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PARTICULATE MATTER IN URBAN AIR: HEALTH RISKS, INSTRUMENTATION AND MEASUREMENTS, AND POLITICAL AWARENESS

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Particulate matter, urban air quality, vehicle emissions, health effects, particle number, particle mass, chemical composition, sampling, analysis, SJAC, CPC, LAS-X, policy.

CONTENTS

SUMMARY

The influence of traffic on the presence of particulate matter (PM) in the air within large urban agglomerations is the major issue of this study. This report describes the scientific results obtained last year concerning instrumental development and measurement campaigns. In the Appendix findings are given of a study in which the political attitude of Dutch authorities with respect to the Particulate Matter issue have been analysed. This work is carried out in close co-operation with ECN Policy Studies.

The major origin of particulate matter (PM) in urban air is vehicular traffic. Due to the intensity of traffic in large cities considerable quantities of primary particles (composed mostly of soot and organic material), as well as certain gases (acting as precursors for new particle formation) are emitted. PM due to vehicular emissions consists of a large part of ultrafine particles (diameter smaller than 0.1 μm) that may penetrate deep into the human lung system. Also, it is known that a number of possible causative agents is present such as policyclic aromatic hydrocarbons (PAH) and toxic metals. However, for several aspects quantitative data is still missing on concentrations and composition of PM, and its relationship with health effects. It is the concern on health effects that is the determining factor for current (and future) scientific research (and associated financial funding) and for the political measures.

Our study addresses four items. The relationships between adverse health effects and the physical and chemical characteristics of PM in air are described (Ch. 2) as far as these are found in the scientific literature available at present. An important conclusion from this study is that there is evidence that the health effects in urban air are due to the increased presence of toxic, insoluble components (soot, PAHs and metals). These components are part of the smaller particles (i.e. a diameter less than 1 μm) being predominantly emitted in very large numbers by vehicular traffic. The outcome of this study is used as a leading guide in the selection of the other items in this project.

Ch. 3 deals with some further refinements of (existing) measuring techniques operated at ECN. In 2000 the Condensation Particle Counter (CPC), an instrument for measuring the number of particles, was further investigated. The comparative study reveals that one of the three available types (CPC-3022) is the best option for measurements of PM in urban air. This is because the measurement range of this instrument corresponds most with the size spectrum of particles emitted by traffic. The other two types can better be integrated with an SMPS system.

The measurements of two experimental campaigns is the topic of Ch. 4 and 5. In these experiments number and mass concentrations in an urban environment at fixed locations or with a mobile unit were measured to estimate their spatial variability in an urban environment. Such measurements are done to discover those locations in a city that show the highest concentration levels.

In the Appendix a study is briefly outlined in which the political awareness of the various Dutch authorities (national, provincial and municipal) of the Particulate Matter problem issue has been analysed. It appears that the expertise available on this topic at the different authority-levels is limited. This makes it difficult to develop an effective local policy. The need for a policy framework to motivate measures to be taken on a local scale is expressed. Today, the Dutch Ministry of Environment still initiates the development of the policy regarding airborne PM. A far-going decentralisation of responsibility (from national to provincial or local level), being characteristic for other topics in the field of air quality, is not anticipated. Also, policy makers

interviewed expect that the future EU PM_{10} directive will not be met on a local scale. As in the past failures like this do not lead to sanctions at whatever authority level.

SAMENVATTING

Een van de belangrijkste bronnen van fijn stof in een stedelijke omgeving is het verkeer. Deze bron stoot grote hoeveelheden kleine stofdeeltjes uit die voornamelijk bestaan uit roet en organisch koolstof, en gassen die als voorlopers kunnen optreden voor de vorming van nieuwe deeltjes of een bijdrage leveren aan de groei van bestaande deeltjes. Het aandeel zeer kleine deeltjes in uitlaatgassen is relatief zeer groot en kent een groot aandeel aan chemische stoffen (Policyclische Aromatische Koolwaterstoffen (PAK's) en toxische metalen) die mogelijk nadelig zijn voor de gezondheid. Er zijn echter nog betrekkelijk weinig gegevens bekend omtrent de grootte van emissies van het fijn stof afkomstig van het verkeer en de mechanismen die de gezondheidsproblemen veroorzaken. De invloed van het verkeer op de aanwezigheid van fijn stof in de lucht binnen stedelijke agglomeraties en langs drukke snelwegen, oorzaak van de gezondheidseffecten en de mogelijke inzet van schoon (elektrisch aangedreven) vervoer is het onderwerp van deze studie.

Het werk in 2000 was daartoe in een viertal onderdelen gesplitst:

a) De zorg om de effecten op de menselijke gezondheid is de aanleiding voor het huidige en toekomstige onderzoek op dit terrein (en de bijbehorende financiering). In een studie op basis van beschikbare literatuur worden de relaties tussen enerzijds de negatieve effecten op de gezondheid en anderzijds de fysische en chemische eigenschappen van fijn stof in stadslucht geinventariseerd.
b) Uitbreiding en verbetering van bestaande meettechnieken die ingezet kunnen worden in uit te voeren meetcampagnes.
c) Uitvoering en beschrijving van de resultaten van een tweetal meetcampagnes in een stedelijke omgeving en langs een snelweg. In deze experimenten zijn aantallen deeltjes en massa op vaste locaties of al rijdende geregistreerd. De bedoeling is vast te stellen op welke plaatsen in een stad de massa en aantallen deeltjes het hoogst zijn en mogelijk een gevaar opleveren voor de directe omgeving.
d) Samen met de Unit Beleidsstudies is middels interviews geïnventariseerd in hoeverre beleidsmakers van diverse overheden (nationaal, provinciaal, gemeentelijk) werken aan de oplossing van het optreden van luchtverontreiniging in het algemeen en het probleem van het fijn stof in stadslucht in het bijzonder. Apart gespreksonderwerp hierbij was hun opvatting ten aanzien van de inzet van schone elektrisch aangedreven transportmiddelen.

De twee belangrijkste vragen die de aanleiding vormden voor de literatuurstudie waren:

1. welke deeltjes eigenschappen indiceren het sterkst de kans op optreden van negatieve gezondheidseffecten?
2. is verkeer een belangrijke bron van deeltjes met deze eigenschappen?

Uit het literatuuronderzoek komt naar voren dat fysische eigenschappen als (aard van het) oppervlak, de (on-)oplosbaarheid en mogelijk de aantallen de voornaamste indicatoren zijn. De toepassing van PM_{10} en $PM_{2.5}$ als indicator (zoals in de huidige EU richtlijn) is minder geschikt omdat er geen causaal verband bestaat met de gezondheidseffecten. Chemische eigenschappen die een rol spelen zijn is de aanwezigheid van metalen, roet, PAK's, endotoxines en zuren. Duidelijk is dat een combinatie van bovengenoemde fysische en chemische eigenschappen het grootste gevaar vormen. Zo blijkt bij ultrafijne deeltjes (diameter kleiner dan 0.1 μm) dat toxische stoffen zich kunnen hechten aan de niet oplosbare kern van deze deeltjes. Daar komt bij dat deze deeltjes tot in de longblaasjes kunnen doordringen waar de gebruikelijke verwijderingmechanismen niet in staat zijn deze te verwijderen omdat ze te klein zijn. Uit de bestudering van de samenstelling van de verkeersemissies blijkt dat deze vooral bovengenoemde chemische stoffen bevatten (black carbon, PAK's en metalen) en deze deeltjes uitstoten in de fijne en ultrafijne mode. Opgemerkt moet wel worden dat de studie naar de oorzaken nog in volle gang is en er nog vele vraagtekens zijn. Samenvattend kan als belangrijkste oorzaak gekenmerkt worden de verhoogde aanwezigheid van toxische,

onoplosbare componenten (roet, PAK's en metalen) die vooral bij de kleinere deeltjes (diameter minder dan 1 μm) lijken voor te komen en die door vooral het verkeer worden uitgestoten.

Naar aanleiding van de uitkomst van de literatuurstudie is een (data-)onderzoek verricht naar de invloed van verkeersemissies op de toxiciteit van het fijne stof in lucht. Deze studie is gebaseerd op verschillen in chemische samenstelling, deeltjesgrootte, massa en aantallen zoals gevonden langs een snelweg (A9) en vergeleken met achtergrond. De belangrijkste vooralsnog kwalitatieve constatering is dat in door verkeersemissies verontreinigde lucht de toxiciteit per $\mu g/m^3$ fijn stof hoger ligt dan in de achtergrond. In achtergrondaërosol zijn lood en zink de belangrijkste metalen. De bijdrage van het verkeer bestaat voornamelijk uit zink, lood en koper. De concentraties in de lucht zijn benedenwinds van de weg hoger (hetgeen ook de verwachting was). Dit onderzoek wordt vervolgd in 2001.

Een ander belangrijk wapenfeit is de studie naar massa en aantallen met behulp van een rijdende meetwagen. Het blijkt dat langs het traject Petten-Amsterdam concentraties geleidelijk toenemen bij het naderen van de stedelijke agglomeratie van Amsterdam. Binnen Amsterdam worden de hoogste aantallen en massa aangetroffen op de ringweg (A10), nabij kruispunten. Extreem hoge aantallen zijn te vinden in tunnels, iets dat zou moeten worden meegenomen in de discussie over het aanbrengen van overkappingen en bebouwing boven snelwegen. Deze toegepaste 'mobiele' meetmethode geeft de mogelijkheid om tegen relatief lage kosten snel een overzicht te verkrijgen van die locaties in een stad waar de hoogste concentraties van fijn stof zich voordoen. Het vormt aldus een aanvulling op het vaste meetnet in een stad waarvan de meetapparatuur doorgaans op plekken staat waar stadsachtergrond gemeten wordt.

Opbouw van gezamenlijke kennis vond plaats met de unit Beleidsstudies (Appendix). Geïnventariseerd is hoe de verschillende overheden omgaan met het fijn stof probleem en hoe zij denken dit op te lossen. Gesprekken zijn gevoerd met verantwoordelijke beleidsmakers van VROM, provincies, en enkele grote steden. De belangrijkste bevindingen zijn dat

i) het kennisniveau bij met name de stedelijke overheid omtrent de fijn stof problematiek (nog) betrekkelijk gering is,
ii) dat VROM voorlopig het voortouw wil houden in de aansturing met het oog op de herziening van de Europese richtlijn in 2003, en
iii) dat alternatief vervoer door de verschillende overheden vooralsnog alleen als lange-termijn oplossing gezien wordt (mogelijk toepasbaar *na 2010*). Op de korte termijn verwacht men meer heil van schone dieseltechnologie. Men ziet nauwelijks mogelijkheden om grootschalige implementatie van alternatief vervoer te versnellen met behulp van beleidsmaatregelen.

ABBREVIATIONS

PAHs	polycyclic aromatic hydrocarbons
PM	particulate matter
$PM_{0.1}$	particle mass fraction smaller than 0.1 μm: ultrafine particles
$PM_{2.5}$	particle mass fraction smaller than 2.5 μm: fine particles
PM_{10}	particle mass fraction smaller than 10 μm: inhalable particles
$PM_{10-1.5}$	particle mass fraction between 2.5 and 10 μm: coarse particles
TSP	total suspended particulate
UFP	ultrafine particles

1. INTRODUCTION

At the department of Air Research and Technology studies are conducted in order to characterise ambient particulate matter (PM) levels in the Netherlands and to assess the contribution of traffic to these levels. The presence of PM in outdoor air causes pollution and human health problems. In this introduction a short overview on the backgrounds of the problem is given.

1.1 Past

Though air pollution dates back in the Middle Ages, the recent history regarding PM starts in London in the 1950s, at the time of the so-called "Big Smoke". During these events a growth in the number of death occurred especially among the elderly and people suffering from asthma and bronchitis. It was indicated that the origin lay in soot particles that were the result of the huge emissions of SO_2 due to the burning of coal for house heating and industry. Since then, scientists and policy makers gradually became aware of the seriousness of the problem. Governmental measures concerning PM and other pollutants were taken. These were (and still are) ultimately directed to the reduction and prevention of acute human health problems. In the Netherlands as well as other European countries one started to work with limit air concentration values based on mean daily and annual values as well as 'smog alarm phases' based on actual hourly concentrations in air (Kroon, 2000).

