Toxicology
- Exposure assessment
- Atmospheric chemistry
- Risk assessment
- Public health policy

Committee Members

- Jonathan Samet
- Glenn Cass
- Judith Chow
- Robert Forster
- Daniel Greenbaum
- Maureen Henderson
- Philip Hopke
- Petros Koutrakis
- Daniel Krewski
- Paul Lioy
- Joe Mauderly
- Roger McClellan
- Gunter Oberdorster
- Rebecca Parkin
- Joyce Penner
- Richard Schlesinger
- Frank Speizer
- Mark Utell
- Warren White
- Ronald Wyzga

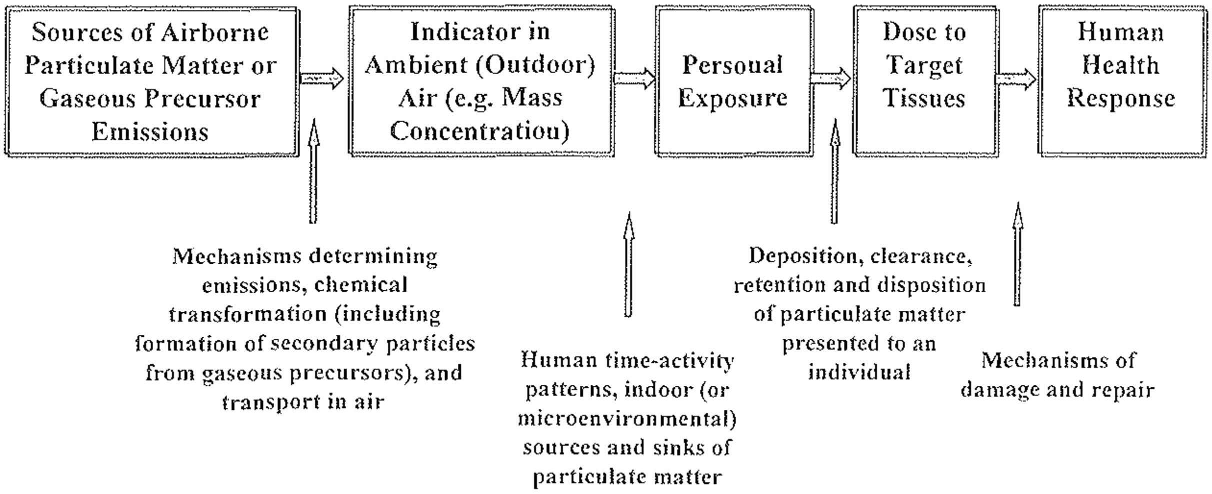
Committee Charge

- Five-year charge, 1998-2002
- Four reports covering:
  - Short- and long-term research needs
  - Research monitoring strategy
  - Progress towards meeting the research agenda

Committee Approach

- Adopt source-to-response framework
  - use framework to identify key uncertainties
- Establish criteria for assigning priorities
  - Scientific value, decisionmaking value, feasibility and timing,
- Develop integrated research "portfolio"
  - 10 component recommendations
- Estimate total and annual costs for each recommendation

Source-to-Response Framework



Sources of Airborne Particulate Matter or Gaseous Precursor Emissions → Indicator in Ambient (Outdoor) Air (e.g. Mass Concentration) → Personal Exposure → Dose to Target Tissues → Human Health Response

Mechanisms determining emissions, chemical transformation (including formation of secondary particles from gaseous precursors), and transport in air

Human time-activity patterns, indoor (or microenvironmental) sources and sinks of particulate matter

Deposition, clearance, retention and disposition of particulate matter presented to an individual

Mechanisms of damage and repair

Source-to-Response Framework

Source → Concentration (or other indicator)

- Contribution of various emission sources to ambient and indoor particulate-matter concentrations
- Relative contribution of various sources to the most toxic components of particulate matter

Source-to-Response Framework *(con't)*

Concentration (indicator) → Exposure

- Relationship between ambient (outdoor) particulate matter and the composition of particles to which individuals are exposed
- Contribution of ambient particulate matter to total personal exposure for:
  - Susceptible sub-populations
  - General population
- Variation in relationship of ambient particulate-matter concentrations to human exposure by place
- Variation in contribution of ambient particulate matter to total human exposure over time
- Covariance of particulate-matter exposures with exposures to other pollutants
- Relationship between outdoor ambient and personal exposures for particulate matter and co-pollutants

Source-to-Response Framework *(con't)*

Exposure → Dose

- Relationship between inhaled concentration and dose of particulate matter and constituents at the tissue level in susceptible subjects
  - Asthma
  - Chronic Obstructive Pulmonary Disease (COPD)
  - Heart Disease
  - Age: infants and elderly
  - Others

Dose → Response

- Mechanisms linking and mortality to particulate-matter dose to or via the lungs
  - Inflammation
  - Host Defenses
  - Neural Mechanisms

Criteria for Assigning Priorities

- Scientific value
  - contribution to knowledge
  - innovation
- Decisionmaking value
  - reduction of uncertainty
  - relevance to risk assessment
- Feasibility and timing
  - operational, technical, and financial feasibility

The 10 Research Recommendations

1. Outdoor measures vs. actual human exposures
2. Exposures of susceptible subpopulations to toxic particulate-matter components
3. Source-receptor measurement tools
4. Application of methods and models
5. Assess hazardous particulate-matter components

The 10 Research Recommendations

6. Dosimetry: Deposition and fate of particles in the respiratory tract
7. Combined effects of particulate matter and gaseous copollutants
8. Susceptible subpopulations
9. Mechanisms of injury
10. Analysis and measurement

Additional Recommendations

- Overall coordination
- Periodic reassessments
- Intramural and extramural talent
- Sustaining adequate research support

The Committee's Second Report

- Reassessment of priorities
- Additional source-receptor model development
  - key for implementation
- Strategy for monitoring research progress
  - need for research inventory

13-Year Costs

Fiscal Year	Estimated Cost ($ million, rounded)
1998	40
1999	46
2000	51
2001	57
2002	55
2003	46
2004	31
2005	31
2006	19
2007	19
2008	19
2009	15
2010	15

PETER BRUCKMANN

Landesumweltamt, Germany

Setting up a monitoring network in Europe: Anticipating Key needs for PM characterization and source apportionment

Abstract

Peter Bruckmann, Landesumweltamt NRW, D-45133 Essen

The most important task for monitoring particulates in ambient air is, from a regulatory point of view, the compliance checking of limit values. The EU Framework directive on ambient air quality as well as the first daughter directive contain important requirements for monitoring networks, which are outlined.

The PM10 burden throughout the territory of the member states has to be assessed, and the monitoring has to cover areas representative for the exposure of the general population as well as hot spot situations. The assessment strategy is based on the monitoring of typical micro-environments, for which minimum numbers of fixed measurement sites are prescribed. Requirements are set up for micro-scale and macro-scale siting of stations, for quality management (e.g. minimum accuracy), and for PM2.5 monitoring. Ultimate aim is to designate all zones not in compliance with the PM10 limit values, and to obtain a representative picture of the PM2.5 burden. The use of a combined system of assessment techniques (monitoring, modelling, emission inventories and indicative measurements) is encouraged.

Whereas this zoning is quite practicable on a regional scale, it can rise considerable problems for hot spots, where distinct spatial gradients of PM concentrations may exist, and modelling may be very inaccurate due to ill defined diffusive emissions.

For zones not in compliance with the limit values action plans to get into compliance have to be set up. This leads to the second task for monitoring, source apportionment, which is necessary to adress the most relevant sources. Because of the considerable uncertainties of emission inventories for particles, source apportionment cannot be based on modelling alone. Analysis of the main constituents (inorganic and organic) of the aerosol yields valuable information, which may be interpreted with the aid of fingerprint and tracer techniques. Consequently, source apportionment by means of the analysis of aerosol constituents should be performed in zones not in compliance with the limit values and with ill defined sources for particulates.

- 2 -

Although compliance checking and zoning of the territory in respect to the PM burden is already a crude exposure assessment, a network preliminary designed for compliance monitoring has considerable shortcomings for health effects research. Other parameters (e.g. PM1, number of ultrafine particles) than PM10 and PM2.5 seem to be more health relevant, and the assessment of personal exposure is quite inaccurate by routinely operating networks. The first difficulty could in principle be overcome by a future change from PM10 or PM2.5 to PM1 or particle numbers, and the pros and cons of measurement techniques for these parameters are discussed. It should be borne in mind, however, that monitoring is only one part of an integrated system of air quality management (other important elements being, inter alia, emission measurements, emission inventories and modelling), and that it is of no use of changing only one element, because all parts must fit together. Therefore, a future change to finer particle fractions or particle numbers should only be performed if the better health relevance of these parameters has unanimously been demonstrated and if the whole air quality management system can be changed accordingly. The shortcomings of routine monitoring for health effects research should be overcome by supplementory monitoring, limited in space and time and specifically designed for the assessment of health effects.

Landesumweltamt NRW, D 45023 Essen

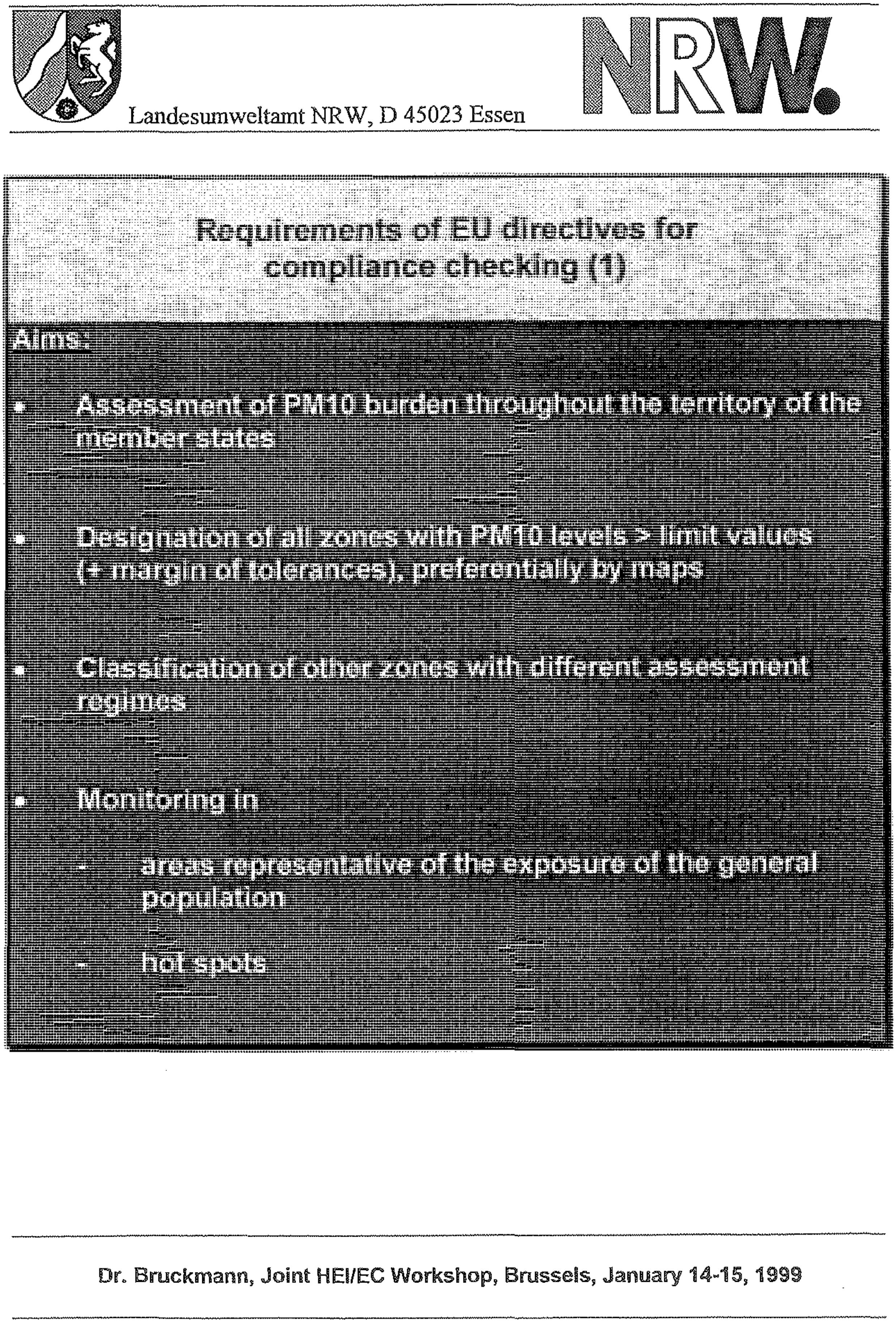
NRW.

Setting up a monitoring network in Europe: Anticipating Key needs for PM characterization and source apportionment

P. Bruckmann,
D-45133 Essen, FRG

Why do we monitor?