1.2 Present

Various studies in the USA and Europe have shown that a relationship exists between PM_{10} in air and certain health effects. Epidemiological studies revealed that acute health effects especially occur in the risk group of those elderly already having problems with the airway system or cardiovascular system. The life shortening probably is a few weeks for these groups. However, chronic effects due to long exposure to high concentrations of PM may be far more serious and may lead to a life shortening of a couple of years.

The University of Wageningen has investigated if and to what extent the lung system of children was affected when visiting a school not far from a busy motorway. The measured air pollution levels in the area were increased when the location of the school was closer to the motorway and when traffic intensity was higher. A relationship was found between air pollution levels (especially originating from heavy traffic) and typical lung systems complications, especially with children already being more sensitive to these kind of airways health problems. It was found that children being exposed most to the prevailing air pollution showed twice as much problems. Also, the percentage of children already possessing allergic antibodies or an increased lung irritability was considerable, namely 40% (VROM, 1999).

Due to these findings current policy has become directed to the prevention of the (long-term) health effects. However, it has not been possible yet to determine a lower threshold level below which no adverse health effects are measured; even at very low PM levels health effects are still observed. It is therefore very difficult to define directives that decrease the risk for health effects to zero. Other complicating issues are the time scale over which regulating measures can become into effect as well as the far-going economical consequences of measures for various emission sources.

In 1997 the primary PM emission to air in the Netherlands was 40 kt. The most important anthropogenic sources of PM in the Netherlands are traffic, industry, households and refineries. Secondary aerosol is produced by SO_2, NO_x, VOS en NH_3. Natural sources of PM are sea salt and soil dust. In figure 1.1 the emission distribution has been displayed for the various sources.

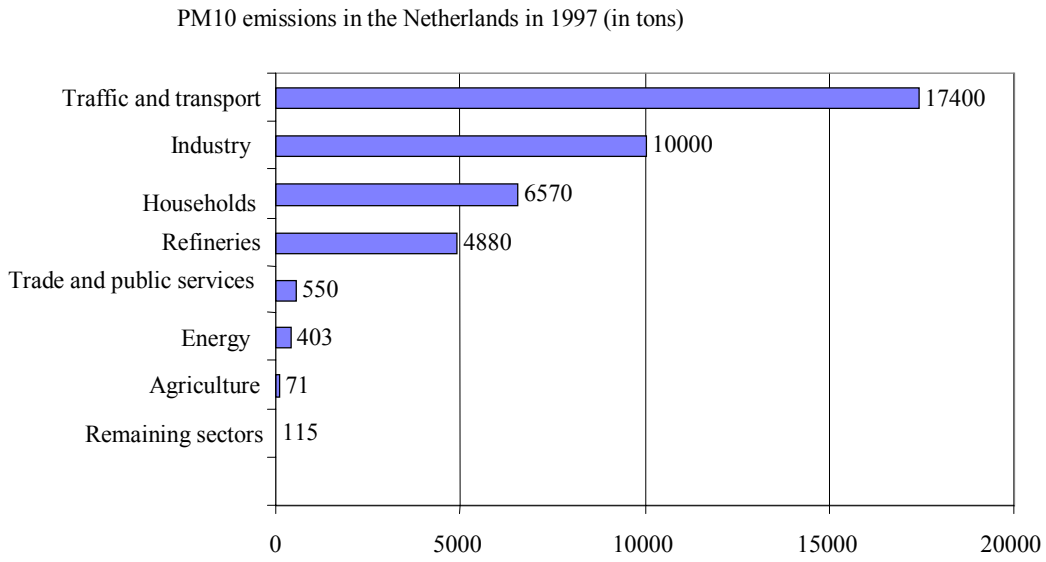

Figure 1.1 *Primary PM emissions in the Netherlands in 1997*

Clearly traffic and the transport sector (heavy trucks and inland shipment) have the largest emissions of PM_{10}.

Another complicating problem is that the exact physiological mechanism responsible for the health effects in human beings is not exactly known. In the past much attention has been paid to the relation between mass of the inhaled particles and effects. Nowadays, one stresses possible relations with numbers, size and chemical composition of the particles. The diameter of a particle determines the location where it deposits in the lungs. Therefore a growing attention exists for the smaller-sized mass fractions (like PM_1 and $PM_{0.1}$) being characterised by very large numbers but hardly adding up anything to the mass concentration. Furthermore, the impression exists that the particles of anthropogenic origin (soot, secondary aerosols) are mostly found in such smaller mass fractions while larger particles have a natural origin. In general, due to these considerations scientific as well as political attention has shifted gradually from the PM_{10} mass fraction to $PM_{2.5}$ or fractions with even smaller sizes (Krijgsheld, 1999).

1.3 Future

The development of effective control strategies in the near future requires a better understanding of the properties of PM but large gaps still exist in our understanding. These gaps are due to the fact that PM is a complex mixture of multi-component particles whose size distribution, composition, and morphology can vary significantly in space and time. Atmospheric aerosol particles vary in size (from a few nanometers to tens of micrometers). Major components include sulfate, nitrate, ammonium, organic material, elemental carbon (or soot), trace elements (including toxic and transition metals) and crustal components. PM is emitted directly from sources such as diesel and gasoline engines and is also formed in the atmosphere from gaseous precursors.

It is important to notice that serious doubt exists on the applicability of the current PM_{10} EU-guideline of 40 $\mu g/m^3$ which is to be reviewed in 2003. First, PM_{10} concentrations hardly show any variation over the Netherlands. Secondly, measurements indicate that the fraction between PM_{10} and $PM_{2.5}$ consists mainly of soil dust, seasalt, etc.. But perhaps most important is the revelation due to German and America research that (statistical) relationships exist between chemical composition and/or the number of ultrafine particles and the frequency of health effects in cities. This finding favors the introduction of a guideline on $PM_{2.5}$ which implies a

substantially different abatement strategy with, for example, introduction of clean electrically driven vehicles in city areas.

The comprehension of the PM characteristics and its sources requires a high-quality execution of experimental measurements. Standard PM measurement techniques like the collection of PM only partly fulfill this desire due to the prerequisite long-term measuring periods in order to collect enough matter on the filter, the burden of huge number of analyses and possible sampling artifacts. The difficulty and cost of such PM measurements are hindering the characterization of temporal and spatial variability, the proper understanding of the processes that control their formation and removal, and the quantification of the exposure of populations to them. In order to overcome these difficulties the evaluation of existing PM measurement methodologies and development of new technologies is required, followed by appropriate experiments which ultimately will allow for an investigation of the causal relationships that are necessary to develop (cost-)effective abatement measures.

One of the major sources of PM in urban environment is the vehicular traffic. The mobile sources emit large quantities of primary particles, composed mostly of soot and organic material, as well as gases that can act as precursors for new particle formation or can assist the growth of existing particles. The emitted PM has a larger proportion of ultra-fine particles and contains a number of other possible causative agents, such as policyclic aromatic hydrocarbons (PAH) and toxic metals. There are, unfortunately, still few data on the emission factors for these PM characteristics. The influence of traffic on the presence of PM in air within large urban agglomerations and along busy motorways is the major issue of this study.

2. LITERATURE STUDY ON THE INFLUENCE OF PARTICULATE MATTER ON HEALTH PROBLEMS IN LARGE CITIES[1]

2.1 Introduction

The possible role of traffic emissions on the occurrence of adverse health problems in large cities can only be determined when the underlying mechanisms are clear. To this purpose a literature study is performed to find answers to two questions:

i) Which properties of particulate matter may explain the adverse effects on human health?
ii) To what extent do traffic emissions affect the properties to be found relevant?

It is notable that a third question has come up only recently, namely:

iii) is PM in urban air solely responsible for the negative effects or is it the combination with other air pollutants?

In a separate section (2.5) a review of literature on this last subject is presented. Obviously, this study is the result of knowledge that is currently available in the open literature.

The mass of particles with an (aerodynamic) diameter smaller than 10 μm is denoted by PM_{10}. The EU uses PM_{10} as an indicator for limit values to be imposed on ambient concentrations in air. These constraints on PM_{10} were defined when it became clear that adverse effects on human health are occurring (for more details see also Chapter 1: Introduction). In the United States, however, a limit value is now imposed on the $PM_{2.5}$ mass fraction because it is believed to be a better indicator for the negative effects than PM_{10}. Not surprisingly, this discussion continues and constraints on the PM_1 fraction or $PM_{0.1}$ (i.e., the 'ultrafine' fraction) are now being considered. The reason for this continuing discussion is that the underlying biological mechanisms are not understood. The *statistical* relationships between the PM mass indicators and health effects are apparent but a clear *causal* explanation for the adverse effects is missing. Such mechanisms may be specific for an aerosol of a certain diameter (range) and/or chemical composition and are not, or only partially, related causally to mass fractions like PM_{10} or $PM_{2.5}$.

There are two methods to find a possible relationship between health damage and traffic-related PM. The first method is a physical and chemical identification of PM in urban air. Statistical relationships are normally calculated for various physical parameters of the aerosol (diameter, number, surface and mass). The chemical composition of the aerosol can be classified into several elements being toxic to a varying degree. Combining such an identification with knowledge of physical and chemical content of traffic emissions with the presence of the (toxic) compounds in aerosols, may lead to a better understanding about traffic related health effects.

The second method is to characterise directly the health effects due to the emissions of traffic, thus without a physical, chemical and toxicological characterisation of the aerosols. This approach is used because a complete determination of the aerosol parameters is very laborious. A number of toxicological and epidemiological studies determine directly the effect of sources (instead of the effects of aerosols); traffic is an important source in these studies.

The remainder of this chapter is organised as follows: section 2.2 describes the various physical parameters (diameter, number, surface and mass) of aerosols. In section 2.3 a classification of aerosols is made according to their chemical composition (inorganic compounds, carbonaceous compounds and metals). In section 2.4 an inventory of health effects that are possibly related to

[1] This chapter is a summary of an internal report written by A. Even (2000).

traffic emissions is given; this is followed by a discussion of the significance of size and chemical composition of aerosols in this respect. Finally, section 2.5 summarises the present discussion whether adverse health effects are due to the presence of the particulate matter in the air or are due to the total mixture of air-pollutants.

2.2 Physical parameters

The biological mechanisms that may explain the health effects indicate the importance of other aerosol parameters than the mass fraction indicator, i.e. size, number and surface. A relationship between health effects and mass can not be translated simply into a relationship with aerosol number or surface. In figure 2.1 it is shown that in urban air the largest numbers occur for particles smaller than 0.1 μm, having a maximum at 0.05 μm; particles having relatively large surfaces (with respect to their diameter) are found in the size range between 0.02 μm and 1 μm.

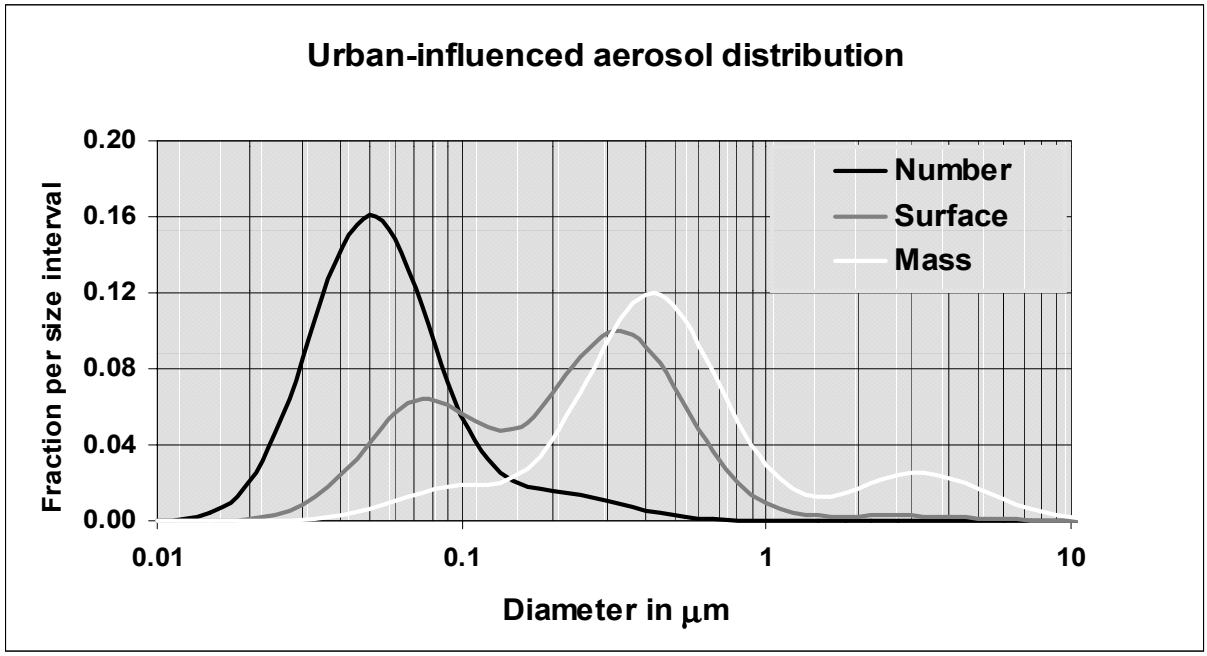

Figure 2.1 *Number, surface and mass distribution of urban air particles*

Only particles with diameters above 0.2 μm contribute substantially to the mass parameter. Due to these properties the relationship with health effects have to be established for each separate parameter. Complicating aspect in this respect is the solubility that is closely linked to both the particle's size and surface. In the next subsections the various parameters are discussed, starting with aerosol size.

2.2.1 Size distribution

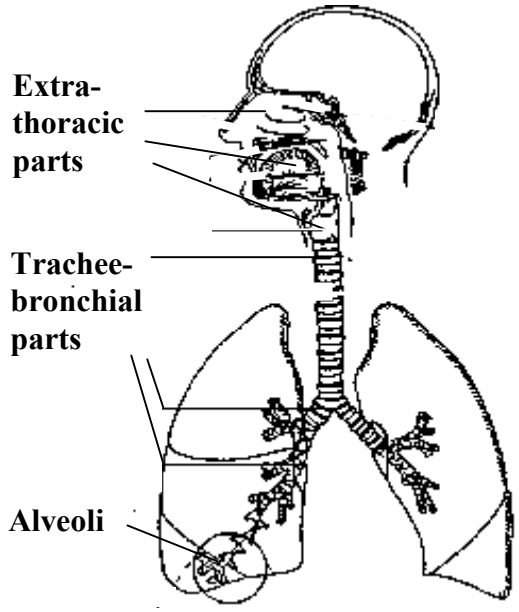

Figure 2.2 *The respiratory system*

Particle size determines whether a particle can be inhaled and in which part of the lung it can be deposited. Particles can be inhaled when they are smaller than 10 μm. Coarse aerosol and part of the fine aerosol (0.5 - 2.5 μm) are deposited in the extra-thoracic and the trachea-bronchial parts (see figure 2.2). The mechanisms are impaction, settling and sedimentation. Particles smaller than 1 μm penetrate into the pulmonary alveoli. Deposition by diffusion causes the particles smaller than 0.2 μm to deposit in the alveoli. For sizes between 0.2 and 0.5 μm, the sum of the results of the various deposition mechanisms have a minimum and a large number leaves the lung again. Combined with the distributions in figure 2.1 it is concluded that most of the mass is deposited in the upper airways, whereas the highest numbers deposit in the alveoli. No indications have been found yet that one size fraction is more important than another in the origin of health effects (Van Bree and Cassee, 1999).

When aerosol particles reach the alveoli they may be removed by dissolution. However, insoluble compounds are removed by uptake (endocytosis) by cells present in the alveoli that transport the particulate matter to other parts in the body for excretion (macrophages). The ultrafine particles evade this removal mechanism because they are so small that they end up in the interstitial spaces of the alveolar lung tissue where they can not be reached by the macrophages. This is important as in outdoor air the insoluble part of aerosols is large for the ultrafine and coarse fraction.

2.2.2 Number

The number size-distribution obtains its maximum in the ultrafine fraction (figure 2.1). These particles might attribute to the health effects due to their following properties:

1) Small enough to be taken up in the vascular system (EPA, 2000).
2) Small enough to reach the interstitial spaces of the lung tissue thereby evading clearance by macrophages (see earlier remark in subsection 2.2.1) (Oberdörster, 1992a,b).
3) High deposition efficiency in the alveoli and extra-thoracic airways, the vulnerable parts of the airways (EPA, 2000).
4) Large surface-to-diameter ratio such that toxic compounds can be released quickly (Diabaté, 2000).

Apart from these theoretical considerations, toxicological and epidemiological studies have shown statistical correlation between health effects and number. For example, in an epidemiological study by Peters et al. (1997) it was found that the number of ultrafine particles is a better indicator than particle mass. In a toxicological study, Oberdörster et al. (1992a)

showed the negative impact of ultrafine particles with non-toxic, insoluble compounds. On the other hand, Churg and Brauer (1997), found that only 5% of the number of particles in human lungs appeared to be ultrafine and that 96% was smaller than 2.5 μm. Apparently, ultrafine particles are removed or dissolved after a certain time. This is consistent with the finding by Oberdörster (2000) that a large part of the inhaled ultrafines turns up in other parts of the body.

2.2.3 Surface

As can be seen in figure 2.1, about one half of the particles in the surface-size distribution is in the ultrafine fraction; a relatively large proportion of these particles is insoluble. The other half has diameters between 0.2 and 1 μm. A larger surface tends to increase the particle's toxicity, for example, Oberdörster et al. (1992b), exposing rats to insoluble, moderately toxic aerosols, found an explicit correlation with surface magnitude for those particles that are small enough to enter the interstitial lung-tissue. However, for another toxic compound, PTFE, the toxicity showed an increase with decreasing size of the administered particles, (Oberdörster, 1995). It is important to note that these findings are only observed for ultrafine particles.

The statistical correlation with a physical parameter results from the combination of this physical property with the chemical composition. Because of the large contribution of ultrafine aerosols to surface magnitude of PM, their chemical composition is of special importance (for more information see chapter 2.3).

2.2.4 Mass

In the period 1996-1999, nineteen epidemiological studies reported a correlation of increased risk for premature death with mass (a/o. Kelsall et al. (1997); Burnett et al. (1998); Borja-Alburto et al. (1997); Loomis et al. (1999); Morgan et al. (1998b)). The risk on premature death is expressed as a relative risk, i.e. the increase in risk (in percents) for a certain increase in particle mass. The increase in risk is usually a few percents per 10 $\mu g/m^3$.

Reported relative risks vary with place and season. For the Netherlands, a much smaller risk is found than in the USA: a 2% increase per 100 $\mu g/m^3$ (Hoek et al., 1997). A possible explanation might be the seasonal variation (see below). It is emphasised that the death do not only occur among the elderly and diseased, but also concern very young and even unborn children causing a large decrease of life years (Brunekreef, 2000). Apart from studies showing a significant increased risk, there are also a (much smaller) number of studies that do not find a significant increase (Gamble, 1998; Levy, 1998; Pereira et al., 1998; Lee et al., 1999). The latter two are multi-pollutant studies.

In a number of studies a seasonal variation in relative risk is found (Moolgavkar en Leubeck, 1996; Anderson et al. ,1996; Hoek et al., 1997; Michelozzi, 1998). The risk is always largest in summer. Reasons for this are diverse: seasonal variation in composition, increased exposure indoors due to open windows, seasonal variation of co-pollutants and an increased mean temperature. The lower relative risk in the Netherlands compared to the studies in the USA may also be explained by a different annual variation of the ambient PM concentrations. The PM concentrations in the USA typically increase in summer, thereby contributing largely to the annual average of the relative risk; in the Netherlands, however, high levels occur in winter (Rombout et al., 2000).

Epidemiological studies correlate air pollution with acute health effects. In cohort studies individuals are followed for a longer period of time to observe possible long-term effects. In the cohort study of Abbey et al. (1999) an increased risk for PM_{10} was measured as a function of the frequency that the PM-level exceeded 100 $\mu g/m^3$. The population examined was divided into subgroups defined by age, socio-economic index and smoking. For all subgroups, a significant

increase in the relative risk was observed when the level of 100 μg/m^3 for (outdoor) PM concentrations was exceeded more often. Another long-term study describes the effect for groups that are exposed to small differences in PM (Brunekreef, 1997); it was calculated that the average life expectancy diminished with more than one year while for the people that actually died it meant a shortage of more than ten 10 years!

Finalising, though epidemiological and cohort studies do not produce any indication of size-fraction and/or typical chemical component that might explain the adverse effects on health, it can be concluded that the outdoor aerosol results in an enlarged risk on premature death when people are exposed to current levels.

2.2.5 Conclusions

Based on epidemiological and cohort studies particulate matter in air appears to have an adverse effect on human health. The aerosols in outdoor air result in an enlarged risk on premature death when humans are exposed at the present levels. Unfortunately, the underlying causal relationship(s) is (are) not known yet with certainty and therefore highly speculative. Properties mentioned in literature are mass, size, number, surface and solubility.

Particle mass, when expressed as PM_{10} or $PM_{2.5}$, is not a very suitable indicator because these fractions include particles of a very different physical nature and chemical content. On the other hand, a strong statistical correlation has been found with particle numbers. The magnitude of the surface correlates fairly strong with health effects for insoluble particles within the ultrafine size range.

In this respect, it may be a combination of surface magnitude, chemical composition and solubility that determine the toxicity of the particle, and particle size, determining the place of deposition, that may be held responsible. And if this were true for a single particle, it will be definitely valid for a large number of those particles, increasing the correlation when numbers increase.

2.3 The Chemical Composition

2.3.1 Introduction

The composition of outdoor particles is divided into three major classes:

1. inorganic compounds (acids, sulphates, nitrates) carbonaceous compounds (soot, policyclic aromatic hydrocarbons (PAHs, endotoxines) belong in this class
2. metals

Toxic effects of an aerosol partly depends on its size fraction. The major mass fraction of the inorganic compounds is present in the fine aerosol fraction; in the case of nitrate it is also partly present in the coarse fraction. The different carbonaceous compounds occur in the ultrafine, the fine and the coarse size fractions. Metals are mostly present in the fine and ultrafine fraction.

Another important difference between these three classes of compounds is the spatial variation in concentration. Inorganic compounds are produced by secondary production mechanisms and are more or less uniformly distributed over large distances (a few hundreds of kilometres). The carbonaceous particles, on the other hand, are largely emitted as primary particles; concentrations of these particles are increased in their source areas and in cities.

2.3.2 Inorganic compounds

The most important inorganic compounds are sulphate, nitrate, ammonium and acids. Research has been carried out for several years on the toxicity of acids. In a report of the EPA (1996a) and confirmed by Abbey et al. (1999) it is concluded that the contribution of sulphates and acids to the toxicity of outdoor particles is small. For example, Frampton et al. (1992) found no or only small adverse health effects of sulphates and acids even at high concentration levels; the small effect is attributed to acidity. One has to keep in mind that acid is not the direct cause for the effects but influences the defence mechanisms against other perpetrators; for example, defence mechanisms fighting bacterial lung infections are influenced by the presence of acids. Also, the presence of acids may intensify the damage caused by ozone. Recent research has reported small effects at specific sites (Burnett et al. 1998, Fairley 1999). The study of Fairley et al. (1999) found an effect of nitrate on mortality. In summary, inorganic compounds may attribute to the health effects, but in general this is not the case.