- To check compliance with EU limit values
- To improve data base for action plans (source apportionment)
- To assess the exposure for health effects research



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Requirements of EU directives for compliance checking (1)

Aims:

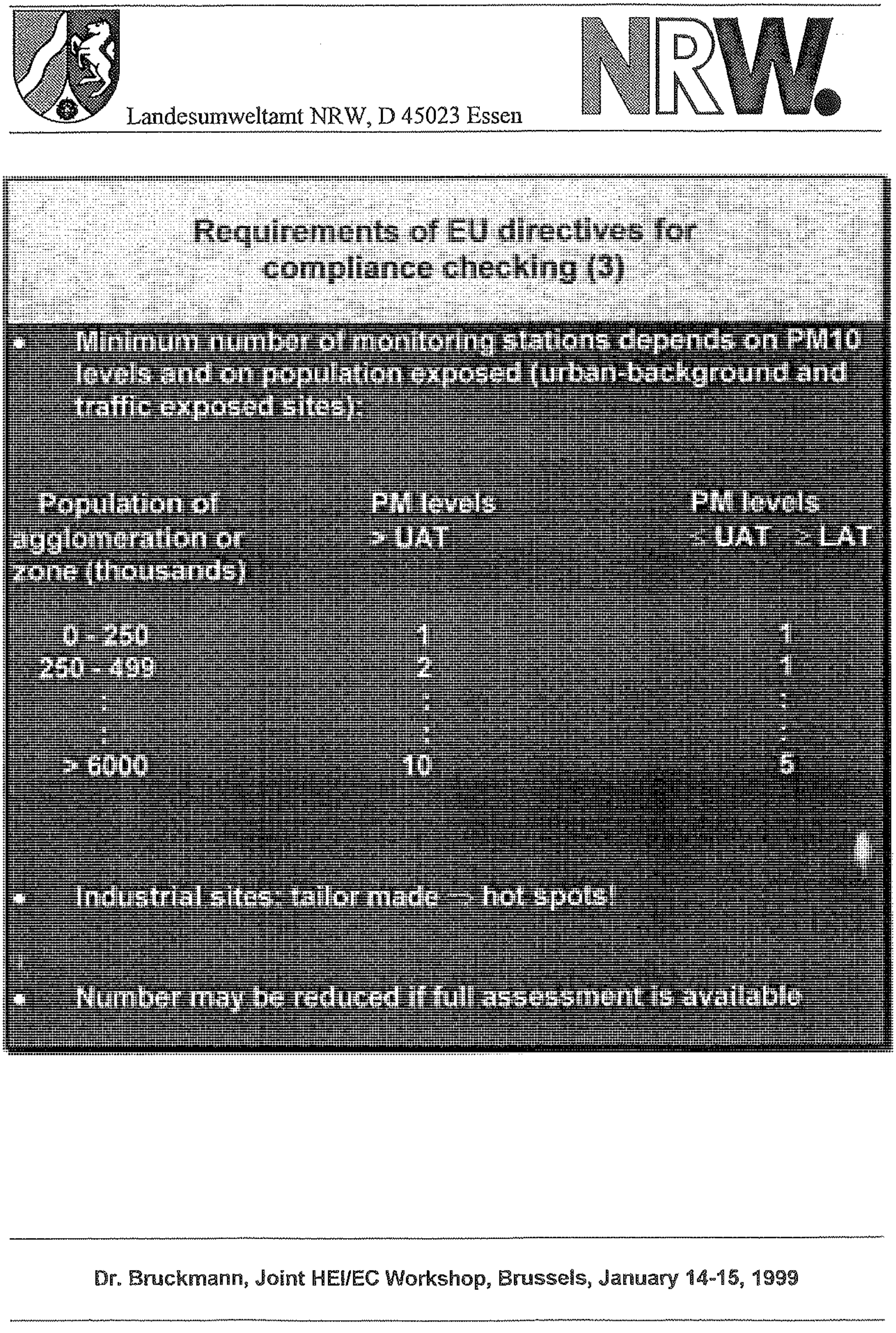
- Assessment of PM10 burden throughout the territory of the member states
- Designation of all zones with PM10 levels > limit values (+ margin of tolerances), preferentially by maps
- Classification of other zones with different assessment regimes
- Monitoring in
  - areas representative of the exposure of the general population
  - hot spots

Dr. Bruckmann, Joint HEI/EC Workshop, Brussels, January 14-15, 1999

Requirements of EU directives for compliance checking (2)

Assessment strategy:

- Monitoring of micro-environments
  - urban-background (representative for several km^2)
  - traffic-oriented (representative for at least 200 m^2)
  - vicinity of industrial sources
- Exposure oriented
  - exposure time must correspond to averaging time of LV
- Assessment preferentially by an integrated system of monitoring, emission inventories, diffusion modelling



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Requirements of EU directives for compliance checking (3)

- Minimum number of monitoring stations depends on PM10 levels and on population exposed (urban-background and traffic exposed sites):

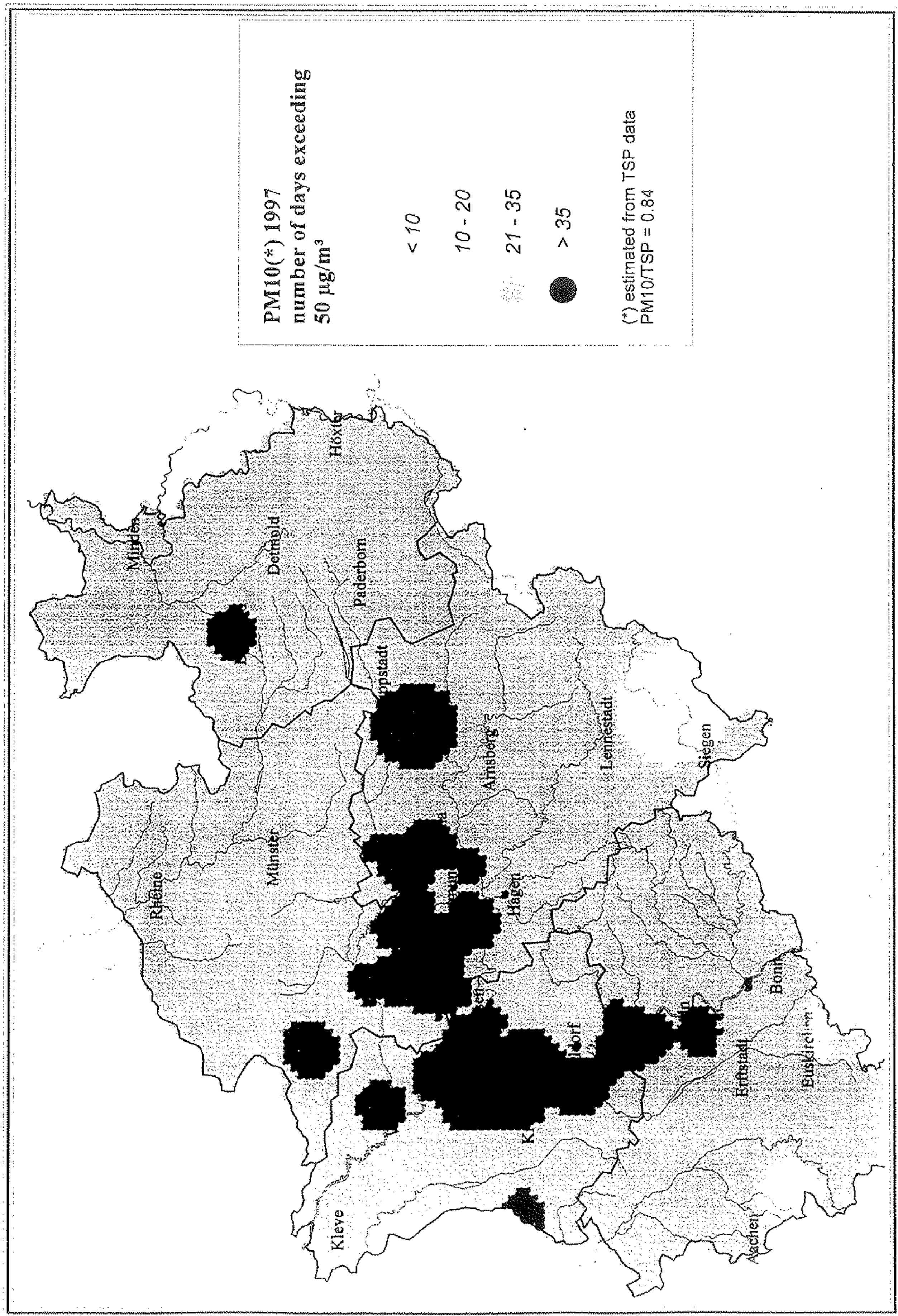
Population of agglomeration or zone (thousands)	PM levels > UAT	PM levels ≤ UAT, ≥ LAT
0 - 250	1	1
250 - 499	2	1
:	:	:
> 6000	10	5

- Industrial sites: tailor made ⇒ hot spots!
- Number may be reduced if full assessment is available

Dr. Bruckmann, Joint HEI/EC Workshop, Brussels, January 14-15, 1999

Requirements of EU directives for compliance checking (4)

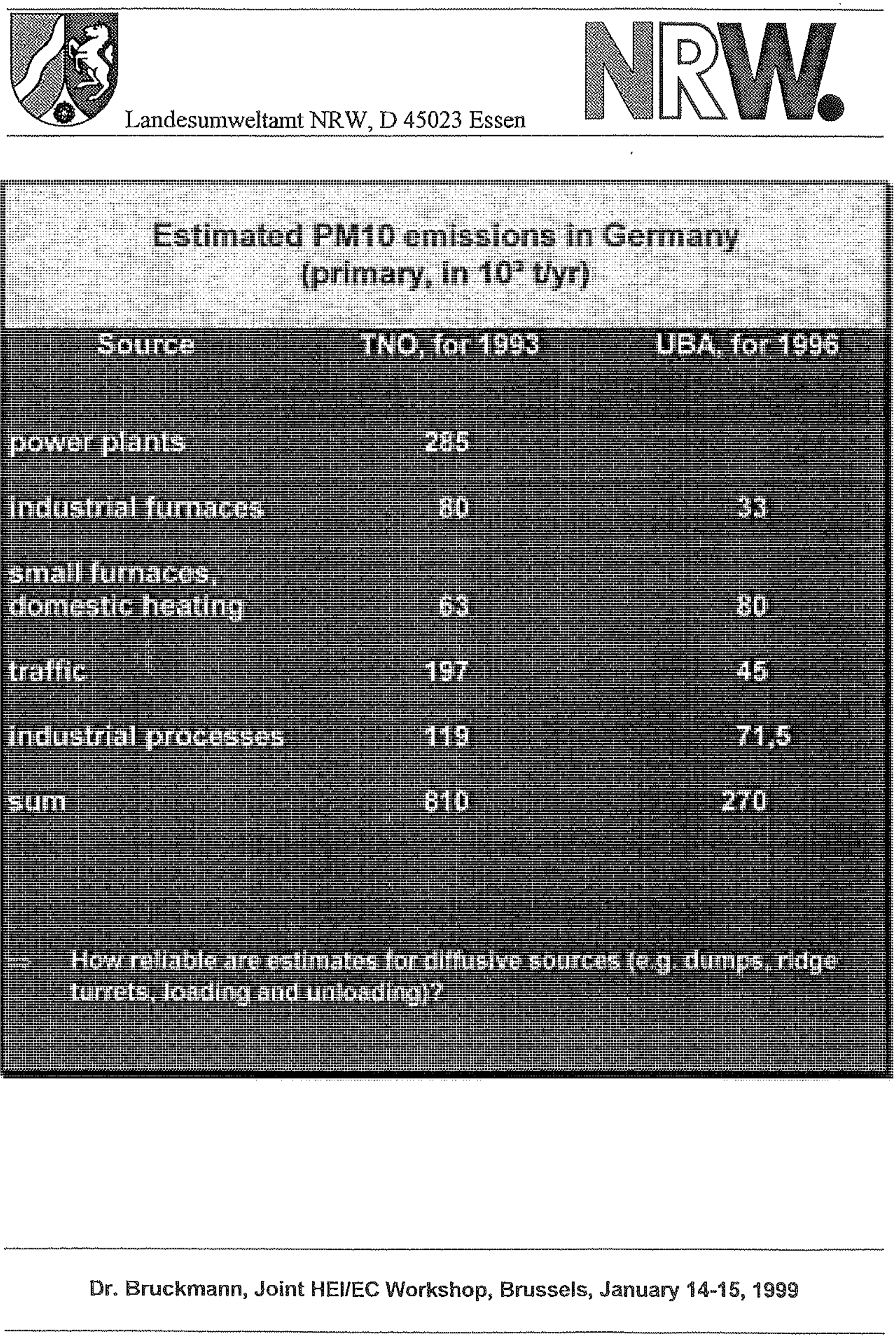
- Requirements for micro-scale siting (annex VI)
- Reference methods:
  - PM10: pr EN 12341
  - PM2.5: CEN WG
- Data quality objectives:
  - accuracy (bias + 2s) 25 %
  - minimum data capture 90 %
- Requirements for PM2.5 monitoring:
  - data must be representative of PM2.5 concentrations in member states
  - collocation with PM10 monitors, if possible
- Monitoring network fully operating in 2001
- Preliminary assessment in 2000


PM10(*) 1997
number of days exceeding
50 µg/m³
< 10
10 - 20
21 - 35
> 35
(*) estimated from TSP data
PM10/TSP = 0.84
Minden
Detmold
Höxter
Paderborn
ppstadt
Arnsberg
Lennestadt
Siegen
Rheine
Münster
Hagen
Bonn
Erftstadt
Kleve
Aachen

Source Apportionment

Necessary in zones not in compliance:

- Action plans heart of the framework directive!
  - identification of main emission sources (problem: diffusive sources!)
  - reduction measures must efficiently address relevant sources
- Considerable uncertainties of emission inventories
- Role of secondary particles
- Share of natural sources and road sanding



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Estimated PM10 emissions in Germany (primary, in 10^3 t/yr)

Source	TNO, for 1993	UBA, for 1996
power plants	285	
industrial furnaces	80	33
small furnaces, domestic heating	63	80
traffic	197	45
industrial processes	119	71,5
sum	810	270

⇒ How reliable are estimates for diffusive sources (e.g. dumps, ridge turrets, loading and unloading)?

Dr. Bruckmann, Joint HEI/EC Workshop, Brussels, January 14-15, 1999

Shortcomings of a network designed for compliance monitoring for health effects research

- Other parameters not routinely monitored (PM1 mass concentration, number of fine and ultrafine particles) seem to be more health relevant

- Assessment of personal exposure quite inaccurate:
  - indoor/outdoor ratios and activity patterns (e.g. commuters)
  - diurnal cycles not reflected by daily means (⇒ weighted daily means?)