2.3.3 Carbonaceous compounds

Four groups of carbonaceous compounds can be discerned:

1. *black carbon* (BC), also called elementary carbon (EC) or soot, associated to diesel emissions. and generally considered to be a toxic fraction in PM
2. *policyclic aromatic hydrocarbons* (PAHs), whose presence can be related to black carbon, but semi-volatile and may adsorb at particles of all sizes, hence, not only present in the small particle fraction in which they are originally emitted
3. *acids*, present in the water-soluble fraction of carbonaceous compounds, having a small toxic effect
4. *endotoxines*, a group of compounds showing similarities with the toxic compound lipopolysacharide (LPS).

Pedersen et al. (1999) examined the toxicity of the total carbonaceous particle fraction at different places and in different seasons as a function of the organic carbon content of the atmosphere. They concluded that the toxicity of the urban organic aerosol is higher due to higher prevailing concentrations. They further reported a seasonal variation in the toxicity per μg organic carbon which may be due to a difference in chemical composition. Also, one has to bear in mind that there is no causal relationship between one μg organic aerosol matter and adverse health effects as the organic fraction consists of hundreds of different compounds that are toxic to a different degree and are present in particles of different sizes.

The complementary part of the organic fraction in carbonaceous aerosol is *black carbon*. In general, BC causes acute and chronic effects. The relative risk on premature death (see section 2.2.4) increases with 1% per 1 $\mu g/m^3$ black carbon (Sunyer et al., 1991). This is 5-10 times larger than the relative risk of PM_{10}. An important property of black carbon is its presence in small aerosol fractions and its related large surface area which both enhance its toxicity. An illustration of this aspect is the observation of large inflammations in rats when exposed to ultrafine black carbon (20 nm), whereas no reaction was observed after exposure to fine black carbon (200 nm) (Li et al. 1996, 1997).

The second group, the policyclic aromatic hydrocarbons, has a similar (main) source as black carbon, i.e. traffic. It has been extensively shown that PAHs are carcinogenic and genotoxic (De Raat, 1994) and the EU is preparing specific legislation on this subject. The size distribution of PAHs depends on the distance travelled after emission and molecular weight; this affects its toxicity. In general, the freshly emitted PAHs are associated with ultrafine aerosol at emission, but the more volatile they are, the quicker they redistribute to the fine and also to the coarse aerosol fractions. Because PAHs can be present in all size fractions, they can end up in all parts of the airways.

Endotoxines are harmful carbonaceous compounds of a biologic origin. Sources of endotoxines are mainly bacterial activities. As an example, pig farmers may be exposed to LPS that causes adverse health effects after inhalation (Michel et al. 1997). Concentrations were high in this case, but also at lower concentrations endotoxines may cause damage (Rose et al. 1998). Bonner et al. (1998) showed that endotoxines are partially responsible for the harmful effects of PM_{10} at macrophages. Becker and Mohn (1998) suggest that endotoxine may play a role in the potential of coarse aerosol to induce adverse effects.

In general, many authors (Godleski et al. 1996, 1997; Gordon et al., 1998; Watkinson et al. 1998; Killingsworth et al., 1997; Rombout et al., 2000; Bree and Cassee, 2000) confirm the importance of the carbonaceous compounds in particles above that of the inorganic compounds. The presence of two toxic groups in the carbonaceous compounds particles being emitted by traffic, black carbon and PAHs, is reason to believe that traffic emissions may play an important role in inducing the adverse health effects.

2.3.4 Metals

Metals are the last group of important toxic compounds. In the case of lead the EU has already imposed regulation. Besides lead, the group of transition metals may be important: these catalyse the production of reactive oxygen species (ROS). In the last EPA overview (1996) it was concluded that, though the metals arsenic, cadmium, copper, vanadium, iron and zinc play an important role in occupational health, they are not harmful at outdoor concentrations (1-14 $\mu g/m^3$). Recently, however, toxicological and epidemiological indications have been found that metals can cause adverse effects at outdoor concentrations (Tsuchiyama et al. 1997, Osman et al. 1998, Lay et al. 1998). Also, ultrafine particles collected through autopsy from human lungs were shown to consist mainly of metals (Churg et al. 1997). According to toxicological studies, the metals present in urban aerosol contribute significantly to two biological mechanisms that cause damage in the alveoli (Goldsmith et al. 1998). These two mechanisms are production of ROS and interaction with macrophages (cytokine production), which were also shown to be important effects of PM_{10} (Li et al. 1996, 1997).

Kodvanti et al. (1997) examined the toxicological importance of these metals. It appears that effects of exposure to nickel can only be measured after a number of days. But at that time it is the most toxic transition metal. Kennedy et al. (1998) showed the effects of copper at outdoor concentrations and even reached the conclusion that the toxicity of copper explains the toxicity of the total outdoor aerosol in places with strong local sources. Finally, Dusseldorp et al. (1995) observed negative effects of the exposure to iron particles. Interestingly, sulfate may play a role in connecting the generation of ROS by functioning as a ligand for particle associated iron. Hence, sulfate does not act as a direct toxicant but facilitates the toxicity of reactive metal constituents (Ghio et al. 1999).

2.3.5 Conclusions

The ambient particulate mass can be divided into three groups of chemical compounds: inorganic compounds, carbonaceous compounds and metals. Health effects of the inorganic compounds are small, though acids may interfere with the defence mechanisms of the lungs. Though effects of particulate matter show spatial and seasonal variation, the two remaining groups were found to be related to health effects.

The first group are the carbonaceous compounds, especially those due to the primary emitted combustion aerosol containing BC and PAHs. The presence of these two toxic compounds in particulate traffic emissions point to the important role of traffic with respect to the adverse

health effects of particulate matter. Apart from this carbonaceous aerosol fraction, a biogenic fraction may possibly be important: the endotoxines.

The second important group are the metals. Though it is not clear which specific metals contribute most to health effects, there are indications that metals might explain toxicological effects of particulate matter.

2.4 Traffic emissions

It is expected that (health) problems resulting from traffic emissions are largest in cities. This is supported by measurements of Morawska et al. (1999b). They measured the size-distributions of aerosol at different locations and found a distinct size-distribution at every location. Only the size distributions of urban and traffic-influenced aerosol resembled each other. Furthermore, Janssen et al. (1999) report a high correlation between the degree of urbanisation in an area with typical traffic indicators like NO_2, soot and benzene; it was remarkable that no high correlation with $PM_{2.5}$ was found.

Traffic emissions do not only contribute to outdoor PM levels, but also to the indoor levels. Because the PM in traffic emissions is in the (ultra)fine particle range, it may efficiently penetrate buildings via open windows, cracks and meshes and may even penetrate mechanic filter systems (Morawska, 1997). EPA (2000), for instance, has found that 30% of the indoor particulate originates from diesel particles.

A number of studies is conducted on the effects of total traffic emissions. One of these studies dealt with the relation between the lung-function of children and traffic-emitted particles in The Netherlands. The conclusion was that traffic-emitted particles have a significant adverse effect (Brunekreef et al. 1997, Van Vliet et al. 1997). In a later study by Van Vliet et al. (1999) it was shown that this relation was due to the emissions of heavy-duty traffic alone.

Other research confirms the picture that PM emitted by (heavy-duty) traffic has adverse effects on airways and the immune system and are potentially carcinogenic (Ye et al., 1999). Pope et al. (1999) attribute the largest part of the effects to mobile sources. When factor analysis is applied usually one factor is related to traffic (e.g., Burnett et al., 1998). As an example, Özkaynak et al. (1996) found a significant traffic-related factor for both total premature mortality and premature cancer, cardio-vascular, respiratory and pneumonia deaths.

These findings are supported by toxicological measurements on mutagenity of outdoor aerosol. The results of these measurements showed that the mutagenity of outdoor particles was 1.5-2 times larger per m^3 in urban areas than in rural areas (Pedersen, 1999).In the densely populated urban areas the contribution of traffic emissions is larger than in rural areas which suggests that the larger toxicity is caused by traffic-emitted particles.

2.4.1 Particulate traffic emissions

Physical characterisation of particles emitted by traffic produces an unambiguous picture. The freshly emitted particles are small, with a mass-median diameter of 0.15 – 0.30 μm (Morawska et al. 1999[a], Weingartner et al. 1997) and a number median of 0.02 – 0.07 μm (Maricq et al. 1999, Morawska et al. 1999[b], Weingartner et al. 1997, Yin and Harrison 1999). A small fraction of the total emissions is in the coarse mode which is generally less than 30% (Weingartner et al. 1997). Hence, a large part (number) of the emitted particles is in the ultrafine fraction and will deposit in the vulnerable parts of the airways.

The chemical composition consists mainly of carbonaceous compounds and metals. Organic and elementary carbon make up more than half of the emitted mass (Kirchstetter et al. 1999); PAHs contribute about 1% (Weingartner, 1997). Vermeulen et al. (1999) measured a significant contribution of PAHs to traffic emissions as well as from metals (Cr, Cu, Zn, Cd and Pb). Both

were found in the smallest size fraction (<0.5 μm). The compounds present in aerosol fractions have a small mass-median and a relatively large surface enhancing their toxicity. From these two observations, it can be concluded that traffic aerosol contributes to a large extent to the toxicity of outdoor aerosol. Nikula et al. (1995) found (unspecified) toxic matter on diesel particles. Some of the toxic effects were not related to PAHs but to surface reactivity of the diesel particles. In general, the response to combustion derived particles correspond with levels of soluble compounds adsorbed on the carbonaceous cores of particles (Bree and Cassee, 2000).

To measure the presence of particles emitted by traffic a number of indicators can be used. Gaseous indicators are NO_2, NO_x and CO (Hitchins et al. 2000, Yin en Harrison 1999, Harrison et al. 1999, Väkevä et al. 1999). Indicators for the particulate phase are absorption of light, the particle number, benzo[a]pyrene and PAHs (Hitchins et al. 2000, Yin en Harrison 1999, Harrison et al. 1999, Väkevä et al. 1999, Fischer et al. 2000).

$PM_{2.5}$ and PM_{10} are explicitly not-useful indicators of traffic emissions. In a number of studies the distance up to which the influence of traffic emitted particles reaches was measured. Hitchins et al. (2000) measured an increase of a factor 7 of the particle number at 15 m from the road and still an increase of 3.5x at 150 m, for which the aerosol distribution did not change with respect to that at 15 m; Morawska et al. (1999a) measured similar ratios. Measuring around high buildings (Väkevä et al., 1999) revealed that the fine fraction behaves like gases and only show elevated concentrations at the first meters above roads.

2.4.2 Conclusions

Studies aimed at health effects of total emissions of pollutants by traffic show clear effects. But more important, the emitted particles consist of precisely those compounds that are toxic and possess a size distribution that enhances toxicity due to a relatively large surface area. Those compounds are soot, PAHs and metals in the ultrafine and fine aerosol fraction. These compounds are also indicators of traffic, just like NO_2, CO and the particle number. Traffic contributes to the toxicity of outdoor aerosol, in particular in urban areas and along roads. Especially at short distance of the road, the contribution can be large. $PM_{2.5}$ and PM_{10} are not considered as specific indicators of traffic emissions.

2.5 Particulate matter as part of the total air pollution mixture

The most important indications for the adverse effects of PM result from epidemiological studies. In these studies, variations in outdoor PM and variations in indicators for public health are correlated. The concentration of outdoor PM varies with meteorological conditions, like the concentrations of gaseous pollutants. The measured effects of PM can therefore also be correlated with the entire pollutant mixture.