⇒ Health effects research should be combined with specifically designed monitoring (limited in space and time) to supplement routine monitoring

⇒ Routine monitoring should only change to PM1 if

- greater health relevance has clearly been demonstrated
- the whole system of air quality management can be changed accordingly

PM1 versus PM2.5 and PM10 (1)

- PM1 seems to be more health relevant
- PM1 less influenced by coarse mode particles (erosion, resuspension)
- PM1 mass concentrations:
  filtration samplers with PM1 inlet ≈ 20.000 ECU

advantages:

- easy handling
- cost efficient
- possibility of chemical analysis
- implementation of standards fits into air quality management system

disadvantages:

- low mass concentrations ⇒ relatively high error in mass determination
- PM1 number concentrations perhaps more health relevant

PM1 versus PM2.5 and PM10 (2)

- PM1 number concentrations:

- Condensation nucleus counter (CNC) with PM1 inlet:

  - range 20 nm - 1 µm: ≈ 35.000 ECU
  - range 3 nm - 1 µm ≈ 50.000 ECU

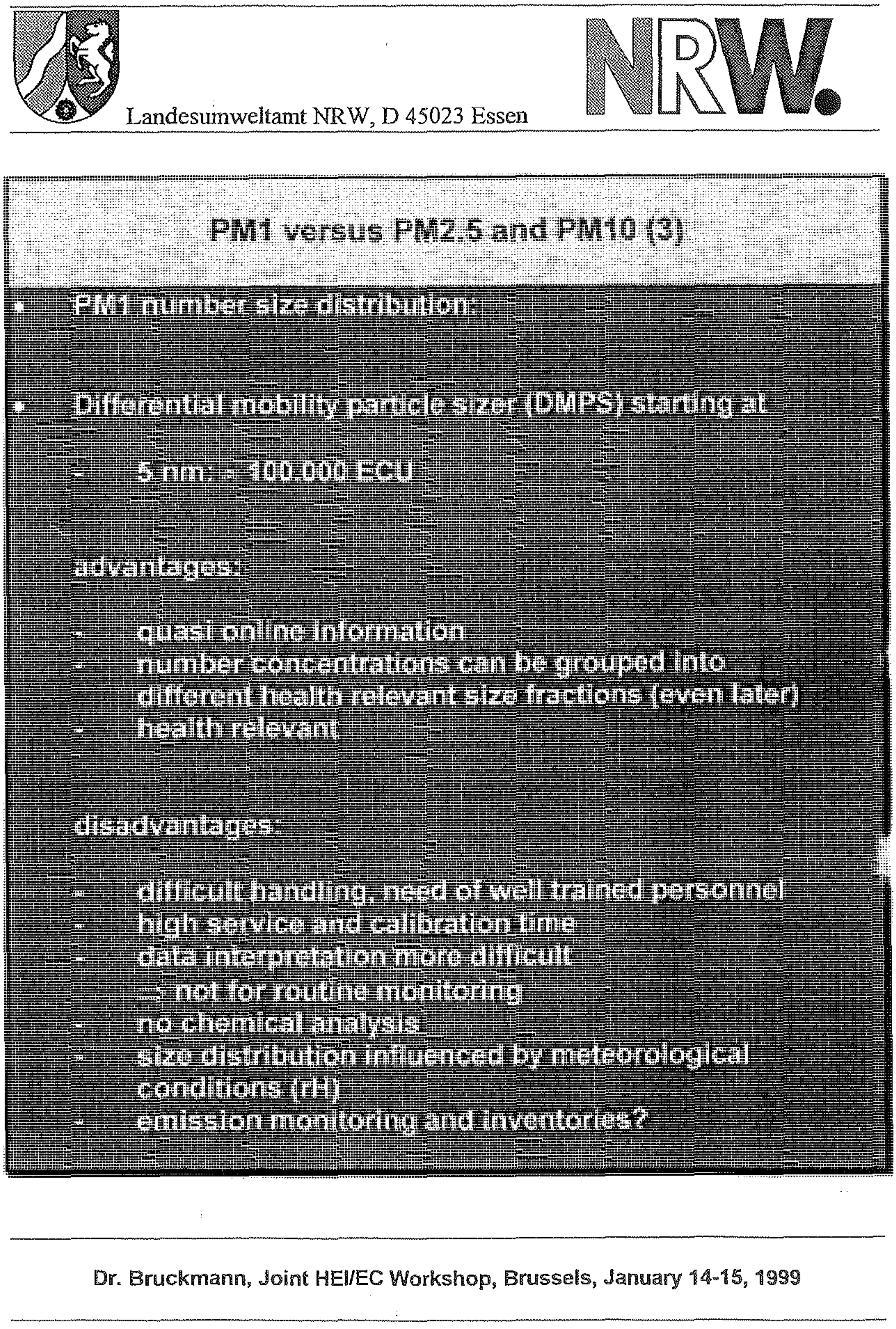
advantages:

- health relevant
- routine monitoring may be possible

disadvantages:

- bias to number of ultra fine particles
- no chemical analysis
- emission monitoring and inventories?

Caveat: ambient air quality monitoring and standard setting are parts of an integrated system of air quality management. All parts must fit together!



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PM1 versus PM2.5 and PM10 (3)

- PM1 number size distribution:
- Differential mobility particle sizer (DMPS) starting at
  - 5 nm: ~ 100.000 ECU

advantages:

- quasi online information
- number concentrations can be grouped into different health relevant size fractions (even later)
- health relevant

disadvantages:

- difficult handling, need of well trained personnel
- high service and calibration time
- data interpretation more difficult
  ⇒ not for routine monitoring
- no chemical analysis
- size distribution influenced by meteorological conditions (rH)
- emission monitoring and inventories?

Dr. Bruckmann, Joint HEI/EC Workshop, Brussels, January 14-15, 1999

Landesumweltamt NRW, D 45023 Essen NRW.

Points for further discussion:

- How relevant are small hot spots (= several hundred people exposed) for compliance checking?
- How do we combine monitoring data with diffusion modelling?
- Do we have sufficient knowledge of diffusive sources, and how can monitoring results help to improve them?
- Are there new tools for source apportionment?
- How do we adress secondary particles (⇒ acidification strategy, emission ceilings?)?
- Shall we switch to other PM parameters like PM1, number concentrations or number size distributions even for routine monitoring and standard setting?
- How can we improve our characterization of personal exposure?
- How important is indoor exposure?

CHARLOTTE BRAUN-FAHRLÄNDER

University of Basel, Switzerland

The European Science Foundation Programme on Environment and Health (ENHE)

Presentation of PD Dr. med. Charlotte Braun-Fahrländer at the meeting 'The Health Effects of Fine Particles: Key Questions and the 2003 Review', Brussels January 14 and 15, 1999, jointly organised by the Directorate General XI of the European Commission and the Health Effects Institute,.

Before focusing on the research recommendations themselves, I will briefly introduce the **background and scope of this ESF initiative.**

The 1994 Helsinki Declaration on Action for Environment and Health in Europe recognised that further scientific research was required to help policy makers to take effective preventive and remedial action. Although various research programmes in Europe investigate environment and health issues at both a European and national level it was felt that there is still a need for further initiatives, especially to inform decision makers on how best to address issues of public health concern. The Helsinki Declaration recommended that the European Science Foundation (ESF) which is an umbrella organisation of the National Research Organisations work with the World Health Organisation (WHO) and the European Commission (EC) to propose research areas to address this need. As a result an ESF Task Force embarked on a programme of scientific consultations on Environment and Health (ENHE) involving observers from WHO and EC.

The ambition of the ENHE Programme was not to cover all possible areas exhaustively where science driven advice may be of use but rather to focus on a limited number of issues (in line with those priority areas identified in the Helsinki Declaration) where policy advice is urgently needed and where pan-European research can make a difference. Therefore, the research needs depicted were not only focused on basic science questions but also related to the translation of basic science results into policy support and to research needs related to risk management principles.

How the recommendations were arrived at

- For the areas identified by the Steering Committee, workshops have been organised and mini-reviews were prepared by the ESF. Eleven areas of research were identified, Ambient Air Particulates being one of them. These issues were discussed by over 150 scientists from a broad spectrum of disciplines and the results were published by ESF in an integrated document, entitled 'An Environment for Better Health' (http://www.esf.org).
- To focus the proposed research areas further, the integrated document was discussed in depth at an ESF update meeting in June 1998, where a multi-disciplinary group of scientists examined the more than 80 detailed recommendations and drew up a shortlist of 24 priority issues outlined in the document 'Scientists' recommendations: Environment and Health Research for Europe, an ESF position paper' (http://www.esf.org) .
- Subsequently, policy makers, scientists and representatives from NGOs and industry discussed this shortlist at a joint ESF/EC/WHO consensus conference in October 1998. A common position on research recommendations has been developed, using scientific and policy criteria, which will be submitted to the Third Ministerial Conference on Environment and Health to be held in London in June 1999.(www.who.dk/london99/topics01/htm)
- Designing details of a research programme, by taking into account the latest developments in the area of environment and health, will be the next step following a positive decision by the Ministers in June 1999.

Research Recommendations relating to Particulate Air Pollution

The research and policy needs for particulate air pollution in Europe have been identified during an international expert workshop 1996 in Bilthoven, based on a background review paper prepared by a group of European scientist. The over 20 main recommendations which came out of the Bilthoven Workshop have subsequently been reduced to a shortlist of 8 main recommendations which will be cribedin more detail.

In addition to scientific issues, some overarching priorities were noted as a limiting factor, such as the lack of opportunity for European researchers to have interdisciplinary interaction on particulate air pollution and the lack of a Europe-wide monitoring network for particulate air pollution. Te formulation of possible working mechanisms of observed health effects related to PM exposure profiles was identified as an overarching priority in the field of particulate air pollution.

Research recommendations relating to release assessment:

1. Development of appropriate source apportionment of PM in indoor and outdoor air to enable the linking of exposure to the sources of PM. This includes research on particle sources as well as on dispersion of particles.

Research recommendations relating to exposure assessment

2. Characterisation of European air quality according to agreed classification criteria with respect to different measures of particles and their correlation with gaseous pollutants.
   Characterisation of personal exposure to PM.
   This may be implemented by a series of mechanisms:

- Interdisciplinary expert consultation to develop criteria
- Monitoring size, mass, number and surface, chemical composition, spatial and temporal distribution of PM indicators (PM10, PM2.5, PM1 and BS)
- Identification of areas with similar and contrasting PM profiles in Europe for the design of future studies
- Compilation of databases on PM concentrations in different microenvironments
- Determination of the relation between personal exposure and ambient concentrations at fixed-site monitoring stations
- Measurement of personal exposure in subgroups of the population

Consequence (health effect) assessment

3. Application of toxicological and clinical research tools to study acute and chronic respiratory and cardiovascular responses to inhaled ambient particles.

4. Epidemiological studies on effects of long term exposure to respirable particulate matter including prospective studies with precise assessment of exposure and health outcomes as well as retrospective studies with estimation of past exposures.

Risk characterisation and risk management

5. Formulation of a set of (policy) scenarios for PM and its public health impact for public evaluation
   and the
6. Formulation of a meaningful set of health impact indicators to be used for the description of the public health impact of PM is required. This set should ideally

also be applied to describe, compare and prioritise other environment and health problems.

7. Evaluate efficacy of previous and current regulatory approaches in terms of public health gain.
8. Evaluate risk management in different economic growth scenarios for cost-benefit and public health gain.

These 8 research recommendations were the result of the science driven discussions within the ESF process. The final ESF position paper (short-list) condensed them to the following three recommended research issues:

1. Conduct epidemiological studies of the effect of long term exposures to respirable particulate matter. These should include prospective studies with precise assessments of exposures and health outcomes; as well as retrospective studies with estimates of past exposures.

2. Apply toxicological and clinical research tools to study acute and chronic cardiovascular and respiratory responses to inhaled ambient particles.

3. Identify the sources of defined groups of respirable particles, in indoor and external environments, to enable the linking of exposure to particulate matter sources.

For the implementation of these proposal a joint effort of international and national activities as well as a pan-European integration and co-ordination of Environment and Health research is needed. The creation of the EC, ESF and WHO liaison group during the ESF process was an important first step in co-ordinating the discussion on research priorities between EC and national research institutions.

Future European planning efforts in the area of particulate air pollution and more general in environment and health research will greatly profit from the creation of an EC, ESF, WHO-EURO platform as it has been proposed to the Ministers of Environment and Health. The next step planned by ESF is the designing of the details of a research programme, taking into account the latest developments in the area of environment and health. It is conceivable that this programme will be more specific with respect to the research needs for fine particulate air pollution health effects.

The European Science Foundation Programme on Environment and Health (ENHE)

PD Dr. med. Charlotte Braun-Fahrländer

Head of the Department Environment and Health
Institute of Social and Preventive Medicine
University of Basel

Background and Scope

- **1994 Helsinki Declaration on Action for Environment and Health in Europe**
- **Scientific research needed to take effective preventive and remedial action: ESF Task Force together with WHO and EC**
- **Criteria for research areas depicted:**
  - **in line with Helsinki priorities**
  - **European scientific added value**
  - **policy relevance towards sustainability**

ESF Process

- **Scientific consultations on environement and health**
- **Workshops and Mini-reviews relating to 11 research areas**
- **Over 80 research recommendations published by ESF in 'Integrated document' (www.esf.org)**

Identified Areas of Research

1. **Climate change, stratospheric ozone depletion and human health**
2. **Social variations in health expectancy in Europe**
3. **Environmental effects on cognitive function**
4. **Cognitive functions as mediators of environmental effects on health**
5. **Children and accidents**
6. **Ambient air particulates**
7. **Indoor air quality and health**
8. **Water quality and drinking water**
9. **Endocrine disrupters**
10. **Human health effects of immunotoxic agents in the environment**
11. **Chemical risk assessment**

ESF Process

- **Science update meeting (June 1998)**
  Short-list of 24 recommendations published as ESF position paper (www.esf.org)

- **ESF / EC / WHO Consensus Conference (October 1998)**
  Background document with research recommendations to be submitted to the Third Ministerial Conference in London 1999 (www.who.dk/london99/topics01/htm)

- **Detailed ESF research programme following positive dicision of Ministers in June 1999**

Research Proposal submitted to London Conference 1999

Overarching Needs

- **Environment and health indicators**
- **Health and environment geographic information system**

Cross-cutting Issues

- **Risk assessment**
- **Environmental contributions to social variations in health**
- **Cognitive function as mediators of environmental effects on health**

Research Proposal submitted to London Conference 1999

Specific research areas

- **Air Quality**
- **Environmental effects on cognitive function**
- **Children and unintentional injuries**
- **Climate change and stratospheric ozone depletion**
- **Water quality and drinking water**

Release Assessment:

1. ***Development of appropriate source apportionment of PM in indoor and outdoor air to enable the linking of exposure to the sources of PM.***
   This includes research on particle sources as well as on dispersion of particles.

Research Recommendations relating to Particulate Air Pollution

Exposure Assessment

2. *Characterisation of European air quality according to agreed classification criteria with respect to different measures of particles and their correlation with gaseous pollutants.*
Characterisation of personal exposure to PM.

Exposure assessment

This may be implemented by a series of mechanisms:

- **Interdisciplinary expert consultation to develop criteria**
- **Monitoring size, mass, number and surface, chemical**
- **composition, spatial and temporal distribution of PM indicators (PM10, PM2.5, PM1 and BS)**
- **Identification of areas with similar and contrasting PM profiles in Europe for the design of future studies**
- **Compilation of databases on PM concentrations in different microenvironments**
- **Determination of the relation between personal exposure and ambient concentrations at fixed-site monitoring stations**
- **Measurement of personal exposure in subgroups of the population**

Consequence (health effect) assessment

3. *Application of toxicological and clinical research tools to study acute and chronic respiratory and cardiovascular responses to inhaled ambient particles.*

4. *Epidemiological studies on effects of long term exposure to respirable particulate matter including prospective studies with precise assessment of exposure and health outcomes as well as retrospective studies with estimation of past exposures.*

Risk characterisation and risk management

5. *Formulation of a set of (policy) scenarios for PM and its public health impact for public evaluation.*

6. *Formulation of a meaningful set of health impact indicators to be used for the description of the public health impact of PM. This set should ideally also be applied to describe, compare and prioritise other environment and health problems.*

Risk characterisation and risk management

7. *Evaluate efficacy of previous and current regulatory approaches in terms of public health gain.*

8. *Evaluate risk management in different economic growth scenarios for cost-benefit and public health gain.*

ESF Short-list of Research Recommendations relating to Particulate Air Pollution

1. **Conduct epidemiological studies of the effect of long term exposures to respirable particulate matter. These should include prospective studies with precise assessments of exposures and health outcomes; as well as retrospective studies with estimates of past exposures.**

2. **Apply toxicological and clinical research tools to study acute and chronic cardiovascular and respiratory responses to inhaled ambient particles.**

3. **Identify the sources of defined groups of respirable particles, in indoor and external environments, to enable the linking of exposure to particulate matter sources.**

KIRSI HAAVISTO

European Commission, DG XII

At the workshop, Kirsi Haavisto of DG XII presented preliminary information on the Fifth Framework Programme. Below is the more final information on this Programme.