In so-called multi-pollutant studies the following pollutants are mentioned that partly explain the adverse health effects: PM (either $PM_{2.5}$, PM_{10} or TSP), NO_2 or NO_x, SO_2, O_3 and CO. In table 2.1 an overview is given of the number of multi-pollutant studies in which a pollutant was significantly associated with adverse effects (Kelsal et al., 1997; Gamble et al., 1998; Fairley et al., 1999; Özkaynak et al., 1996; Burnett et al., 1998; Borja-Aburto et al., 1997; Loomis et al., 1999; Simpson et al., 1997; Morgan et al., 1998b; Lee et al., 1999). From this table it appears that the entire mixture of air pollutants plays a role.

The effect of the entire pollutant mixture is larger than the sum of particle and gas phase related effects. This can be explained by various mechanisms. First, O_3 stimulates the uptake of particles in airways (Churg et al., 1996). Second, there are two mechanisms in which particles stimulate the uptake of gases (Bree and Cassee, 2000): a) particles can be a carrier of gases, delivering gases deep in the lungs and, b) the lifetime of free radicals is increased from the interaction of oxidants (O_3 and NO_2) with biomolecules in the lung.

Component	Significant contribution (Number of studies)	No significant contribution (Number of studies)
$PM_{2.5}/PM_{10}$/TSP	6	2
NO_2/NO_x	3	2
SO_2	2	1
O_3	4	1
CO	3	0

Table 2.1 *Overview of multi-pollutant studies: number of studies in which a pollutant did or did not contribute significantly to the adverse health effects.*

On basis of table 2.1, it can be concluded that PM has an adverse effect on public health. Again, it is not clear which parameters might explain the causative biological reaction mechanism. There are several hints that they are important but not enough research has been carried out yet to make definite conclusions. Finally, gaseous pollutants also play a role alone and in combination with PM (Bolarin et al., 1997; Churg et al., 1996; Kleinman et al., 1996; Last et al., 1987). Also, the quality of the different studies that are referred to here has not been considered.

2.6 Concluding remarks

Physical and chemical characteristics suspected to contribute to the toxicity of particulate matter as mentioned in literature are summarised and compared with the current knowledge about emissions by vehicular traffic. The important conclusions of this literature study are:

- *The combination of particle size, surface magnitude and chemical composition determine the toxicity of a particle.*

This is substantiated by the following findings in literature:

- particles smaller than 0.1-0.2 μm, the 'ultrafine' fraction, deposit at vulnerable places in the lungs
- toxic insoluble compounds appear more frequently in the smaller particle-size ranges; hence, the toxic potential of such compounds increases with smaller particle-diameters.
- black carbon, PAHs, soluble transition metals and endotoxines are toxic compounds
- acids are not toxic but may interfere with lung mechanisms against air-pollutants and bacteria
- toxicity correlates with the surface of ultrafine insoluble particles. This may be is explained by reactive oxygen species (ROS) that are released by soluble transition metals present at insoluble carbonaceous particle cores
- inorganic compounds only show weak correlation with particle's toxicity
- particle mass, expressed in e.g. PM_{10} and $PM_{2.5}$, as well as number are not causally related to the particle's toxicity; they are containers of compounds with different toxicities. The toxicity measured for PM_{10} or $PM_{2.5}$ fractions varies with location and season

- *Vehicular traffic contributes to the particle's toxicity*

The main source of the primary carbonaceous compounds is traffic. These compounds are likely to play an important role in the toxicity of a particle and are believed to cause damage to public health. They are emitted in the fine particle fraction, where their toxicity is further enhanced by their presence in the (ultra)fine particle fractions. Apart from these compounds, the reactive oxygen species (ROS) are also associated with the emitted insoluble carbonaceous particle cores. Hence, traffic emits those compounds that are listed above as toxic. Although not causally related with the particle's toxicity, their number is an indicator of vehicular emissions.

Furthermore, the associated mass fractions are relatively small; hence, neither PM_{10} nor $PM_{2.5}$ are clearly related to the adverse health effects that ascribed to traffic-emitted particles.

- *Definite conclusions can not be drawn at the current state of research*

The above listed conclusions are drawn on basis of currently available knowledge. For most of the mentioned compounds the precise biological mechanisms that cause the health damage to the lungs are not known. Also, the dose-response relations and interactions between particulate pollutants are not determined yet for the mentioned compounds.

From the information gathered here it is clear that the particle's toxicity can not be accurately described by a single parameter like PM_{10} or $PM_{2.5}$. There are several other indicators derived from physical and chemical characteristics. For most of these indicators only limited information is available, and the collected indicators should be used with care. An important indicator of toxicity is the chemical compound (soot, PAHs and metals and to some extent acids) though its effect depends on where it is deposited in the airways and hence its size. The toxicity of these compounds depends on their solubility which becomes enhanced when they have a larger surface area. The smaller the particles, the larger the surface area. Also, another property of these smaller particles is that penetration into the lungs is deeper and might reach vulnerable parts, for example the alveoli.

Measurement of particle toxicity is complicated by the existence of so many indicators. The current instrumentation is designed for the measurement of physical and chemical characteristics of particles. These are measurements of size, number, mass and chemical characterisation. It is concluded here that useful measurements in future will involve particle size-distribution, the soluble fraction for this size-distribution and the chemical compounds soot, PAHs, metals and total organic compounds (preferably including the size-dependence). In future, analysis methods will be established to measure also the potential of reactive oxygen species (ROS).

It will be labour-intensive to measure all the mentioned indicators. An easier method of approach is measurement of the total particles mass to get a rough indication of potential damage caused by PM but one has to keep in mind that the particle's mass bears no correlation with traffic and has no clear causal relation with toxicity. The toxicity per μg of PM varies with place, season and sources. Because of its large contribution to particle toxicity, traffic is a source of special interest. The particle number is an indicator of traffic and can be used as such, though it has no causal relation with toxicity. Measurements of BC, particulate PAHs or NO_2 serve the same purpose.

Finally, the particle's mass seems to play an important role in the total pollutant mixture which is concluded in epidemiological studies. Van Bree and Cassee (1999) posit that the few toxicological studies that have been conducted so far, have not yet produced enough evidence to conclude that the current concentrations of outdoor particles can play a plausible role in the adverse health effects of air pollutants. On the other hand, it can also not be concluded with certainty that they do not play an important role. Rombout et al. (2000) conclude on basis of a literature study of epidemiological and toxicological results that PM_{10}, $PM_{2.5}$, $PM_{0.1}$, secondary and primary PM all could be indicators of the effects of PM. Besides these, other anthropogenic and biogenic compounds, such as LPS, pollen and a combination of PM and gases, cannot be excluded either.

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3. CONTRIBUTION OF TRAFFIC TO TOXICOLOGICAL EFFECTS OF PARTICULATE MATTER[2]

3.1 Introduction

The combination of chemical composition and particle size determines the toxicity of particulate matter, as described in the previous chapter. Traffic emissions have a distinct influence on composition and size and hence particle toxicity. Black carbon, PAHs and transition metals are important toxic components in outdoor aerosol, and a substantial part of these components are emitted by traffic. Further toxicity per unit mass of particles is enhanced in case of particles in the smaller size range, because of an increased availability of toxic compounds from their surface. The particles emitted by traffic are mainly in the fine and ultrafine fraction, for which toxicity is enhanced because of their small diameter. Hence, one of the main conclusions was that traffic seems to emit toxic compounds in those size classes that, by nature, already enhance the toxic property.

To measure the influence of vehicular emissions on the particle's toxicity, differences in size and concentration of air influenced by traffic and the 'average' background can be compared. To discover air masses being influenced by traffic the number concentration is used as an indicator (Hitchins et al. 2000, Yin en Harrison 1999, Harrison et al. 1999, Väkevä et al. 1999) though one has to realize that there is no causal relation with the particle's toxicity. The measure of toxicity used here are the following compounds: metals, PAHs and black carbon, together with the particle size distribution.

Concentration and size distribution measurements of metals in aerosol measured along a motorway are re-evaluated here. From chapter 2 it can be expected that metals are present in ultrafine PM emitted by traffic. In outdoor aerosol they are present in the larger size ranges due to agglomeration of particles and condensation. PAHs were also measured in this study. PAHs are emitted by traffic and other combustion sources, initially in the ultrafine mode, like metals; due to agglomeration and condensation they become present in larger aerosol size fractions. Also important are black carbon (BC) and organic carbon (OC). These are examined in a data set derived from a study in a suburb of Amsterdam.

Two questions are discussed here: first, in which typical size ranges does traffic emit toxic compounds, and second, are these different from those in ambient aerosol. In fact, these questions are similar to those treated in the literature study of chapter 2.

3.2 Description of field measurements

3.2.1 Campaigns

3.2.1.1 A9-highway

Measurements of car-exhaust were made along a highway near Spaarnwoude in September '98. The selected highway was the A9 that runs from Haarlem to Alkmaar. The maximum speed of the cars is 120 km/h, the average traffic density was 6360 vehicles/hr with 5% heavy traffic. Results from two measurement point were used, one background point at 30 m east of the road and one point at 10 m west of the road. Measured parameters are the particle number, the mass size-distribution and the size-distributions of two types of toxic compounds, the PAHs and

[2] This chapter is an adaptation of an internal report written by A. Even (2000).

metals. During the collection of the samples the wind speed was moderate and constant at 3-4 m/s, the atmosphere was slightly unstable. For further details see Vermeulen et al. (1999). Air advected from the east is clean after passing the IJsselmeer, but also slightly influenced by nearby cities. Downwind of the road the traffic emissions are added, therefore the contribution of traffic can than be discerned by comparing with the air upwind.

3.2.2 Amsterdam suburb

The second campaign providing data on traffic emissions was the Ultra-2 campaign located in a suburb in Amsterdam called Gaasperplas (December 1998 - June 1999). The measurement location in the Ultra-2 project receives two different types of air masses dependant on the wind direction. Air masses from the Southeast bear traces from source regions in Germany. During transport to the measurement site, the particles age, i.e. they increase in size. Further the concentration increases under influence of Dutch emissions, thus large particle mass concentrations are reached. With northerly and westerly winds, relatively clean air is advected from the North Sea. Flowing over land few pollutants are added, until the air reaches Amsterdam. In the city emissions from all types of sources are added while close to the experimental site emissions are added from two mayor highways and two highway junctions. Thus in these airmasses enhanced particle number concentrations are expected to be present.

Measured parameters are the fine mass concentration and number, and black carbon and organic carbon concentrations in the ultrafine and fine mass fractions. Fine particles are defined as particles with an aerodynamic diameter smaller than 2.5 μm, and ultrafine particles are defined as being smaller than 0.14 μm. Also, the mass, number and size distribution of the particles were measured. Particle number, mass and size measurements were made throughout the campaign. Additional carbon measurements were made in three periods: December 1998 and February and April 1999.

3.2.3 Measurement methods

The aerosol parameters were measured as follows. The suburban fine particle mass was collected at an Anderson Teflon filter with a pore size of 2 μm, installed in a holder behind a size-selective inlet with a cut-off diameter of 2.5 μm. The filters were weighted with a 10 μg precise microbalance after equilibrating at an RH of 45% relative humidity at 20°C. The suburban particle number was determined using a CPC model 3022. This CPC measures the sum of the particle number for particles with a diameter larger than 8 nm. The mass median diameter was measured as the volume mean diameter, using a combination of SMPS and LAS-X. The SMPS measured the particle size distribution from 8-265 nm, using an electrostatic classifier model 3071 combined with CPC model 3010. The LAS-X measured the particles from 265 nm –10 μm. For further details on data collection and handling see Khlystov et al. (2000). Measurements of the black and organic carbon content were made with an ACPM model 5400. The ACPM was modified in order to measure both the ultrafine and fine particles. This was done by placing a filter and an impactor with a 0.14 μm cut-off in the two lines of the instrument. Thus, the fine fraction and the fraction larger than 0.14 μm are measured alternately. From the difference the ultrafine fractions of black and organic carbon was determined.