I. The Fifth Framework Programme

I.1. Objectives

The Fifth Framework Programme, adopted on 22nd December 1998, defines the Community activities in the field of research, technological development and demonstration (hereafter referred to as "RTD") for the period 1998-2002.

The Fifth Framework Programme differs from its predecessors. It has been conceived to help **solve problems** and to respond to major **socio-economic challenges** facing the European Union. It focuses on a limited number of objectives and areas combining technological, industrial, economic, social and cultural aspects.

Priorities have been chosen according to three basic principles which will apply for all levels: the Framework Programme as a whole, the Specific Programmes implementing it and the RTD activities covered by those programmes.

- ***European "value added" and the subsidiarity principle***, *for example, to reach a critical mass or contribute to solving problems of a European dimension,*
- ***Social objectives***, *such as quality of life, employment or protection of the environment in order to meet the expectations and concerns of the Union's citizens,*
- ***Economic development and scientific and technological prospects*** *in order to contribute to the harmonious and sustainable development of the European Union as a whole.*

I.2. Structure and contents

The Fifth Framework Programme consists of seven Specific Programmes, of which four are Thematic Programmes and three are Horizontal Programmes.

The Thematic Programmes are:

- **Quality of life and management of living resources**
- **User-friendly information society**
- **Competitive and sustainable growth**
- **Energy[1], environment and sustainable development.**

In line with the provisions set out in the EC Treaty, the widely ranging Horizontal Programmes underpin and complement these Thematic Programmes.

The Horizontal Programmes are:

- **Confirming the international role of Community research**
- **Promotion of innovation and encouragement of participation of small and medium-sized enterprises (SMEs)**
- **Improving human research potential and the socio-economic knowledge base.**

One essential new characteristic of the Fifth Framework Programme is the **integrated, problem-solving approach**. Integration is strengthened at three levels:

- ➢ By the **key action concept** in the Thematic Programmes. Key actions are major innovations of the Fifth Framework Programme. They will enable the many and varied aspects of the economic and social issues to be targeted, by integrating the entire spectrum of activities and disciplines needed to achieve the objectives.
- ➢ By **integration between Horizontal and Thematic Programmes objectives.**

  International co-operation

  Participation by entities of third countries and international organisations will be possible in all Programmes in addition to opportunities for participating in the Horizontal Programme "Confirming the international role of Community research". Conditions for participation, including possible financial arrangements, are specified in section III of this document. Box 1 describes the opportunities for bursaries for young researchers from developing countries.

Innovation and participation of SMEs

Measures encouraging SME participation in RTD activities will be carried out in all Thematic Programmes and the Innovation and SME programme. Details on SME stimulation measures will be found in a special information brochure devoted to them. In addition, each Thematic Programme will interface with the Horizontal Programme "Promotion of innovation and encouragement of SME participation" in order to develop awareness and help technology transfer and use of the results of the Thematic Programme.

Socio-economic and training aspects

Socio-economic research can be funded by both the Thematic Programmes and by the key action on "Improving the socio-economic knowledge base" of the Horizontal Programme "Improving the human research potential and the socio-economic knowledge base". Socio-economic research is present in the Thematic Programmes as an integral part of the technological research activities. Training opportunities for researchers are assured through the Marie Curie system of fellowships that can be implemented by Thematic Programmes as well as by other specific training activities in the Human Potential Programme. The fellowships system is described schematically in Box 2.

- By **integration between Thematic Programmes**. Complementary and synergistic interactions will be ensured in implementing the Programmes.

I.3. Implementation

I.3.1. WORK PROGRAMME

A Work Programme has been drawn up for each Specific Programme, describing the specific activities and the various research areas. The Work Programme will be revised regularly with the assistance of Advisory Groups of independent experts to ensure its continued relevance in the light of evolving needs and developments. Potential proposers should therefore ensure they are consulting the **current** version of the work programme when planning a proposal. The Work Programme appearing at the Specific Programme Web site is always the current version.

The Work Programme includes an indicative timetable or "***roadmap***", which indicates which parts of the Work Programme will be opened, by calls for proposals, and deadline(s) involved. This provides a means of focusing attention on areas or sub-areas, thereby optimising opportunities for launching collaborative projects and establishing thematic networks.

The Commission will manage the Specific Programmes to ensure that links in thematic content between the programmes are exploited in a synergistic way. This may occasionally require joint or synchronised calls for proposals. Where necessary, co-ordination measures such as these will be indicated in the announcement of the calls for proposals, and in the Work Programme.

I.3.2. TYPES OF ACTIONS SUPPORTED

The Community will contribute financially to the RTD[2] activities, carried out under the Specific Programmes implemented within the Fifth Framework Programme. The general rules[3] are as follows:

(a) Shared-cost actions

- **Research and technological development (R&D) projects**[4] – projects obtaining new knowledge intended to develop or improve products, processes or services and/or to meet the needs of Community policies (financial participation: 50 % of total eligible costs[4,5]).
- **Demonstration projects**[4] – projects designed to prove the viability of new technologies offering potential economic advantage but which cannot be commercialised directly (financial participation: 35 % of total eligible costs[5]).
- **Combined R&D and demonstration projects**[4]– projects combining the above elements (financial participation: 35 to 50 % of total eligible costs[4,5]).
- **Support for access to research infrastructures** – actions enhancing access to research infrastructures for Community researchers. Support will cover maximum of 100 % of the eligible costs necessary for the action.

- **"SME Co-operative" research projects**[4] – projects enabling at least three mutually independent SMEs from at least two Member States or one Member State and an Associated State to jointly commission research carried out by a third party (financial participation: 50 % of total eligible project costs[4]).
- **"SME Exploratory" awards** – support of 75 % of total eligible costs[6] for an exploratory phase of a project of up to 12 months (e.g. feasibility studies, validation, partner search).

(b) Training fellowships

Marie Curie fellowships are either fellowships, where individual researchers apply directly to the Commission, or host fellowships, where institutions apply to host a number of researchers (financial participation: maximum of 100 % of the additional eligible costs necessary for the action[7]).

(c) Research training networks and thematic networks

Training networks for promoting training-through-research especially of researchers at pre-doctoral and at post-doctoral level - *and thematic networks* for bringing together e.g. manufacturers, users, universities, research centres around a given S&T objective. Support will cover maximum 100% of eligible costs necessary for setting up and maintaining such networks.

(d) Concerted actions

Actions co-ordinating RTD projects already in receipt of funding, for example to exchange experiences, to reach a critical mass, to disseminate results etc. (financial participation: maximum of 100 % of the eligible costs necessary for the action). These include co-ordination networks between Community funded projects.

(e) Accompanying measures

Actions contributing to the implementation of a Specific Programme or the preparation of future activities of the programme. They will also seek to prepare for or to support other indirect RTD actions (financial participation: maximum of 100 % of total eligible costs).

I.3.3 CLUSTERS

The cluster is a defined group of RTD projects. Its aim is to guarantee complementarity among projects, to maximise European added value within a given field and to establish a critical mass of resources at the European level.

An integrated approach towards research fields and projects financed is needed to solve complex multidisciplinary problems effectively. The clusters reflect this **problem-solving approach.** Indeed, in a cluster projects are joined together because they complement each other in addressing major objectives in the context of a key action or a generic activity (sometimes even across different key actions or specific programmes). Clusters are expected to optimise scientific networking, management, co-ordination, monitoring, the exchange of information and, on voluntary basis, the exploitation and dissemination activities. The cluster may thus become a natural process to generate European added value, wherever it makes sense, beyond the limited resources of an isolated project.

All types of projects can be assembled and integrated within a cluster, including those funded by different EU RTD activities (key action, generic activity, infrastructure). By the same token, and as part of an overall European approach, relevant activities under other research frameworks (notably EUREKA, COST) could also be taken into account whenever this can reinforce synergy.

I.3.4. GENDER EQUAL OPPORTUNITIES

In line with the Commission's strategic approach of mainstreaming equal opportunities in all Union's policies, particular account is taken in the Fifth Framework Programme of the need to promote the participation of women in the fields of research and technological development.

INVITED COMMENTS

What Are the Priority Research Questions Relative to Public Health and Regulation?

What Can Be Accomplished in Time to Inform the 2003 Review?

FRAZIER GOODWIN
European Federation for Transport and Environment,

T&E How serious is the problem?

- What are actual Exposure levels
- What are the effects of such exposures both in chronic and acute terms
  Helpful here could be the multi centre studies
- Are there sub groups at higher risk ?
  Old, young, heart and lung patients, pregnant mothers

T&E Are we solving the problem

- PM modelling
- what has technical adaptation to standards produced ?
- What is the impact of new technology sources GDI for example

T&E The Size of the problem

- Standardised reference measurement for PM 2.5
- Characterisation of particles in terms of :
  - Where they come from / sources
  - What compoiunds are on / in particles

T&E Air Pollution Problem?

Questions the Research Community should address:

- What is the Size of the problem
- How serious is the problem
- Are we doing enough to solve the problem

MICHAEL SPALLEK

Volkswagen AG, Germany

Fine Particles - Key Questions

Uncertainties on the correlationship between reduced PM mass and PM numbers

- Influence of new engine technologies and fuel qualities ?

Standardized and reliable measurement method is needed

- Definition of ultrafine particles
- Reliable counting systems
- Accurate source relationships
    - Combustion vs. other sources
    - Individual exposure vs. stationary exposures

Fine Particles - Key Questions

Need for plausible biological causes for the observed statistical associations between changes in PM and different health effects

There´s a need for a more holistic view of the PM-associated problems

(MONICA-Project WHO: weather condition/temperature/air pressure and CHD)

What about an „acceptable risk“ for PM effects in comparison to other environmental or „individual“ risks ?

WIM TORDOIR

CONCAWE, Belgium

CONCAWE
SHARED INTERESTS IN PM RESEARCH

IMPROVEMENT OF HAZARD IDENTIFICATION*

THROUGH EPIDEMIOLOGY	BY 2003	AFTER 2003
Mortality/morbidity time series studies		
- evaluate clinical characteristics of cases: excess versus non-excess situation	y	
- reconstruct personal exposures	y	
- evaluate confounding factors at individual level	y	
Cohort studies		
- retrospective	y	
- prospective		y
THROUGH MECHANISTIC TOXICITY STUDIES		
Physical		
size mass number		y
Chemical composition		
primary secondary anorganic/organic		y

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* Current epidemiological studies are only suitable for generating hypotheses, not for quantitative risk assessment

CONCAWE
SHARED INTERESTS IN PM RESEARCH

IMPROVEMENT OF EXPOSURE ASSESSMENTS

	BY 2003	AFTER 2003
• EXPAND DATA BASE ON PERSONAL EXPOSURE (AMBIENT- RURAL/URBAN/INDUSTRIAL VERSUS INDOOR)	y	
• EVALUATE STATIC AREA SAMPLING AS PREDICTIVE FOR PERSONAL EXPOSURE	y	
• DEVELOP MODELS PREDICTING PERSONAL EXPOSURE OF SPECIFIC SUBPOPULATIONS	y	

CONCAWE
SPECIFIC INTERESTS OF OIL INDUSTRY IN PM RESEARCH

		BY 2003	AFTER 2003
HAZARD IDENTIFICATION			
Mechanistic toxicity studies			
PM from	- gasoline vehicles		y
	- diesel vehicles		y
	- heavy fuels from powerplants and ships		y
EXPOSURE			
Personal exposure studies			
PM from	- gasoline vehicles		y
	- diesel vehicles		y
	- heavy fuels from powerplants and ships		y
AIR QUALITY			
• Emission inventories		y	
• Source apportionment		y	
• Modelling:	emissions -> air concentrations -> personal exposure		y

PREREQUISITE:
- proper characterisation
- proper generation

Posters: New PM Research Results from Europe and the USA

Exposure Assessment Studies

Epidemiology Studies

Toxicology Studies

EXPOSURE
ASSESSMENT
STUDIES

CELINE BOUDET
University of Grenoble, France

PM exposure assessment in Grenoble

Zmirou D.[1], Boudet C.[1], Déchenaux J.[2], Personnaz MB.[3]

[1]Grenoble U. Public Health Laboratory
[2]Pollution Prevention Association (APPA)
[3]Grenoble Air Quality Network (ASCOPARG)

Personal/population exposures to air pollutants are often assessed indirectly and ecologically, which may lead to substantial misclassification. Many studies suggest that $PM_{2.5}$ are more noxious than PM_{10}. In Europe, $PM_{2.5}$ are not routinely monitored by the Air Quality Networks. Moreover, $PM_{2.5}$ personal exposure data are scarce. In this context, the EXPOLIS study has been supported by the E.U. It took place in Helsinki, Athens, Basel, Prague, Milan, Bilthoven and Grenoble. It aimed at characterizing exposure distributions of adult urban populations. This poster presents some results from the French EXPOLIS center, and additional local data on particulate exposure.

Non-smoker volunteers (adults) were selected in summer 96 (n=40), in winter 97 (n=40) and in summer 98 (n=20). They carried a personal PM2.5 monitoring case during 48 h. and they filled time-activity diaries as well as questionnaires on their life-environments. Particles were collected on Teflon filters and masses were determined by deionised weighing and reflectometry.