The traffic emissions determined upwind and downwind of the highway, the A9, are the 16 EPA PAHs and 5 metals. The 16 EPA PAHs are naphthalene, acenaphthalene, acenaphtene, fluorene, phenantrene, anthrancene, fluoranthene, pyrene, benzo[a]anthrancene, chrysene, benzo[b]fluoranthene, benzo[k]fluoranthene, benzo[a]pyrene, dibenzo[a,h]anthracene, benzo[g,h,i] perylene and indeno[1,2,3-cd]pyrene. They were collected with a Sierra cascade impactor, thus size-dependant data were collected. Analysis was done by HPLC model HP1100 using an acetonitryl/water mix and a Supelcosil LC-PAH column. For further details see Vermeulen et al. (1999). The 5 metals that were collected are chrome, copper, zinc, cadmium and lead. They were also collected using a Sierra impactor. Analysis was done by graphite oven AAS with Zeeman correction. Filter material was extracted using nitric acid. Also here, for

further details see Vermeulen et al. (1999). The particle size distribution only was measured 10 m downwind of the road, using a Berner impactor. Mass on the impactor foils was determined using a 1 μg precise balance, after equilibrating of the samples at 20°C and 45% relative humidity.

3.3 Results

In this chapter the differences of the particle toxicity in traffic emitted particles and 'bulk' ambient particles are described. In paragraph 3.3.1 and 3.3.2 results from the highway experiment are described; in paragraph 3.3.3 and 3.3.4 results from the suburban experiment, including a comparison. Finally, in section 3.3.4 some of the implications are discussed.

3.3.1 Policyclic Aromatic Hydrocarbons

In figure 3.1 the sum of the EPA-PAHs is plotted for each size range as measured at 10 m upwind and downwind from the road. Also given is the contribution due to the traffic emissions on the motorway. The maximum concentration in the latter distribution as well as in air influenced by traffic appears in the smallest size fraction[3], $PM_{0.5}$. The traffic emitted PAHs in the $PM_{0.5}$ fraction are fluoranthene, pyrene, dibenzo[a,h]anthracene and benzo[g,h,i]perylene; in the $PM_{0.5}$ fraction in the background aerosol they are fluoranthene, benzo[g,h,i]perylene and phenanthrene.

A second maximum is present in the 1.5-3 μm interval for a number of PAHs in traffic-influenced aerosol. These PAHs were pyrene, fluoranthene and benzo[b]fluoranthene. The vapor pressure of these PAHs is relatively low and they are present in both the particles and the gas-phase. Because of the equilibrium between both phases, these compounds move from the small particles to larger particles in the accumulation mode resulting in a bimodal size-distribution also known in literature (Venkatamaram et al., 1994).

The size distributions up- and downwind are similar for total PAHs, except for the second maximum in the 1.5-3 μm interval that may be removed by deposition. Overall, of the PAHs in the fine aerosol fraction (PM_3) 59% is present in the $PM_{0.5}$ fraction in case of particles emitted by traffic; this is 55% in background air. It is noteworthy that only the absolute concentrations up- and downwind differ, and not the relative size-distribution.

3.3.2 Metals

The size distribution of the metals strongly resembles that of the PAHs. Again, a maximum appears for aerosols smaller than 0.5 μm, both in aerosol measured upwind and downwind of the road and in the net distribution. The size distribution of the sum of the measured metals is plotted in figure 3.2.

In background aerosol lead and zinc appear to be the most abundant. In traffic emissions lead, zinc and copper have the largest concentration. Again the absolute concentrations are higher downwind than upwind, but size-distributions are similar. A large fraction of the metals present in the fine particle range (PM_3) is in the smallest measured aerosol fraction ($PM_{0.5}$), i.e. 55% for traffic emitted particles and 53% for background particles. Furthermore, a small peak seems to be present in the coarse aerosol fraction but it is not significant in size.

The size-distributions of metals and PAHs are compared with that of the total mass distribution at 10 m downwind of the road (figure 3.3). Whereas the maximum for the mentioned species is in the smallest aerosol fraction, the maximum of the total mass is in the 0.5-1 μm interval. Apparently, the smallest size-fraction is more enriched with metals and PAHs. This result will

[3] The PAHs in this size range were measured at a backup filter placed behind an impactor and may be overestimated by adsorption of gases. This was done at both upwind and downwind location.

be used in the next sections, when air masses containing PM with different size distributions are discussed.

Two observations should be made. First, when plotted in one figure, it is clear that the size distributions of metals and PAHs resemble which is natural as the two compounds are emitted by the same source. This suggests that the effect of possible sampling artifacts for the PAHs might be small in these measurements. And secondly, the size-distributions of the PAHs and metals upwind and downwind also resemble each other, only the absolute concentrations differ. A possible explanations may be that the background aerosol already has been influenced by traffic emissions upwind of the road.

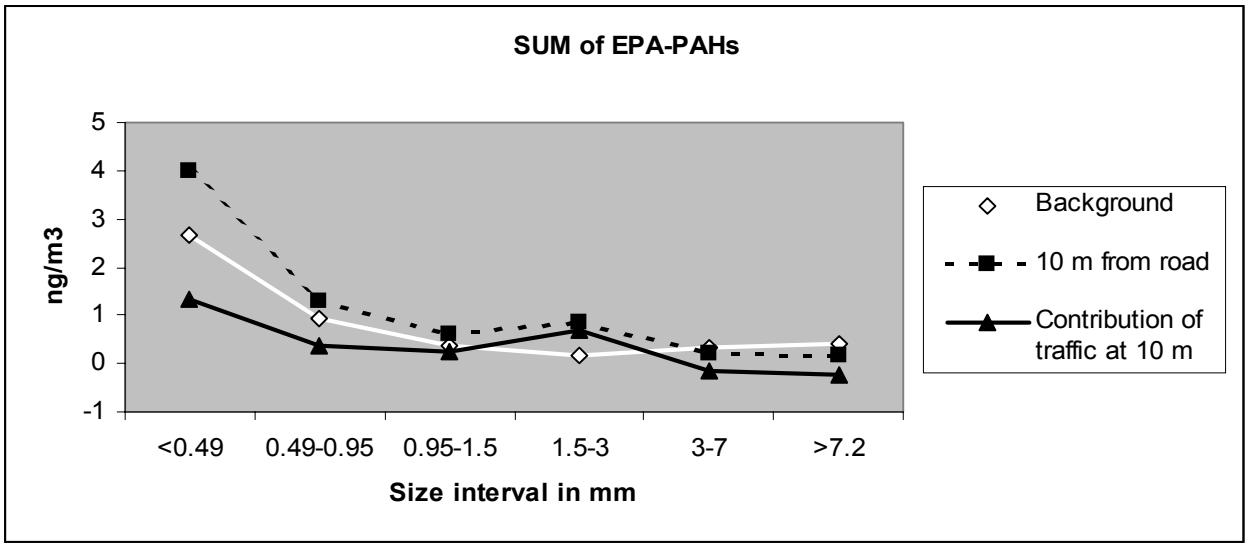

Figure 3.1

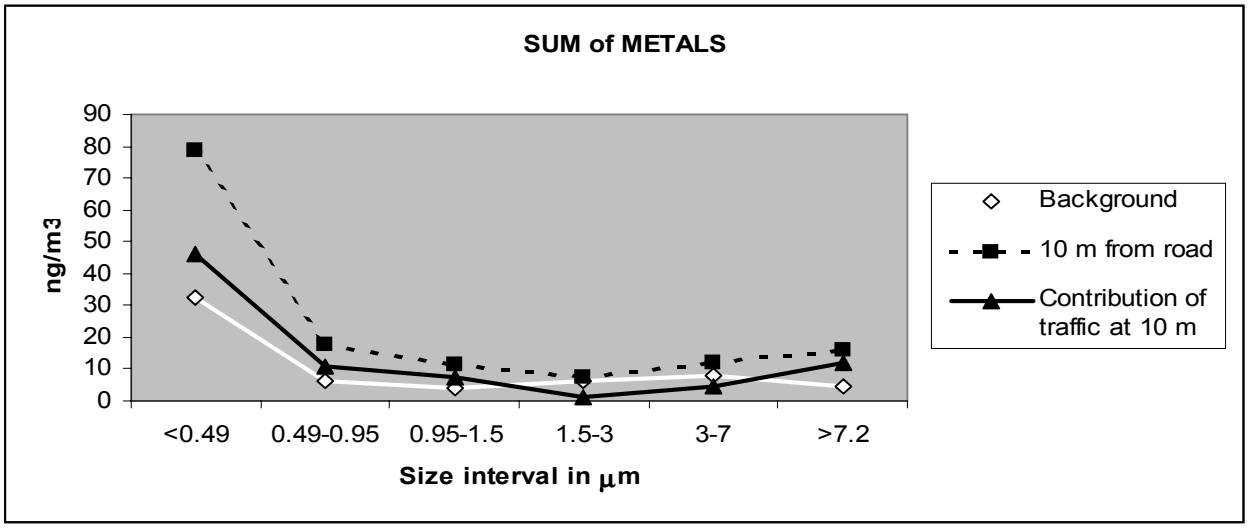

Figure 3.2

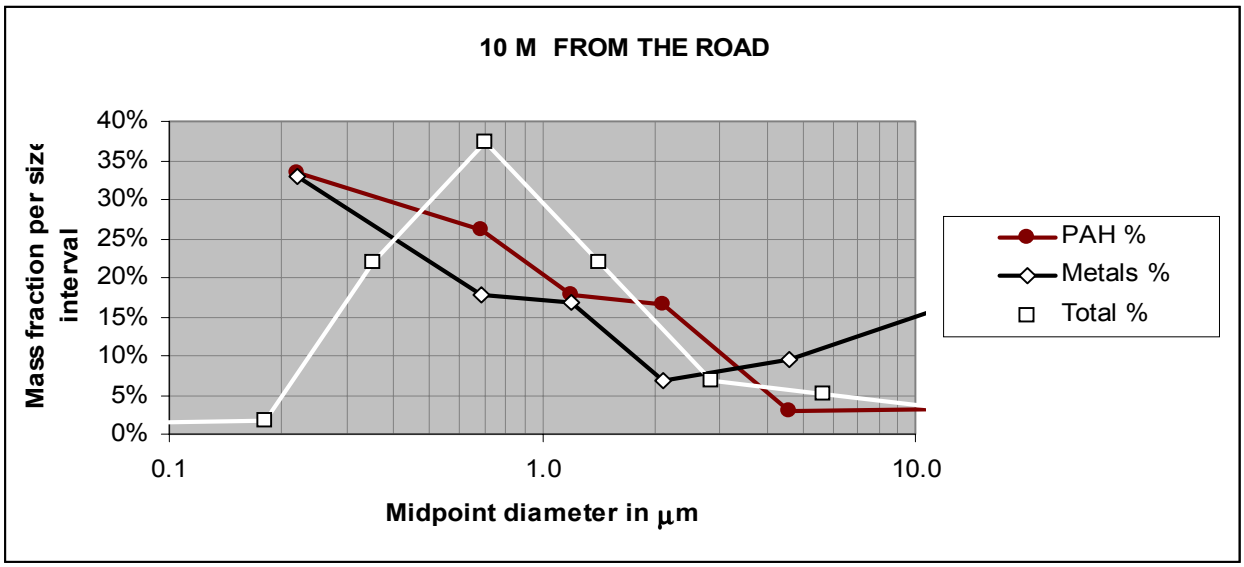

Figure 3.3

3.3.3 Black and Organic Carbon

The black and organic carbon findings obtained in the Amsterdam suburb campaign are given in figures 3.4-3.9. Before ascribing results of carbonaceous carbon to the influence of traffic, it has to be ascertained whether there are two different wind sectors distinguished with and without the contribution of recent traffic emissions. The presence of recently emitted traffic exhaust in air masses is indicated by an elevated particle number concentration.