In Grenoble, $PM_{2.5}$ total personal exposure is similar to results from Basel or Athens, with greater average values in winter (36.7, 25.6, 35.6 $\mu g/m^3$ respectively) than in summer (21.9, 19.7, 26.2 $\mu g/m^3$ respectively). Helsinki shows the lowest values (15 $\mu g/m^3$), with no variation between summer and winter. Prague finds much greater values in summer (43.4 $\mu g/m^3$ on average) than in winter (32.4 $\mu g/m^3$). The relationship between the mass and blackness of the particles deposited on filters is dependent upon the particles sources, and thus varies across locations and periods. In spite of the high percent of time spent indoors (about 90 %), time spent outdoors is associated with a great fraction of the total personal exposure (23 % in winter, 35 % in summer). These results under-estimated the contribution of outdoor sources since they do not take into account the fraction of indoor particles that penetrates from outdoors. Cross-sectional analyses of the relationship between personal $PM_{2.5}$ exposure and PM_{10} ambient air levels show poor correlations, as usually described in the literature. However, the field study design in Grenoble (3 phases) allows a longitudinal approach on a restricted number of volunteers (n = 6) with repeated measurements. There are high correlations between the time variations of personal exposures and of ambient air levels (R = 0.78 on average). The Grenoble Air Quality Network measures PM_{10} at a high traffic proximity site and on a urban background site, using a TEOM. Punctual $PM_{2.5}$ measurements (GRIMM G1105) allowed us to calculate some $PM_{2.5}/PM_{10}$ ratios so as to look at the spatial and seasonal variability of the PM ambient air levels. Ratios show some consistencies across seasons and locations (about 0.3). Preliminary findings in Europe show a high variability of $PM_{2.5}$ personal exposures within and between towns. As a result, there is a need for further research in order to describe the determinants of this variability.

MATTI JANTUNEN

National Public Health Institute, Kuopio, Finland

THE ROLE OF MULTICENTER STUDIES IN AIR POLLUTION RESEARCH

THE *EXPOLIS* EXPERIENCE

Matti Jantunen, KTL, Environmental Hygiene, Kuopio, FINLAND

ABSTRACT

EXPOLIS goals

Epidemiological literature of the 1990's has revealed surprisingly large public health impacts associated with present common air pollution levels in North American and European cities. Any causal explanation of the health effects of air pollutants must go through exposure, yet, prior to *EXPOLIS* no large, population based air pollution exposure studies have been conducted in Europe, and consequently no European database of air pollution exposures of urban populations has existed until now. *EXPOLIS* is a European multicentre study for measurement of air pollution exposures of working age urban populations during workdays. The selected urban areas are Athens, Basel, Grenoble, Helsinki, Milan and Prague. The main objectives of *EXPOLIS* are:

* To assess the exposures of European urban populations to major air pollutants.

* To analyse the personal and environmental determinants and interrelationships to these exposures.

* To develop an European database for simulation of air pollution exposures.

These objectives were pursued by measuring the personal exposures, home indoor and outdoor and workplace levels of $PM_{2.5}$, VOCs and CO of approximately 500 subjects representing the adult populations of the selected cities.

Field work

continued from fall of 1996 to winter of 1997-98. To assure comparability of the data from the 6 cities in 6 countries, a strict QA/QC protocol was established, standard operating procedures were prepared for all subject, laboratory and field procedures, and identical sampling equipment, operating procedures, time-microenvironment-activity diaries, questionnaires, database and data entry tools were used in each centre. VOC laboratory analyses were intercalibrated by the European Commission / Joint Research Centre (EC/JRC) Environment Institute in Ispra. Other techniques were intercalibrated between the teams.

Reasons and consequences of multiple centres

The multicentre nature of the *EXPOLIS* study is based on the need to acquire exposure data for modelling purpose from a wide variety of qualitatively and quantitatively different populations, cultures, climate zones and urban centres, bu also on the need to involve the best available expertise from exposure monitoring, modelling and exposure data application points of view.

A multicultural and -lingual study like *EXPOLIS*, where multiple compounds are monitored in multiple microenvironments, needs a great deal of practical everyday problem-solving and other communication to ensure on the one hand a common practice and comparable study results, and on the other hand minimum data losses. The junior researchers were trained at the different phases of the study together in *EXPOLIS*-Workshops in Prague (Apr. 1996), Helsinki (Sept. 1996), Grenoble (Mar. 1997), and Bilthoven (Feb 1998). In each center one team member was assigned to one or more of the following contact groups: *Equipment*, *Database* and *VOCs*. *QA/QC* and *Privacy Protection* responsibilities lay within the principal investigators. Communication occurred mostly via E-mail, but each team member was also assigned a GSM telephone with contact numbers of all other team members to ensure fast access when and where problems were encountered in the field or laboratory.

Session: EXPOLIS, Abstract Number: 263 P

Expolis Database for Exposure Analysis and Simulation with Data from Six European Cities

Exposure to air pollution have been shown to cause substantial health effects in urban populations. The exposures and related factors of European urban populations have been measured in the Expolis project and the results are published as a common CD-ROM database called EADB.

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1. Introduction

Real exposures of individuals determine the health effects of air pollution. While fixed station monitoring gives a rough estimates of the average exposures of the populations, their exposure distribution are usually not known (*Jantunen et al 1998*).

Population based exposure studies, like the Expolis study in Europe, produce detailed exposure descriptions and distributions in the measured population and the measurement time frame. With statistical and modeling tools these results can be generalized to other populations. (*Jantunen et al 1998*). Thus, extensive set of such data would be useful in many research setups concerning population exposures

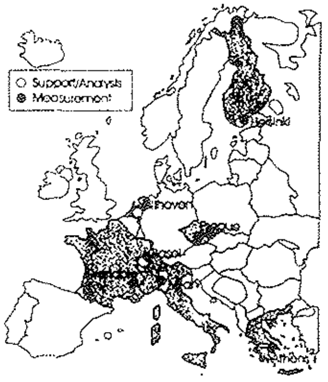

Figure 1. The Expolis Centers. From these six cities the exposure, concentration and other data is stored in the EADB.

Table 1. Population sample sizes in the Expolis Centers.

Center	Primary	Exposure	Diary
Athens	2.000	50	50
Basle	3.000	50	250
Grenoble	-	40	40
Helsinki	2.523	201	240
Milan	3.000	50	250
Pregue	-	50	50
Total	10.523	480	880

Table 2. Summary of measured compounds and sample types.

Measurement	$PM_{2.5}$	VOC 30 compounds	CO	NO_2
Personal	cyclone, filter day/night	tenax day/night	continuous (1min)	Palmes tube 48 h
Home indoors	impactor, filter night	tenax night	-	-"-
Home outdoors	-"-	-"-	-	-"-
Workplace	impactor, filter day	tenax day	-	-"-

2. Materials and methods

The personal air pollution exposures of urban populations have been measured in the Expolis study during 1996-1997 in six European cities (see figure 1).

The study subjects are randomly drawn samples of 25-55 year old inhabitants of each city. Population samples are stratified so that civil register data is gathered for largest number of subjects (*Primary sample*) and questionnaire and diary data for a somewhat smaller sample (*Diary sample*). Smallest number of subjects participated the exposure and concentration measurements (*Exposure sample*) (see table 1).

Measurements included personal (PEM) and microenvironmental (MEM) measurements of PM2.5, 30 VOC compounds, CO and NO2 (see **table 2**). Microenvironmental samples were programmed to run during the time the subject was present, e.g. daytime at workplace.

3. Results

The Expolis measurement results, such as questionnaires, diaries, exposures and microenvironmental concentrations, as well as simultaneously measured ambient air quality and meteorological data are stored in an international MS-Access version 7 (Office 95) database called **EADB**.

The basic structure of the EADB is shown in figure 2. The detailed structure will be published in 1999 and the database will be made available for researchers for specified purposes.

4. Discussion

Data collected in the Expolis study provides distributions of personal exposures and related microenvironmental concentrations in the subject's home and workplace, as well as the time activity patterns of the subjects.

The effects of factors like smoking, cooking and commuting can be statistically analyzed from the database. Observed empirical relations, supplemented with other data from the literature can be used in many in simulation and statistical modeling situations.

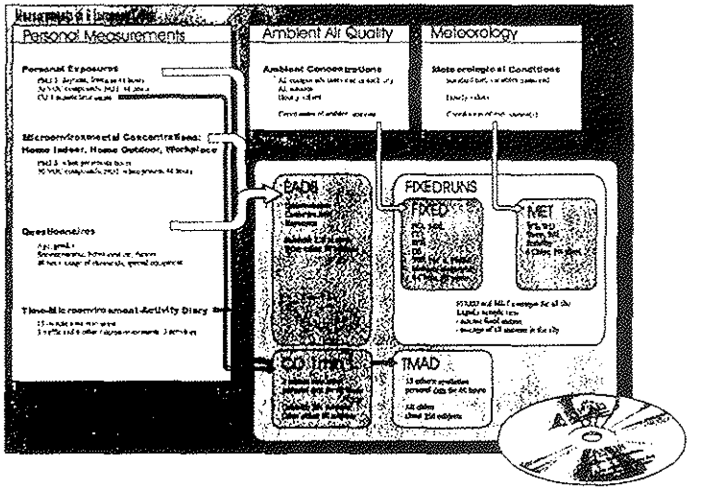

Figure 2. The structure of the Expolis database. The detailed structure of the finished database will be published in 1999 and the data will be made available to research teams for specified analyses.

Thus the EADB, containing comprehensive set of exposure related data, will be very useful in many research setups.

Uses of the EADB

The EADB can readily be used in analyzing the relationships between behavioral, socioeconomic and other factors and the personal exposures. The database is suitable also for comparing the relationships of personal exposures and microenvironmental concentrations to the ambient air quality results from monitoring networks in different cities.

The EADB data, combined with simulation tools, can also be used to assess exposures of urban populations, where no measured exposure data are available.

The EADB can be used in selecting population groups for panels and cohorts in epidemiological studies and to assess sample sizes. It can also be used to evaluate the effect of measurement error on power and on distortion of the exposure - response relationship.

Acknowledgments

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EXPOLIS
Exposure
Distributions
of Adult Urban
Populations

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Comprehensive Integrated Risk Assessment for Risk Management Purposes

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Introduction

Population exposure to ambient particulate matter (PM) is associated with serious risk to public health. It is not known yet whether this association is causal nor what fraction of PM is most powerful in rendering health effects. However, it appears biologically plausible that exposure to PM and/or mixture of PM and gasses is causally related to widespread morbidity and mortality in the entire population. Further, for risk management purposes it is prudent to assume that this relationship is causal. Although, the more costly the abatement measures, the better science has to underpin the benefits not only in improvement of air quality but also in terms of risk reduction. The challenge of PM research lies therefor in diminishing uncertainties. Not only with respect to causality but also for finding cost-effective ways for risk reduction. This callenge calls for a comprehensive integrated risk assessment aimed at, and governed by risk management objectives.

Methods

By combining data of measured PM_{10} levels with exposure-response relationships we arrived at a quantitative risk assessment of PM exposure of the Dutch population. Thereafter we collected information on all European sources (alloted to a number of source categories) of primary PM and inorganic precursor gasses. We then calculated the annual averages of PM_{10}, $PM_{2.5}$, inorganic secondary PM and carbonaceous PM for the rural region, a city, a heavy traffic street and an industrial area in the Netherlands. We have chosen a number of indicators for PM as we do not know the causal factor. By calculating the influence of various abatement scenarios on the relative reduction of the levels of these indicators we may support or refute a "no-regret" policy. By modelling the subsequent reduction of actual population exposure and by combining this increased exposure with exposure-response data, we calculate the accompanying risk reduction. All information on emissions, spatial distribution of sources, modeled air quality, actual population exposure and exposure response data have been put into a model. These various activities amount to some 25 man year/year over 6 years.

Results of the first phase of the PM model

- a large gap exists between calculated and measured PM levels, partly since non-anthropogenic sources have not yet been included.
- the contribution of various source categories to public health risk has been quantified.
- the impact of policy measures, economic scenarios, future trends in emissions (AUTO-OIL II, UN/ECE, NOx-Protocol, etc.), were evaluated.
- inhalation toxicological research on the effect of particle size and composition has been performed as well as toxicological research with concentrated $PM_{2.5}$
- epidemiological research points to the mixture of PM and gasses as important explanatory variables for morbidity and mortality.

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EXPOSURE AND RISK ASSESSMENT FOR FINE AND ULTRAFINE PARTICLES IN AMBIENT AIR (ULTRA).

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Aims: 1) to compare available aerosol spectrometers to measure particle size and number distributions in urban air, 2) to characterize urban particles in Germany, Finland and the Netherlands, and 3) to study their health effects.
Methods: 1) three intercomparisons in ambient conditions and a laboratory comparison 2) air pollution monitoring study in winter 1996/1997, and 3) an epidemiological panel study in winter 1998/1999 in the three countries.
Results: 1) In the ambient side-by-side comparisons, the three aerosol spectrometers were found to be very well comparable in the measured total number concentrations and the concentrations of particles in the ultrafine (0.01 - 0.1 μm) and accumulation fractions (0.1 - 0.5 μm). The average concentrations differed by less than 20% and all correlations were above 0.9.In the laboratory comparison, the comparability of the instruments with laboratory aerosols of diverse chemical composition was also good in this size range. However, in the coarse fraction (particles larger than 0.5 μm in diameter), aerosol spectrometers were less comparable.
2) In the winter 1996/97, particle number and size distributions were monitored with the aerosol spectrometers in three European cities for three and a half months. There were clear differences in the concentrations of accumulation particles, PM2.5 and the blackness of the PM2.5 filters between the cities, but not in the concentrations of ultrafine particles. Correlations between ultrafine particles and PM2.5 were 0.23 in Helsinki, 0.36 in Alkmaar and 0.61 in Erfurt. Correlations of blackness of PM2.5 filters with accumulation particles were 0.7-0.8. According to principal component analysis airborne particulate pollutants seem to be divided into two source categories, one related to number concentrations and the other to mass-based information
3) In each of the three centers, 50 elderly persons with chronic cardiovascular disease will be followed up for 6 months in winter 1998/99 with biweekly intensive clinic examinations, which include measurements of cardiopulmonary function (ECG monitoring, spirometry, blood pressure) and of urinary biomarkers for lung damage. The subjects will also keep daily symptom diaries. Concurrently with the panel study, the intensive particle monitoring described above will be performed. In addition, with funding from HEI, personal exposure to PM2.5 and indoor PM2.5 levels will be monitored for 24 hours before each clinic visit in Amsterdam and Helsinki.
Conclusions: The present results underline the importance to use particle number counts together with particle size distributions, and not only mass-based measures, to characterize levels of urban air particulates.

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ORIGIN OF HIGH PM10 AND TSP EVENTS IN EASTERN SPAIN

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This study shows preliminary results obtained from a research project supported by the Spanish CICYT (AMB98-1044) on the interpretation of source origins of PM10 in Eastern Spain. To this end, levels of suspended particles (TSP and PM10) as well as major gaseous pollutants and meteorological parameters were monitored (in July 1995-July 1997) at four permanent rural stations in Easter Spain (Figure 1). In addition, field campaigns were performed to investigate possible short term variations in the particulate characteristics related with a large SO_2 emission source (particulate measurements and characterisation during direct plume impacts, formation of secondary particulate pollutants, re-distribution of particulate pollutants by local wind circulation).