In figure 3.4 the particle number concentration (Ncpc) is plotted as a function of wind direction. Clearly, in the wind sector 235°-315° the numbers are elevated. Additionally, Khlystov et al. (2000) corrected the particle number concentrations in Amsterdam by subtracting the number concentrations per wind sector at the background location Petten. Their results strengthen the conclusion that the particle number in the wind sector 235°-315° is elevated. Apparently, this wind sector is influenced by recent traffic emissions. The fine particle mass also is plotted in figure 3.4 ($PM_{2.5}$). The mass concentration is elevated in the wind sector 75°-175° as is everywhere the case in the Netherlands. The particle mass is *not* elevated in the traffic-influenced wind sector 235°-315° indicating that air influenced by traffic exhaust does not correspond to an elevated particle mass concentration.

The absolute concentrations of black and organic carbon are distributed over the wind sectors similar to the fine particle mass concentration (figure 3.6). That is in the southeasterly windsectors the mass concentrations are larger than in the ones from the west. In the figures 3.8 and 3.9 the mass fractions of black and organic carbon in the total fine mass are plotted. In contrast to the absolute carbon concentrations, the mass fractions of both black and organic carbon are elevated in the westerly wind sectors that are influenced by traffic. Clearly, the presence of carbonaceous material is associated with traffic emissions.

For both carbonaceous aerosol compounds the ultrafine particle fraction was measured. On average ca. 40% of the black carbon is in the ultrafine mode. This fraction is larger in the traffic influenced wind sector, but not significantly (figure 3.7). Of organic carbon ca. 50% is present in the ultrafine aerosol, and also this does not vary significantly for different wind sectors (figure 3.7). This is surprising, since it was expected that the ultrafine organic carbon fraction would be lower in polluted air, because of redistribution of semi-volatile compounds to larger aerosol sizes and condensation of secondary organic carbon on the same sizes. Perhaps in future this can be demonstrated by measurement methods with a better accuracy.

3.3.4 Particle mass and number size-distributions

In the Ultra-2 project the size distribution was determined by LAS-X and SMPS, which allowed the determination of the number distributions. From the number distribution the mass size distribution was calculated. The particle mass concentration is reflected by the mass median diameter. In the south-east wind sector, where aged aerosol is present, the mass median diameter is significantly larger than in the other wind sectors such as the traffic influenced sector (see again figure 3.5). The difference in the mass median diameter between these sectors is 80 nm.

In figure 3.10 the size distributions for both windsectors (denoted by Ultra-2 southeast and Ultra-2 west) are plotted using SMPS data. Note that the particle number is plotted on a logarithmic scale. Both distributions are bimodal. There is one mode (ultrafine) for particles up to 20 nm, from which only a tail is measured. This mode is produced by combustion sources, an important one is traffic (Hitchins et al. 2000, Yin en Harrison 1999, Harrison et al. 1999, Väkevä et al. 1999). The other mode is the accumulation mode. Its sources are coagulation of ultrafine particles and condensation of secondary aerosol compounds.

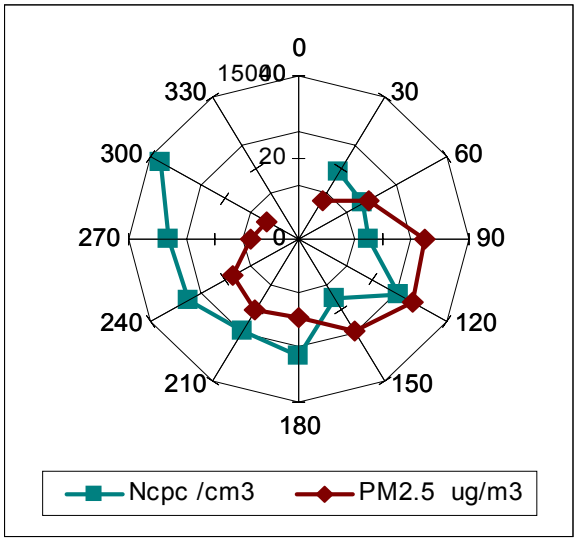

Figure 3.4

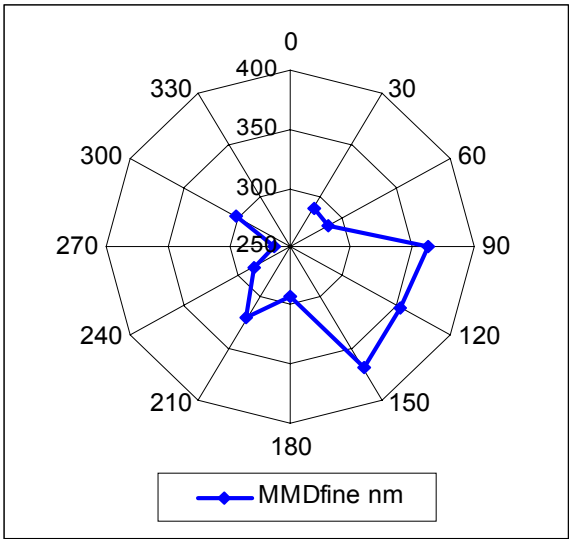

Figure 3.5

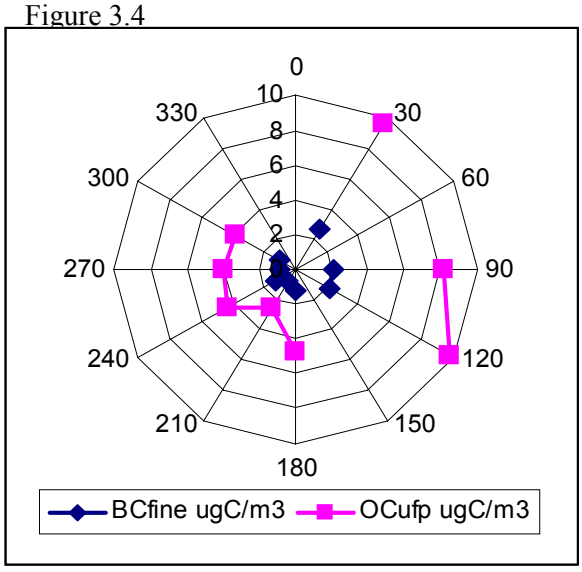

Figure 3.6

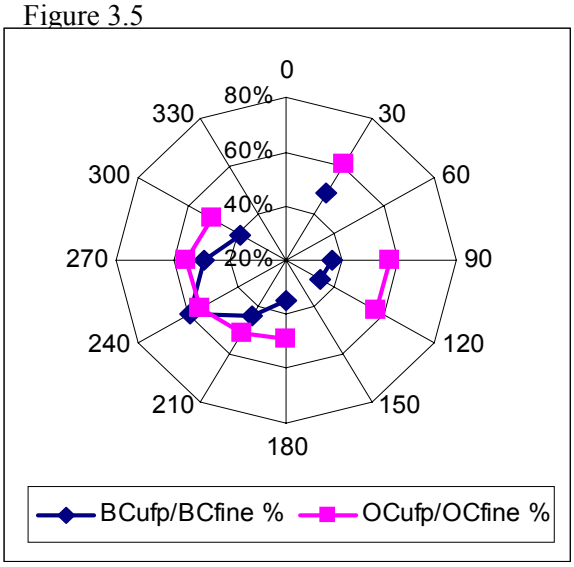

Figure 3.7

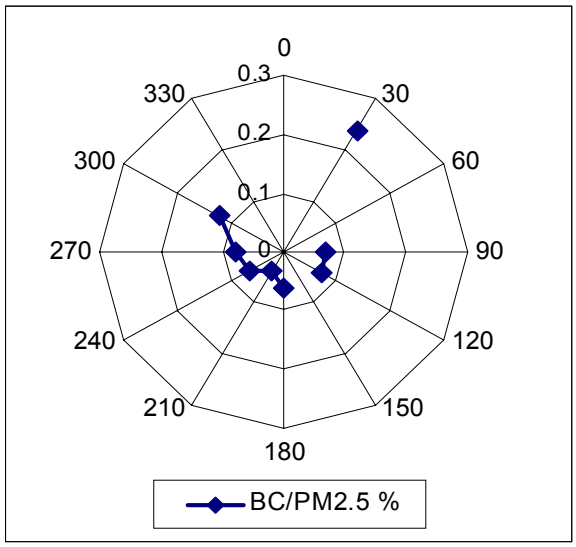

Figure 3.8

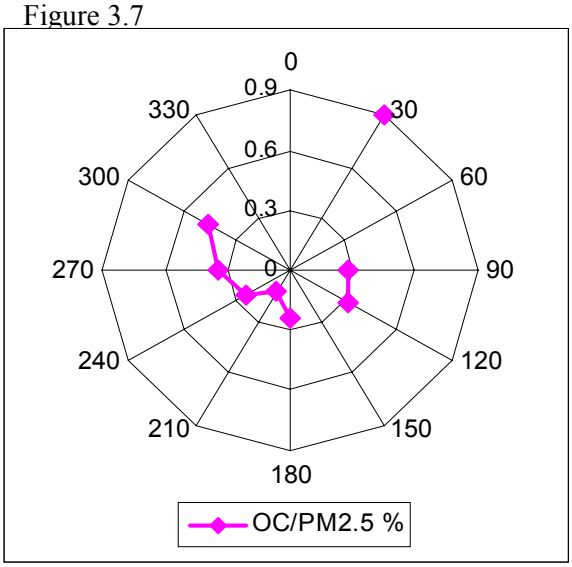

Figure 3.9

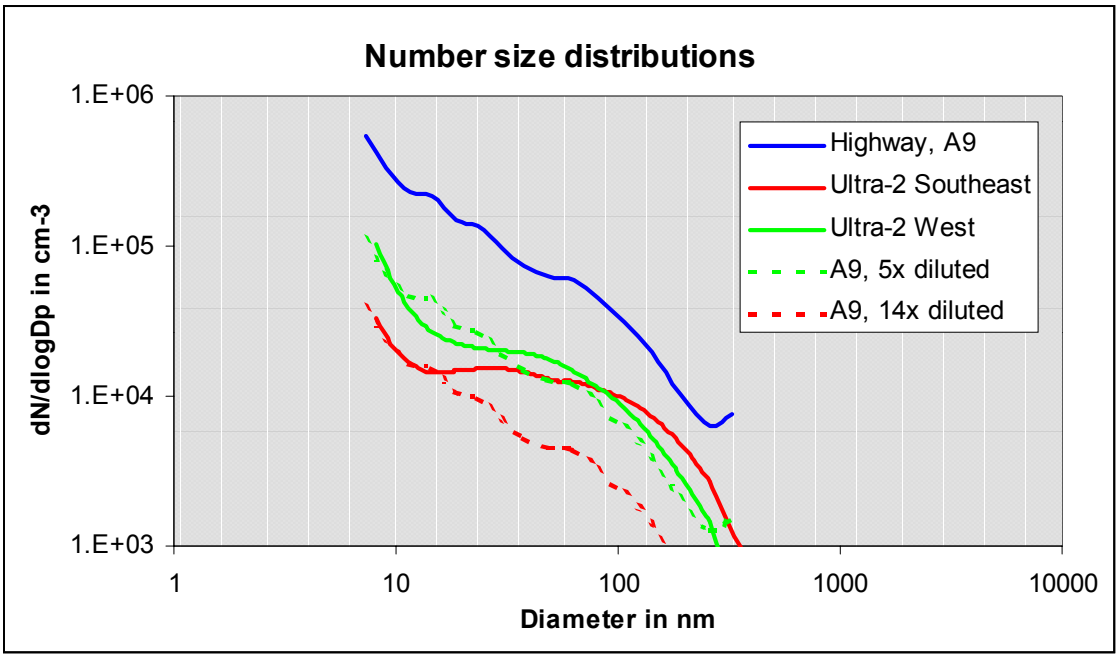

Figure 3.10

Two differences between both size distributions are visible. As expected the ultrafine mode is larger in the traffic influenced west sector. Since this mode contributes largely to the particle number, this is consistent with the earlier observation of larger particle numbers in this sector. The total particle number in the west sector is on average $3.0*10^5$ per cc while in the southeast sector it is $1.9*10^5$ per cc (a difference of 30%.) In reality the difference between the total particle numbers will be even larger, since the method that was used to determine the particle number only measures particles larger than 7 nm (see chapter 4 on CPCs). Also in figure 3.10 it can be seen that the smaller the particles, the larger the difference in particle number is. For particles larger than 100 nm the particle number is slightly larger for the east sector. Since most of the fine mass is present in this larger than 100 nm mode, the particle number is slightly larger for the east sector.