The results showed a marked trend with higher particulate (TSP and PM10) levels in spring-summer lower winter levels (Figure 2). The origin of this time series distribution was interpreted in prior studies (Querol et al., 1998-a). In addition to this annual pattern, short term high PM10 events were recorded 4 to 6 times per year (Table 1). Long range transport particulate inputs affecting this area of the Mediterranean basin (mainly Sahara air mass intrusions) was found to be the origin of these events (Table 1). This findings together with an statistical relationship with daily PM10 levels and demonstrates that even PM10 levels are still highly influenced by natural particulate sources and long transport events (Figure 3).

A comparison of time series of TSP or PM10 measured at three distant sites in Spain (NE and SW Spanish coast and central-NE Spain) revealed that a high proportion of particulate peaks were common for the sites (Figure 4), and consequently the origin were related with long transport high particulate events.

Conclusions pointed that for an accurate environmental assessment, PM2.5 should be monitored instead of TSP or PM10, in order to quantify the influence of secondary particulates from gaseous emissions on the bulk atmospheric particulate levels.

Detailed chemical and mineralogical characterisation of TSP as well as a studies on the grain-size fractionation of secondary particulate pollutants and the degree of neutralisation of secondary sulphate and nitrate acidic aerosols for the same study period were reported by (Querol et al., 1998-b).

We would like to express our gratitude to ENDESA and the Direcció Genreal de Qualitat Ambiental from the Generalitat Valenciana for their help in supplying the data from the monitoring stations and for their interest in the development of this study. We would like also to thank and the Direcció Genreal de Qualitat Ambiental from the Generalitat de Catalunya for supplying the TSP time series of Cubelles.

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Table 1. First and second order particulate peaks obtained from the daily mean PM10 levels measured at the MON monitoring station during the period July'95 to August'96, and origin of the particulate peaks as deduced from the air back-trajectory analysis as well as from the comparison between the SO_2 and PM10 levels.

Major PM10 peaks

	Date	**Daily mean (PM10µg.m^{-3}N) (days)**	**Duration**	**Origin**
I.	16 July to 06 August 1995	26-56	22	Sahara
II.	25 October to 11 November	20-67	18	Sahara+PS
III.	05 to 21 June 1996	20-39	17	C.M.+ C.E.+Sahara
IV.	18 July to 02 August 1996	21-58	16	PS+Sahara

Minor PM10 peaks

Date		**Daily mean (PM10 µg.m^{-3}N)**	**Duration (days)**	**Origin**
v.	06 to 14 July 1995	18-30	9	Sahara Int.
vi.	17 to 24 August 1995	20-37	8	PS
vii.	30 Aug. to 03 Sept. 1995	21-28	5	PS
viii.	21 to 25 September 1995	21-40	5	C.E.+PS
ix.	09 to 13 October 1995	21-24	5	C.E.+C.M.
x.	29 to 30 November and	21-22	2	Sahara
	07 to 9 December 1995	21-31	3	Sahara
xi.	16 to 17 January 1996	21-30	2	Sahara
xii.	6 to 7 and 11 to 13 March 1996	21-24, 21-29	2,3	C.E.
xiii.	22 to 25 March 1996	18-29	4	Sahara
xiv.	19 to 22 April 1996	21-37	4	Sahara
xv.	12 to 16 May 1996	12-30	5	PS
xvi.	29 to 31 May 1996	18-41	3	C.E.
xvii.	25 June to 07 July 1996	15-42	14	PS
xviii.	15 to 20 August 1996	21-27	6	Sahara

Sahara, Sahara air mass intrusions; PS, local power station emissions; C.M., long range transport from the Eastern and Central Mediterranean basin; C.E., long range transport from Central and Northern Europe

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ELEMENTAL COMPOSITION OF PM10 SAMPLED IN EUROPEAN COUNTRIES WITHIN THE PEACE PROJECT

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Objectives: To determine easily and sparingly soluble fractions of many elements in PM10 samples.

Material: The samples were collected on Teflon® filters on a daily basis in urban and suburban sites in different countries within the project "Pollution Effects on Asthmatic Children in Europe" (PEACE).

Method: A two-step extraction procedure with 10 mL weak and strong acid solutions, respectively, was used to differentiate between easily soluble and sparingly soluble elements. The weak acid solution contained 0.05 M hydrofluoric and 0.01 M nitric acid, and the extraction time was 4 h. The strong acid solution contained 5.0 M and 1.1 M of the same acids, and the extraction time was 24 h.

The sample extracts were analyzed using low resolution inductively coupled plasma mass spectrometry (quadropole ICP-MS). The instrument was equipped with a Teflon® sample nebulizer, a sapphire plasma torch orifice, and a platinum-tip sampling cone. The samples were analyzed in scan dual mode (analog and puls counting). The strong acid extracts were diluted 10-fold prior to analysis. A total of 1546 PM10 samples were analyzed.

No suitable certified reference filter samples are available for testing of the extraction procedure. A certified multielement standard solution was included repeatedly in the analytical series to test the instrument performance.

Results: For 29 elements (Li, B, Na, Mg, Al, Ca, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Rb, Sr, Zr, Mo, Cd, Sn, Cs, Ba, La, Ce, W, Pb, Bi), the median concentration in the weak acid extracts exceeded the detection limit.

For the major part of the elements, the median fraction extracted into the weak acid was 50%, or more, of the total extractable amount. The highest values, ≥75%, were obtained for Na, Mg, Mn, Zn, Cd and Pb; the lowest, ≤30%, for Li, B, Ti, Cr, Zr, Cs, La and Ce. However, the variation between samples was considerable.

The concentrations of elements extracted in the weak acid varied considerably between sampling locations, and were generally higher in the urban locations than in the suburban ones. For example, median concentrations (ng/m^3) of Si in urban and suburban locations ranged <230-2000 and <230-1300, respectively. The corresponding figures for V were 1.5-8.6 and 1.3-5.3, for Fe 105-1100 and 32-520, and for Pb 5-380 and 5-80. There was a general, increasing trend of the elemental concentrations from northern Europe to southern Europe.

Conclusions: ICP-MS is a most powerful technique for determination of a large number of elements in extracts of airborne particulate samples. The detection limits are, with some exceptions, similar or better than those reported for different non-destructive techniques, such as particle induced X-ray emission, X-ray fluorescence and neutron activation analysis, that have been used for determination of the total contents of elements in aerosol samples. Information on detection limits is, however, lacking in most publications on elemental concentrations in airborn particles.

The present extraction procedure needs further evaluation with regard to its applicability for characterization of the bioavilable fraction of elements.

EPIDEMIOLOGY STUDIES

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Effects of long term exposure to air pollutants ($PM_{10}/PM_{2,5}$) on chronic respiratory diseases, lung function and survival in the Swiss population (SAPALDIA[1] and SCARPOL[2])

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[1] Swiss Study on Air Pollution and Lung Diseases in Adults

[2] Swiss Study on Childhood Allergy and Respiratory Symptoms with Respect to Air Pollution

The studies SCARPOL and SAPALDIA are well established to assess the effect of long term exposure to air pollutants on respiratory health, allergic status, symptoms, diseases and survival.

The cross-sectional parts show effects of PM_{10} on respiratory symptoms (adults and children) and on lung function.
SAPALDIA has established a well defined cohort, which will be reexamined in 2000/2001 (health assessment as in the European respiratory health survey).
SCARPOL has established a system of monitoring of respiratory health in school children with repeated surveys of the same population.

EXPOSURE ASSESSMENT:

- PM_{10} measurements were introduced in all SAPALDIA and SCARPOL locations in 1993 (using Harvard Impactors).
- Also since 1998 $PM_{2,5}$ are monitored in all SAPALDIA and SCARPOL regions.
- At the same time the particulate size distribution was measured in 5 SAPALDIA locations.
- Since 1996 EXPOLIS assesses the personal exposure in Basel and BRISKA the spatial distribution of ambient particulates within one area. These projects contribute to knowledge about exposure in air pollution epidemiology.

FURTHER DEVELOPMENTS:

- The Swiss studies on the effect of long term exposure to air pollutants on human health have developped concepts to improve exposure assessment and to define cumulative exposure to air pollutants over longer periods (between 15-20 years and life long exposure).
- These improved exposure definitions will help in the assessment of changes in the health status and the contribution of air pollution to these changes in cohorts to be followed over the next ten years.

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PERSONAL EXPOSURE TO FINE PARTICULATE MATTER AND ITS RELATIONSHIP TO SHORT-TERM CHANGES IN CARDIOVASCULAR AND RESPIRATORY HEALTH INDICATORS

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The aim of the investigation is to assess the relationship between ambient levels, indoor levels, and personal exposure to fine particles, and cardiovascular and respiratory health of elderly subjects.

In the framework of an EU funded study, panel studies are conducted in Amsterdam and Helsinki. Panels consist of elderly subjects with cardiovascular disease. Subjects are followed for 6 months. Measurements are made of lung function, ECG and heart rate variability using Holter recorders once every 2 weeks including a mild exercise challenge. Subjects keep diaries to record symptoms and medication use.

At a fixed site, ambient particle concentrations (PM10, PM2.5, particle numbers) are measured. Particle reflectance (validated in a subset by measurement of Elemental Carbon) is measured as a marker for diesel exhaust.

With additional HEI funding, measurements of indoor and personal exposure to fine particles are added.

We have conducted several studies on personal and indoor exposure to PM10 as well as Fine Particles (FP). The aim of these studies was to establish time-series relationships between personal exposure to either PM10 or FP and ambient and indoor exposure. The results of these studies showed that the time-series relationship (correlation within subjects over time) between ambient and personal PM10 was reasonably high in adults as well as children. The time-series relationship between personal and ambient FP was found to be even stronger, suggesting that for FP, ambient concentrations are a good proxy for personal exposure. One of the Finnish investigators is currently co-ordinating a EU funded study (EXPOLIS) aimed at measuring and modelling personal exposure to several air pollutants (including PM2.5) in various European cities.

We perform indoor and personal exposure measurements of PM2.5 and its chemical composition in the panel studies described above. The number of subjects in the panels is about 50, and we measure each subjects' indoor and personal exposure biweekly, during the 24-hour period preceding the intensive health status measurements.

The field work of the study has started in the fall of 1998. The study is now fully implemented in the field. Results are not yet available, as the field work will continue until the summer of 1999.

The results of the study will expand the currently small database on personal exposure to FP in relation to ambient and indoor FP considerably. The study will also improve our knowledge on the relative contribution of ambient and personal exposure to FP to explaining acute health effects.

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CESAR - Central European Study of Respiratory Health and Air Pollution. Some Preliminary Findings

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Overview of study

The Central European Study on Air pollution and Respiratory health (CESAR) was funded by the Commission of the European Communities (CEC). A cross-sectional study was conducted in primary school children to assess the effects on respiratory health of long-term exposure to ambient air pollution.

Regions were selected for the study after consultation with local experts. These regions were: Sofia (Bulgaria), Ostrava (Czech Republic), Komarom County (Hungary), Silesia (Poland), Zlatna and Ploiesti (Romania) and Banska Bystrica (Slovak Republic). In each region four study areas were selected. Exposure was measured in all study locations for a year.

The study population comprised 21743 children aged 7 to 11 (about 3500 children per country). In each study area the prevalence of respiratory symptoms and disorders, medication use and allergies was assessed by use of a questionnaire administered to the parent. In the questionnaire, information was also gathered on other risk factors, including indicators of socio-economic status, housing and indoor air quality, family health and nutritional status. Lung function measurements took place in the older half of the children (about 2000 per country). The atopic status of a sub group of the children was assessed with specific IgE in serum.

Preliminary findings

The principal analyses involve the use of regression methods to investigate the association of the symptom prevalence, adjusted for important potential confounders at individual level, with the exposure to particulates. In general there were higher prevalences of bronchitis and reported cough as compared to symptoms of wheeze or diagnosis of asthma. A wide number of potential confounders were identified, but indicators of socio-economic status and exposure to tobacco smoking indoor were judged to be of particular importance. Within each of these, mothers education and smoking habits of both parents showed the strongest associations with several respiratory health measures and have been included in the analyses presented here, along with age and sex. Thus the differences between towns can be seen after having taken into account differences in the patterns of mothers' educational level and parents' smoking habits, and the age and sex of the children.

For several different respiratory outcomes there is a suggestion of a positive association with particulate exposure in between country comparisons. Within country analyses are much less consistent. These preliminary analyses, focusing on the outcomes and exposures of primary concern, suggest that between-country differences in reported childhood illness are more substantial than within-country differences. Between-country differences are not explained by available data on socio-economic status or smoking habits. Differences in air pollution between countries may account for some of the observed health differences, however within-country analyses give limited support at this stage, for this explanation. Full analyses will address this apparent contradiction.

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SHORT-TERM EFFECTS OF AIR POLLUTION ON HEALTH
A EUROPEAN APPROACH TO METHODOLOGY, DOSE-RESPONSE ASSESSMENT AND EVALUATION OF PUBLIC HEALTH SIGNIFICANCE
(THE APHEA2 PROJECT)

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The APHEA2 project started on February 1st, 1998. It is sponsored by the European Commission DGXII, Contract number ENV4-CT97-0534, under the ENVIRONMENT AND CLIMATE 1994-1998 Programme. Twenty research groups, several of which represent research networks, from 19 different European countries, are participating. The objectives of the project are: 1. To investigate dose-response relationships between air pollutants and daily mortality and hospital admissions, 2. To test hypotheses about mortality displacement caused by the short-term effects of air pollution, 3. To investigate regional differences and effect modifiers and 4. To conduct a second stage analysis with data from 32 cities on mortality and 7 cities on hospital admissions and, possibly, a combined analysis with the U.S., H.E.I. sponsored NMMAPS project.

Air pollution data include measurements of ambient particles (either as PM_{10} or black smoke), nitrogen dioxide (NO_2), ozone (O_3), sulphur dioxide (SO_2) and carbon monoxide (CO). Most air pollution measurements will come from operating fixed site monitors. To make exposure data as comparable as possible criteria are applied concerning placement of monitors and measurement completeness. Mortality data will be available from 34 cities, 9 of which are in Central-Eastern European countries. Hospital admissions data from 7 cities will be analysed. Analysis of mortality time series will be done by cause of death and age. Respiratory and cardiac hospital admission will be analysed separately. Currently, the individual city analysis is under way.

The APHEA2 project is a continuation of the older APHEA (Air Pollution and Health: a European Approach) project, sponsored also by the E.C. under ENVIRONMENT 1991-1994. The APHEA project was the first large scale multi-center project addressing short-term effects of air pollution on health and included 11 research groups and data from 15 cities. When the project started (in 1993) there were few available results from Europe analysing current air pollution data. The objectives of this older project were to provide estimates of air pollution short-term health effects in Europe, to standardise the methodology of analysing epidemiological time series data and applying a meta-analysis procedure. The project finished in 1995 and the results have been published and had a considerable impact in policy making at the national and E.U. level. All pollutants studied, with a partial exception of NO_2, were found to have small acute effects on daily total, cardiovascular and respiratory mortality. Particulate matter and ozone levels were consistently associated with respiratory and COPD admissions. NO_2 levels were associated with asthma admissions. The estimated effects for particulate matter were consistent with those observed in the U.S. studies but they were on the lower side of the U.S. range of estimates. The above effects were observed in locations were, on most days, air pollution levels were well below the set standards.

Several open questions remained. Are there regional differences in air pollution health effects? What is the independent effect of each pollutant? Is there synergy? What is the extent of mortality displacement caused by the short-term effects of air pollution? The APHEA 2 project aims at addressing the above questions.

There are advantages and disadvantages in such a large scale multicenter project. Among the advantages, the use of a larger data base and a stardardised protocol, the varying pollutant mixes, the different environmental and socioeconomic conditions, the transfer of know-how and the joining of scientific forces and different perspectives should be mentioned. The major disadvantage is the difficulty in managing and coordinating all the research groups.

GÖRAN PERSHAGEN

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URBAN AIR POLLUTION AND LUNG CANCER - A POPULATION BASED CASE-CONTROL STUDY FROM STOCKHOLM

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Background The epidemiological evidence on urban air pollution and lung cancer is inconclusive and suffers from crude aggregate exposure assessment and lack of adequate information on important confounders.

Methods We conducted a population-based case-control study among men aged 40-75 encompassing all cases of lung cancer 1985-1990 in stable residents of Stockholm County 1950-1990. The resulting study group comprised 1042 cases and 2364 controls. Questionnaires to subjects or next-of-skin elicited information regarding smoking and other risk factors for lung cancer, including occupational and residential histories. Retrospective emission databases were created for SO_2 and NO_x/NO_2 as indicators of air pollution from residential heating and traffic, respectively. Spatial distribution was estimated using validated dispersion models and estimated local annual air pollution levels were linked to residential addresses. The intensity and probability of exposure to seven occupational factors, including diesel exhaust and combustion products, were assessed for every work period.

Results Average traffic-related NO_2 exposure during 30 years in the top decile was associated with a relative risk (RR) of 1.2 (95% confidence interval [CI] 0.8-1.6), adjusting for smoking, residential radon and occupational exposure. A considerable latency period was suggested and the RR for the top decile of NO_2 exposure 21-30 years previously was 1.4, 95% CI 1.0-2.0. A multiplicative interaction between NO_2 exposure and smoking was suggested, however, with a weaker effect among heavy smokers. No associations were observed for SO_2. Cumulative exposure to diesel exhaust and combustion products was related to lung cancer, with RRs in the highest quartile of exposure of 1.6 (95% CI 1.1-2.4) and 1.7 (1.1-2.6), respectively.

Conclusions This study indicates that urban air pollution is a risk factor for lung cancer and suggests that traffic emissions may be particularly important. Further support for a role of diesel exhaust and combustion products in lung cancer induction was provided by data from occupational exposure.

JONATHAN SAMET

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NMMAPS: NATIONAL MORTALITY, MORBIDITY AND AIR POLLUTION STUDY.

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Time-series studies have shown associations between air pollution concentrations and morbidity and mortality. These studies have largely been conducted within single cities with varying methods. Key questions remain unaddressed concerning the findings, including 1) the extent of the heterogeneity of air pollution effects across locations and its sources; 2) the effect of measurement error on the estimated effect of air pollution; and 3) the public health significance of the short-term associations. NMMAPS comprises the development of methods to address these questions and their application to national data sets on mortality and hospitalization among persons 65 years of age and older, as an index of morbidity. For analyzing mortality data from multiple locations and summarizing the findings, a Bayesian hierarchical regression method has been developed and applied to data from 8 cities. Frequency domain regression methods have been used to assess mortality displacement, with the initial finding that air pollution effects can be assessed in time-response bands that are relatively resistant to short-term mortality displacement. The consequences of measurement error are being comprehensively exposured using available data sets. Data bases have been assembled on mortality of the 100 largest U.S. cities and for morbidity. In the next phase of NMMAPS, the morbidity and mortality analyses will be completed and the combined analyses of morbidity and mortality will be undertaken. The methods of NMMAPS should prove useful for future surveillance for the health effects of air pollution.

FRANK SPEIZER (for Douglas Dockery)

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AIR POLLUTION AND INCIDENCE OF CARDIAC ARRHYTHMIA.

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Elevated levels of particulate air pollution have been associated with an increase in hospital admissions and mortality for cardiovascular diseases. This study tests the hypothesis that patients with implanted cardioverter defibrillators (ICD) experience potentially life-threatening arrhythmias associated with particulate air pollution episodes.

Tachycardia and ventricular fibrillation were identified from records of 100 patients with ICD devices at the Beth Israel-Deaconess Hospital Cardiac Device Clinic for the years 1995 through1997. During the same period, 24 hour mean concentrations of particulate matter (PM_{10} and $PM_{2.5}$), Black Carbon, and gaseous air pollutants were measured in the Boston area.

Air pollution concentrations were moderate during this period. However, an increase of 26 ppb NO_2 was associated with increased tachycardia and ventricular fibrillation two days later, odds ratio of 1.8 (95% confidence interval: 1.1 to 2.9). Patients with repeated events (10 or more events during a three year follow-up) were especially at risk of experiencing arrhythmia in association with $PM_{2.5}$ (odds ratio: 1.6 for an increase of 22 $\mu g/m^3$ (95% confidence interval: 1.0 to 2.6)) and NO_2 (odds ratio: 2.8 for an increase of 26 ppb (95% confidence interval: 1.5 to 5.1)).

These results suggest that elevated levels of air pollutants are associated with potentially life-threatening arrhythmia leading to therapeutic interventions by an implanted cardioverter defibrillator.

RADIM ŠRÁM

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MATERNAL EXPOSURE TO PM10 IN VARIOUS GESTATIONAL STAGES AND FETAL GROWTH.

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OBJECTIVES. The aim was to evaluate a possible impact of PM10 on the risk of intrauterine growth retardation (IUGR) in Teplice - a district in highly polluted area of Northern Bohemia.

METHODS. All European single live-births occurred in two year period (April 1994 to March 1996) in the district of Teplice were included. Information on reproductive history, health and lifestyle was obtained from medical records and maternal questionnaires. Mean concentration of PM10 in particular gestational month was calculated for each mother on the basis of continual PM10 monitoring data. Relative risk of IUGR for various PM10 levels and gestational stages was estimated after adjusting for a wide range of covariates using logistic regression models. Three PM10 concentration intervals were used (<40, 40-64 and $\geq 65 \mu g/m^3$).

RESULTS. PM10 daily averages varied between 5 and $330 \mu g/m^3$ and 30day averages between 28 and $98 \mu g/m^3$. Adjusted Odds Ratio (OR) of IUGR for fetuses exposed in the first gestational month to PM10 mean levels $40\text{-}64 \mu g/m^3$ is 1.6 times ($P<0.01$) and for fetuses exposed over $65 \mu g/m^3$ is 1.8 times ($P<0.05$) the risk for those below $40 \mu g/m^3$. No other associations of IUGR risk with particulate matter were found.

CONCLUSIONS. Consistent and significant association of IUGR Odds Ratio with the level of PM10 during the first gestational month was observed. Influence of particles or other associated air pollutants on fetal growth in early gestation is one of several possible explanations of these results. Timing of this effect is compatible with a curreut hypothesis of IUGR pathogenesis. Seasonal factor, one of the other possible explanations, is less probable. More investigation is required to examine these findings and alternative explanations.

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MORTALITY AMONG PATIENTS WITH COPD DUE TO URBAN PARTICLES: A CASE-CROSSOVER ANALYSIS.

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Our objective is to assess the acute association between particulate air pollution and mortality in a population based cohort of subjects recruited through an emergency room admission for a chronic obstructive pulmonary disease (COPD) exacerbation, using a case-crossover analysis. The fact that the unit of observation was the individual allowed us to explore which characteristics of these patients conferred a major susceptibility to air pollution. In addition, the use of a case-crossover design avoids the common concerns about the complex mathematical methods used to assess the acute health effects of environmental exposures. Persons who died during the period 1990-1995 among those residents in the city of Barcelona (Spain), aged over 14 years, who visited emergency room services for a COPD the period from 1985 to 1989 in the four largest urban hospitals were included (a total of 1850 males and 462 females). Vital status was ascertained through record linkage of the cohort individuals with the Catalonia Mortality Registry. Causes of death were based on the underlying cause in the death certificate. Air pollution exposure was measured at the city monitoring stations, which provide an average for the entire city. Particle levels (measured as black smoke) were associated with mortality for all causes of death (odds ratio for an increase of 100 μg/m3, adjusted for temperature, humidity, and influenza = 1.70, 95% confidence interval =1.09-2.64) with a relative risk about 10 times higher the observed when all persons, instead of COPD patients, were assessed in previous studies. The association was particularly strong for respiratory causes (2.31, 1.13-4.73), but is also elevated over the general population results for cardiovascular causes (1.45, 0.66-3.23). The association with SO_2, although positive, was not significant with any of the causes ($p > 0.2$). Older females, patients admitted in intensive care units, patients with a higher rate of emergency room visits, and patients visited in days with high pollution were at higher risk of dying in association with black smoke. These results reinforce the deleterious role of urban pollution, and provide new epidemiological information on possible factors that confers susceptibility to the acute role of air pollution.

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PARTICULATE AIR POLLUTION AND DAILY MORTALITY IN ERFURT (EAST GERMANY): CHARACTERIZATION OF THE AEROSOL SIZE DISTRIBUTION AND ELEMENTAL COMPOSITION

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There is an ongoing debate on the question which size fraction of particles in ambient air may be responsible for short-term effects observed in humans. Some hypotheses aimed at explaining the health effects of ambient aerosol focus on the chemical composition of the particles. A well characterized elemental composition may also provide means for identifying the source of the particles in different fractions. As a part of an ongoing HEI study on the short term effects of particulates on daily mortality, we report here on the results of an extensive program to characterize the size distribution. composition and sources of the local aerosol.

The mean particle size distribution of the ambient aerosol is continuously determined on a daily basis, using a Mobile Aerosol Spectrometer (MAS). Particles in the size range 0.01-0.1 μm are classified by an electrical mobility analyzer, particles in the range 0.1-2.5 μm by an optical particle counter. Size discriminated samples of the aerosol using a 9-stage low pressure cascade impactor (0.06-16 μm) are collected on a weekly basis during the first two years and on a daily basis during the last year of the study. The elemental composition is analyzed using PIXE (proton Induced X-Ray Emission).

Our data show the following: (1) The variability of number and mass concentrations of fine and ultrafine particle fractions can be used to analyse health effects on a daily (or even shorter) scale. (2) In Erfurt, there is a strong shift over the years with decreasing mass concentrations and increasing number concentration. (3) The analysis of the elemental composition helps to identify the sources for particles in different sizes fractions.

In conclusion. the implementation of detailed characterization in the size. the composition and the number concentration of particles within an epidemiological study is a powerful tool to better understand the origin of the observed health effects.

EPIDEMIOLOGICAL STUDIES ON HEALTH EFFECTS OF FINE AND ULTRAFINE PARTICLES IN GERMANY

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Four studies address possible effects of fine (diameter < 2.5 μm) and ultrafine (diameter < 0.1 μm) particles.

(1) A panel study was conducted in adult asthmatics in Erfurt (1991 - 92). Symptoms increased and lung function decreased in association with 5 day means of particle concentrations. The strongest effect was observed for the number concentration of the ultrafine particles.

(2) Daily mortality is studied in Erfurt (1995 - 99) with respect to particles and elemental compositions. Particle mass decreased over time while particle numbers increased. An interim analysis shows borderline significant increases in mortality in association with both fine and ultrafine particles.

(3) An panel study assessing symptoms, EKGs and lung function in elderly subjects with cardiovascular disease is ongoing in Amsterdam, Erfurt and Helsinki (1996 - 2001). Number concentrations (ultrafine particles) are comparable throughout the locations, while particle mass concentrations (fine particles) show differences.

(4) A case crossover study with survivors of an acute myocardial infarction has been started to analyse the possible role of fine and ultrafine particles on the onset of myocardial infarctions in Augsburg (1999 - 2001).
In conclusion, these studies might have major regulatory implications if the ultrafine particles show relevant health effects. Further research on the role of the probably very relevant particle surface, on chemical and source-specific characterization of particles, and on respiratory and cardiovascular endpoints is needed.

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TOXICOLOGY
STUDIES

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BIO-PHYSICOCHEMICAL PROPERTIES AND PULMONARY TOXICITY OF AIR PARTICULATE MATTER

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Epidemiology studies have reported excess mortality and enhanced morbidity in susceptible subpopulations following exposure to ambient air particulate matter (PM) at levels below the National Ambient Air Quality Standard. The bio-physicochemical properties and pathophysiological mechanism(s) responsible for the observed PM health effects must be determined in order to effectively manage this air pollutant. To address these uncertainties in PM health risk assessment, initial research efforts have been directed towards determining the bio-physicochemical properties of combustion emission and ambient air PM which are responsible for their corresponding health effects. Results from these studies have demonstrated the acute pulmonary toxicity of fugitive residual oil fly ash (ROFA) and coal fly ash (CFA) was due to their metal content and bioavailability. This finding was confirmed in studies which demonstrated that similar acute lung injury could be induced by ROFA or CFA when exposures were conducted on the basis of an equi-metal rather than an equi-mass dose-metric, suggesting that chemical composition, i.e., metal content/bioavailability, was responsible for the observed effects. Similar results were obtained when the acute pulmonary toxicity of various ambient air total suspended particulate (TSP) samples were examined on an equi-metal exposure dose-metric. Physicochemical and *in vivo* toxicity analyses performed on size-fractionated ambient air TSP samples revealed the fine (0.4-3.3μm MMAD) fraction to have a higher bioavailable metal content and induced more lung injury than the unfractionated TSP samples. *In vitro* cytotoxicity studies performed on ambient air PM_{10} samples collected from various regions of Mexico City revealed the most cytotoxic sample to be derived from the industrialized northern region. This PM_{10} sample had the highest content of sulfate and metals as well as bioavailable metal. Pulmonary toxicity analyses conducted on size-fractionated ambient air PM collected in 1995 from Washington, DC demonstrated the fine ($PM_{<1.7}$) fraction to be more toxic when compared to the coarse ($PM_{3.7-20}$) fraction. The $PM_{<1.7}$ physicochemical properties were found to resemble particles derived from anthropogenic combustion sources with regards to particle acidity and sulfate content as well as bioavailable metal content. A heat-labile constituent responsible for a significant amount of pulmonary neutrophil influx induced by ROFA and ambient air PM exposure has been detected. Preliminary physicochemical analyses have identified potential causative organic constituents. Particle-associated endotoxin, a biogenic constituent of ambient PM, was found not to contribute directly to the acute lung inflammation induced by any ambient air PM sample examined to date. These data demonstrate that particle size, acidity and composition, such as sulfate content, metal content/bioavailability, and potentially organic constituents, are important physicochemical properties, each of which can influence acute PM toxicity. (This abstract does not reflect EPA policy).

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LOCAL INFLAMMATORY AND SYSTEMIC EFFECTS FOLLOWING SHORT-TERM INHALATION EXPOSURE TO FINE AND ULTRAFINE CARBON BLACK IN RATS

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An ultrafine component (particles with diameter<100nm) has been hypothesised to play a role in mediating the pathogenic effects of PM10/2.5. We have used ultrafine carbon black (uf-CB) 14nm in primary particle diameter, to study the potential pathogenic effects of ultrafine particles in ambient air. As a size control we used carbon black with a primary diameter of 260nm (CB). These two samples were used to investigate the hypothesis that ultrafine particles have effects at low exposure. Inhalation exposure studies in rats with the two carbon black preparations at 1mg/m3 for 7 hours showed a pro-inflammatory and oxidative stress response with the uf-CB; there was also an increase in Factor VII detectable in the blood in the uf-CB-exposed animals. Factor VII is a pro-coagulant factor that is an independent risk factor for cardiovascular disease. CB had none of these effects. Instillation studies revealed that, at low instilled masses, the uf-CB was more inflammogenic on a mass basis but that at higher mass doses the CB became apparently more inflammogenic. This may be explained by increased interstitialisation of uf-CB leading to retention of PMN in the interstitium. More recently, studies with chelators suggest that the inflammation caused by ufCB is not iron-mediated and does not involve any soluble factors. The mechanism of the local and systemic effects of ultrafine carbon black at low exposure require further investigation.

This research was supported by the Medical Research Council

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RESPIRATORY RESPONSES TO URBAN PARTICLES INHALED AT CONCENTRATIONS CLOSE TO AMBIENT LEVELS

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Three longitudinal studies with healthy dogs exposed over periods of a year to hydrophilic sulfur-related fine surrogate particles were performed and the following hypotheses tested:

- particle-associated sulfur-IV induces bronchitic responses
- particle-associated hydrogen ions induce detremental responses
- when sulfur-IV and hydrogen ions are inhaled, responses are enhanced

Considering lung mechanics, integrity of alveolar-capillary barrier, macrophage-associated defense capacity, intrapulmonary particle transport, pulmonary cell number, cell injury, type II cell function, antiproteolytic and oxidant status and extracellular proteins, all three hypotheses had to be rejected:

- chronic exposures induced subtle responses of healthy canine lungs, but no pathological processes were initiated
- sulfur-IV induced early stages of lung emphysema, altered the integrity of the alveolar-capillary barrier and macrophage-mediated particle clearance, and suppressed the defense capacity of macrophages
- hydrogen ions induced early stages of fibrotic leasons, increased proliferation of type II cells and release of alkine phosphatase
- antagonistic rather than synergistic interactions occurred between responses induced by sulfur-IV and hydrogen ions

In addition, three short-term studies with rats exposed over periods of 10 days to hydrophobic ultrafine surrogate particles without surface contaminations (0.02 μm carbon, silver and iron oxide particles at concentrations of 10^6 cm^{-3}) were performed and the following hypothesis tested: ultrafine particles cause acute respiratory responses due to their generic nature as particles. Considering the integrity of the alveolar-capillary barrier, pulmonary cell differentiation, proinflammatory reactions and histomorphology, the hypothesis had to be rejected:

- none of the particles altered integrity of the alveolar-capillary barrier, cell differentiation and histomorpholgy
- none of the particles produced proinflammatory reactions

CHARACTERIZATION OF PARTICLES IN URBAN AIR

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Surrogate particles are often used to simulate human and animal exposure to ambient particles. With these particles physicochemical properties of ambient particles are simulated. The following properties of ambient particles were determined over a long period of time at different urban sites in Central Europe: particle size and mass distributions in the range 0.01 - 2 μm with a mobile aerosol spectrometer, hygroscopic properties with a tandem differential mobility analyser and chemical composition by laser-induced mass spectrometry. The following results were obtained:

- ▩ 0.01 - 0.10 μm particles (ultrafine particles)
    - ▩ their number concentration determines that of urban particles
    - ▩ their number concentration remained unchanged over the last decade
    - ▩ their mass concentration is negligible
    - ▩ they can be hydrophobic or hydrophilic
    - ▩ the major component in hydrophilic particles is sulfur
    - ▩ the major component in hydrophobic particles is carbon

- ▩ 0.1 - 1 μm particles (fine particles)
    - ▩ their mass concentration determines that of urban particles
    - ▩ their mass concentration decreased over the last decade
    - ▩ their number concentration is negligible
    - ▩ they can be hydrophilic or hydrophobic
    - ▩ the major component in hydrophilic particles is sulfur
    - ▩ the major component in hydrophobic particles is carbon

Conclusions: to study respiratory responses initiated by inhaled urban particles in controlled human or animal exposure scenarios, hydrophobic carbon-related ultrafine surrogate particles and hydrophilic sulfur-realted fine surrogate particles are most suitable.

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STUDIES WITH ULTAFINE PARTICLES.

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The ambient fine particle mode consists of ultrafine (~5-80 nm) and accumulation mode (~100-1000 nm) particles. Our hypothesis that ultrafine particles (UP) rather than accumulation mode particles (AP) are causally involved in adverse effects is based on (*i*) results of our earlier studies with particles of both sizes and (*ii*) on the finding that mass concentrations of urban ultrafine particles can reach high levels of up to ~40-50 $\mu g/m^3$, equivalent to number concentrations of 3 - 4 x 10^5 particles/cm^3. The objectives of our exploratory studies were (a) to evaluate basic principles of inducing adverse effects by using UP of known toxicity (although not occurring in the ambient atmosphere); (b) to characterize generation and coagulation behavior of UP of relevance for urban air; (c) to study the influence of age, disease and co-pollutants as modifying factors. We used ultrafine Teflon® (polytetrafluoroethylene, PTFE) fumes (median particle size ~18 nm) generated by heating PTFE in a tube furnace to 486°C to evaluate basic principles of UP toxicity which might be helpful in understanding potential effects of ambient UP. Ultrafine Teflon® fumes at UP concentration of ~50 $\mu g/m^3$ are extremely toxic. We found that the ultrafine Teflon® particle phase alone is not toxic at these exposure conditions and neither are Teflon® fume gas phase constituents; only the combination of both phases causes the high toxicity, suggesting a carrier mechanism of the UP for adsorbed gas phase compounds. We also found a rapid translocation of the UP across the epithelium after their deposition, which appears to be an important difference from larger-sized particles. Furthermore, the pulmonary toxicity of the ultrafine Teflon® fumes could be prevented by adapting the animals with short 5-min. exposures on three days prior to the 15-min. exposure. This shows the potential importance of pre-exposure history for the susceptibility to acute UP effects. Aging of the Teflon® fumes for about 4 mins. leads to a predicted coagulation to >100 nm particles which in combination with the gas phase no longer caused toxicity in exposed animals. This result is consistent with a greater toxicity of UP compared to AP particles. With respect to UP of environmental relevance, UP of carbon, platinum, iron and iron oxide, vanadium and vanadium oxide were generated by electric spark discharge and characterized. The count median diameter of the ultrafine carbon particles was ~26 nm, and of the metal particles 15-20 nm, with geometric standard deviations of 1.4 - 1.7. For UP carbon, ~100 $\mu g/m^3$ is equivalent to 1 x 10^6 particles/cm^3. Homogeneous coagulation of these UP in an animal exposure chamber occurred rapidly at 1 x 10^7 particles/cm^3 so that particle size quickly grew to >100 nm. Thus, controlled aging of UP carbon allows the generation of AP carbon (due to coagulation growth) for performing comparative toxicity studies. We also developed a technique to generate UP consisting of the stable isotope ^{13}C, using ^{13}C graphite electrodes made in our laboratory from amorphous ^{13}C powder. These particles are particularly useful tools to determine deposition efficiencies of UP carbon in the respiratory tract of experimental animals and their translocation to extrapulmonary sites. With respect to compromised animal models we used models of pulmonary emphysema, and a low dose endotoxin inhalation model to prime target cells in the lung. Additional modifying factors were age and co-pollutant (ozone) exposure. Young mice, whether healthy or emphysematous (elastase-induced), were not affected by 6-hr. inhalation of ultrafine carbon or platinum particles at ~110 $\mu g/m^3$. Old healthy mice also were not affected by exposures to these particles, and only old emphysematous (elastase-induced) animals showed a significant, although only slight, pulmonary inflammatory response. Subsequent multi-group (8 groups) inhalation studies in young and aged rats using the LPS inhalation model with UP carbon in combination with O_3 analyzed by a 3-way ANOVA showed (neutrophils in lung lavage): In the young animals, the UP carbon induced a small effect when inhaled alone which was additive to the effects of LPS and ozone; however, although ozone alone also caused a significant effect, ozone was less than additive in the LPS-sensitized lung. In the aged rats, UP carbon alone did not cause a response whereas it was synergistic with the effects of ozone; in the LPS-sensitized lung of the aged rats, the effects of UP carbon in combination with ozone were additive. We conclude from these studies that carbon particles inhaled by rodents as singlet UP of 10 - 50 nm size can induce an inflammatory response in the aged compromised lung. This effect occurs at concentrations which lead to similar deposited lung doses as are predicted to be deposited in humans exposed to episodic high urban UP concentrations. Significant effects of UP carbon are seen only in the compromised or in the primed lung, and are most obvious (synergistic) with co-exposure to ozone. Particles of the size of the ambient AP do not show effects at a lung dose - expressed as particle mass - which shows effects with UP.

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LOW LEVELS OF DIESEL EXHAUST INDUCE AIRWAY INFLAMMATION

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Introduction: Particulate matter (PM) pollution is of considerable concern for airway health. Diesel exhaust (DE) is a major source of PM pollution. This group has experience from a series of controlled chamber exposures with diluted diesel exhaust, using a carefully developed and validated exposure set up, used for studies of DE effects in humans. We have shown that exposure to a DE concentration of 300 $\mu g/m^3$ PM_{10} induced a pronounced airway inflammation in healthy subjects. This was reflected not only in BAL but even more so in the bronchial mucosa, as reflected in immunostained biopsies (Salvi et al. Am J Respir Crit Care Medicine, in press).

Aim: To investigate if exposure to a low concentration of DE (100 $\mu g/m^3$ PM_{10}) would also induce inflammatory responses in the airways. The HEI supported project comprised two study populations, healthy subjects and asthmatics, of which data from the first group are complete at the time of the Brussels meeting.

Methods: 25 healthy volunteers were randomly exposed to DE (100 $\mu g/m^3$ PM_{10}) and air for two hours in an exposure chamber on two separate occasions. Bronchoscopy with bronchial wash (BW), bronchoalveolar lavage (BAL) and biopsy sampling were performed six hours after exposure. Biopsies were processed in GMA and immunostained for determination of cell counts and vascular adhesion molecule expression.

Results: Significant increases in IL-6, IL-8 concentrations and percent neutrophils were found in BW. Endobronchial biopsies revealed a significant upregulation of the adhesion molecule P-selectin and VCAM-1 in the vascular endothelium, but not ICAM-1 at this time point. CD3+ lymphocytes decreased in bronchial epithelium and correspondingly, lymphocyte numbers increased in BAL. Lung function measurements showed a bronchoconstrictive pattern.

Discussion: Exposure to diesel exhaust containing PM 100 $\mu g/m^3$ induces inflammatory reactions in the airways of healthy subjects. The acute cytokine and chemokine presence with IL-6 and IL-8, together with enhanced expression of the vascular adhesion molecules P-selectin and VCAM-1 in the biopsies, suggest an early state of recruitment of inflammatory cells. Mechanisms for neutrophil migration into the airways were present, though not fully developed at 6 hours after exposure. The inflammatory changes showed qualitative similarities, but were less pronounced than in the 300$\mu g/m^3$ diesel study, performed with identical protocol. The profile of the inflammatory markers suggests a slower development of airway inflammation after 100 μg PM /m^3, than after 300 $\mu g/m^3$. A later time point of bronchoscopy, 12-18 hours after exposure, may have reflected a more established state of inflammation. Material from 14 similarly investigated asthmatics is under processing, and may yield additional understanding of diesel exhaust effects.

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HEALTH EFFECTS OF PARTICULATE MATTER - TOXICOLOGY AND DOSIMETRY

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Toxicity studies on components of ambient particulate matter (PM_{10}) are conducted within the framework of a research program on adverse health effects of ambient particulate air pollution. These studies are part of the Dutch "Wintertime Smog and Traffic Program ", performed by order and at the account of The Ministry of Housing, Spatial Planning and Environment. The first part of this program included epidemiological studies on health effects of ambient PM_{10} and the possible role of traffic emissions. The second part of this program includes toxicity studies on ambient PM_{10} and has been designed in accordance with the simplified strategy of the health risk assessment model for ambient PM_{10}, i.e. the "Pentagon" model for possibly main critical fractions with respect to particle size and chemical composition.

This research will be a meaningful help to prove causality and biological plausibility of effects of PM_{10} in healthy people and, in particular, asthmatics as a possible susceptible group, especially when used in combination with ambient PM_{10} deposition models and exposure-dose relationships.

Important questions about causality and biological plausibility of PM-associated health effects are:

- Can ambient PM cause health effects and toxicity?
- Are these effects mechanistically understandable at low PM levels?
- Do effects depend on specific PM subfractions and compositions $PM_{2.5}$ versus PM_{10} versus number concentrations?
- Do effects depend on specific pollution situations and PM mixtures and sources?

Several approaches are applied to search for causality and plausibility.

A. Inhalation studies with model aerosols (carbonaceous and secondary PM),
B. Inhalation studies using ambient PM in a mobile laboratory equipped with an Harvard-USEPA ambient fine particle concentrator, allowing studies on locations dominated by various sources of emissions,
C. Instillation studies PM fractions (ultrafine, fine and coarse fraction PM),
D. *In vitro* toxicity testing investigating possible mechanisms,
E. Dosimetry modelling for animal-to-human extrapolation

Fig. 1 Mobile Exposure Laboratory equipped with an Harvard-USEPA Ambient Fine Particle Concentrator