Downwind of the motorway in the first experiment, the number size distribution was also measured. The result is also given in figure 3.10. The total particle number here was $4.7*10^5$ using the same measurement method as for the Ultra-2 experiment. The size-distribution of particles influenced by the emissions on the motorway resembles the one measured in the west-sector during the Ultra-2 experiment, though the relative contribution of the accumulation mode seems smaller.

In the same figure 3.10 also two other lines are plotted. These are the size-distribution of the motorway as if diluted. In this way the ultrafine particle mode of the Ultra-2 distributions was matched. The differences of these ‘diluted-highway’ distributions and the Ultra-2 distributions is another way to estimate the influence of traffic emitted particles to the suburban aerosol. For the west-sector this renders two almost equal distributions, confirming the strong influence of traffic to it. For the southeast-sector clearly an additional mode is present, confirming the low contribution of freshly traffic-emitted particles to it.

3.4 Discussion and Conclusions

Based on the data from both experiments, the following qualitative conclusions can be drawn towards differences in the toxicity of the particles emitted by traffic and the bulk of particles in ambient air.

- As described in section 3.3.4 the main difference between freshly emitted traffic particles and the 'bulk' of ambient particles is the ratio of the ultrafine to the accumulation mode. In traffic influenced air more than 90% of the particle number is in the ultrafine mode, but this mode contains only a small fraction of the fine particle mass. Most of the mass is in the accumulation mode.

- In figure 3.3 it was shown that the chemical composition of the ultrafine mode differed from the accumulation mode. The graph shows that the smaller the particles, the larger the concentration is of the measured toxic compounds PAHs and metals. From this it is concluded that the ultrafine mode is more enriched with toxic compounds than the accumulation mode in which a large proportion of the mass consists of secondary materials such as sulfate and nitrate that are not toxic. The difference is larger than it appears as half of the mass of the accumulation mode is present in the $PM_{0.5}$ size-interval, thus diluting the contribution of PAHs and metals to it.

- In figure 3.8 it was shown that also the black carbon concentration, being related to toxic compounds, is largely present in the ultrafine particle fraction. Together with the data presented in figure 3.4, this means that there is a difference in particle toxicity in the traffic influenced air masses in the western wind sector compared to the south-eastern windsector with higher particle mass concentrations. In the latter case the large contribution of the accumulation mode to the aerosol mass induces a smaller toxicity per $\mu g/m^3$. Because the ultrafine particle mode only carries a small proportion of the fine particle mass, it does not necessarily result in a large increase on the particle toxicity per µg. However, it does result in an increase of the toxicity of those particles that are deposited in the lung because the deposition efficiency of ultrafine particles is larger than that of accumulation mode particles.

- Because the PAH and metal concentrations are not linearly related to the particle mass concentration, the particle's toxicity is probably not related to mass concentrations either. Furthermore, the concentration of these compounds is larger in the ultrafine particles with a larger deposition efficiency in the lung than in the accumulation mode. This indicates that the toxicity of the particles deposited is not linearly related to the total mass of toxic compounds present in the fine particles. Thus, on basis of the reported measurement results, it can be concluded that the toxicity in particles influenced by recent traffic emissions is larger than for the 'bulk' ambient particles.

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4. COMPARISON OF DIFFERENT TYPES OF CONDENSATION PARTICLE COUNTERS (CPC)

4.1 Introduction

In measuring campaigns characterising urban air the use of Condensation Particle Counters (CPC's, sometimes referred to as Condensation Nucleus Counters) is indispensable. These instruments measure the number concentration of particles in the air; number concentration is one of the physical parameters of particulate matter possibly responsible for the adverse health effects described in Chapter 2. Considering the properties of the smaller-size fractions of PM, their numbers may rise significantly to levels well above one million per cubic centimetre under certain meteorological, infrastructural and traffic conditions in the urban environment.

There are various types of CPC's available on the commercial market. Here, a comparative test is described between three types that are common in current measurements in urban air. The objective of this assessment is to find out which type is technically most appropriate in the determination of the number of particles in urban air. The result of this study will be taken into account in the future work. The TSI-types examined here are CPC-3010, CPC-3025 and CPC-3022.

In general, the technical principle of a CPC is based on measuring the (forward) scattered light of a laser beam. The scattering is caused by a through passage of a uniformly sized droplet grown out the micron or submicron particles. The particles 'grow' to a final size of approximately 5 μm. This is the result of exposure of the particles to a supersaturated butanol atmosphere; the air with the particles in it is lead over a warmed bath with butanol (saturator held at 30 °C) and subsequently travels through a cooling element (condenser, 10°C).

4.2 Present limitations in the measurements with CPC's

Each type of CPC has its specific lower detection limit. This is the result of a combination of flow, exposure time (and thus growth time) and temperature settings of saturator and condenser; the warmer the saturator the more vapour is available to condense onto the particles and the colder the condensor is the more vapour is forced to condense. The efficiency curves can be seen in figure 4.1.

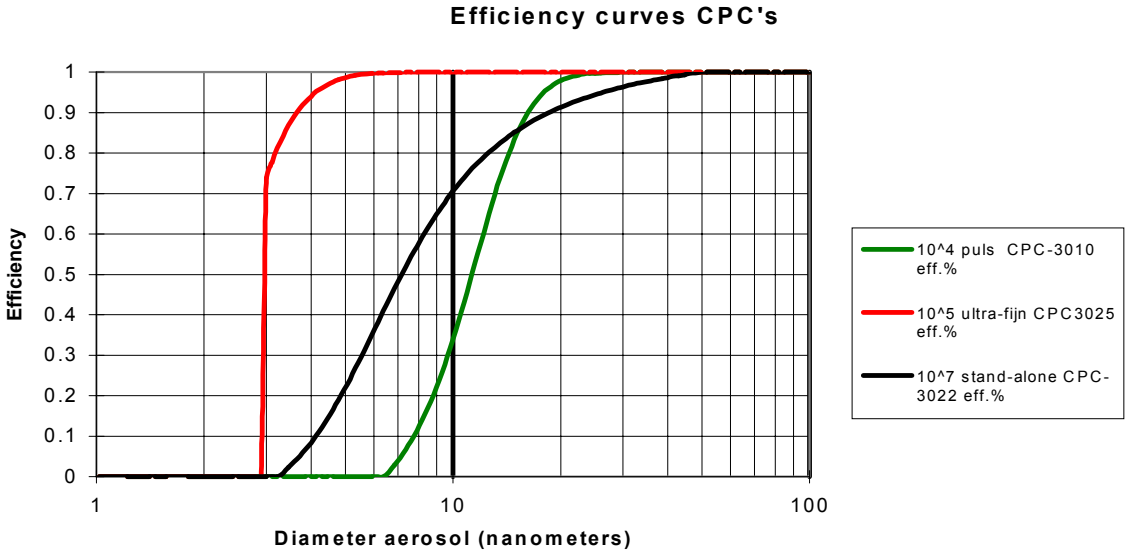

Figure 4.1 *Efficiency curves for the CPC-3010, CPC3025 and CPC3022*

Clearly, the smallest particles can be measured with the CPC-3025: aerosols with diameters of some 3 nm have an efficiency of 75%; above diameters of 7 nm or more the efficiency is 100%. In case of the CPC-3022, 50% of the particles having a diameter of 7 nm is detected while a 100% efficiency is attained at 60 nm. For the CPC-3010 the 50%-efficiency point occurs around 12 nm; at 30 nm 100% of the incoming particles are measured.

The maximum number of particles that can be detected also varies: the CPC-3022 is a typical stand-alone instrument capable to measure as much as 10^7 particles/cm^3; the CPC-3025 has the ability to measure a maximum number concentration of 10^5 particles/cm^3 while the CPC-3010 is limited to 10^4 particles/cm^3. The above results are summarised in the table below (table 4.1).

table 4.1

Type	50% efficiency (nm)	100% efficiency (nm)	maximum number
CPC-3010	12	30	10^4
CPC-3025	3	7	10^5
CPC-3022	7	60	10^7

Concluding, the CPC-3010 is less suitable to be part in urban air concentration measurements due to the limited particle numbers it is capable to measure. Still, the instrument is good enough to be integrated with a SMPS (Scanning Mobility Particle Sizer)[4] when dealing with particles larger than 12 μm.

[4] The SMPS consists of a classifier and a CPC that is connected to its aerosol exhaust. The classifier consists of two concentric cylindrical electrodes. A high voltage supply controlled by a computer maintains the centre electrode at a precise negative potential. The outer electrode is kept at zero potential level. Particles with positive charges are attracted towards the negatively charged electrode. The trajectory of such a particle is a function of flow-rate, analyser geometry, electrical field, particle diameter, and the number of charges on the particle. Only particles within a narrow mobility range pass through a slit near the bottom of the centre electrode. A small air flow carries the particles out of the electrode. The positively charged particles do no longer play a role in the measurement. In order to relate the electrical mobility to particle size, the electrical mobility of a particle must be considered. It is a measure of how rapidly an electrically-charged particle will respond to an electrical field. Particles passing a so-called neutraliser (a Krypton 85 radio-active source) will be charged with elementary charge units or remain neutral. The distribution between the charged and neutral particles is a matter of well-known statistics that is directly related to particle size. This phenomenon is called the "Boltzmann equilibrium". During measurements this effect is used to reduce the data of the SMPS-program with a factor of 30 (see Scheibel and Porstendoerfer, 1983). In the end a size

The CPC-3025 is not very suitable to measure directly near urban sources because of its number concentration limit: on typical urban locations – usually somewhat distant from the traffic flow – its number readings will be close to 100% of its upper limit. However, the incorporation of the CPC-3025 in a SMPS-system is most valuable due to its capability of detecting very small particles.

When dealing specifically with number measurements the use of the CPC-3022 will be most rewarding; its efficiency is a factor 100 lower than that of the CPC-3022.

4.3 Experimental set-up CPC tests

To estimate representativity and accuracy of the CPC instruments their behaviour was studied under similar experimental conditions[5]. The first (laboratory) test was done with particles having a (number-median) aerodynamic diameter of 0.3 μm (S.D. 2.1 μm); more than 99% of the particles have a diameter above 20 nm. Because number concentrations in urban air are regularly in the order of 10^4- 10^6 particles/cm^3 the efficiency of the CPC's within this range is particularly important. Therefore, a second test was executed measuring urban air during fairly stable weather conditions.

spectrum can be related to the number counts measured by the CPC instrument. Due to the number reduction the CPC-3010 and CPC-3025 can be part of this system.

However, reductions may be huge. In this set up measuring an aerosol spectrum from 0.01 up to 1.0 μm is a measurement of the amount of positively charged particles as function of diameter within this size range; the detected amount may be as low as 0.34 % for particles with a diameter of 0.01 μm, increasing up to 24.1% for 0.10 μm followed by a decrease down to 12.7 % at a diameter of 1.0 μm. It is obvious that a correctional factor of 30 for 0.01 μm particles may lead to huge under- or overestimation of the true numbers of particles with this size.

[5] In the testing of the comparability of the CPC's while measuring non-critical aerosol sizes (i.e. with respect to the lower size limits) the experimental set-up consisted of: aerosol generator, vessel of 25 litre filled with Raschich-rings of approximately 1 cm diameter (for removal of the smallest particles by diffusion), neutraliser (Kr-85 2 MCI) and short metal tubes connected to the CPC's under consideration.

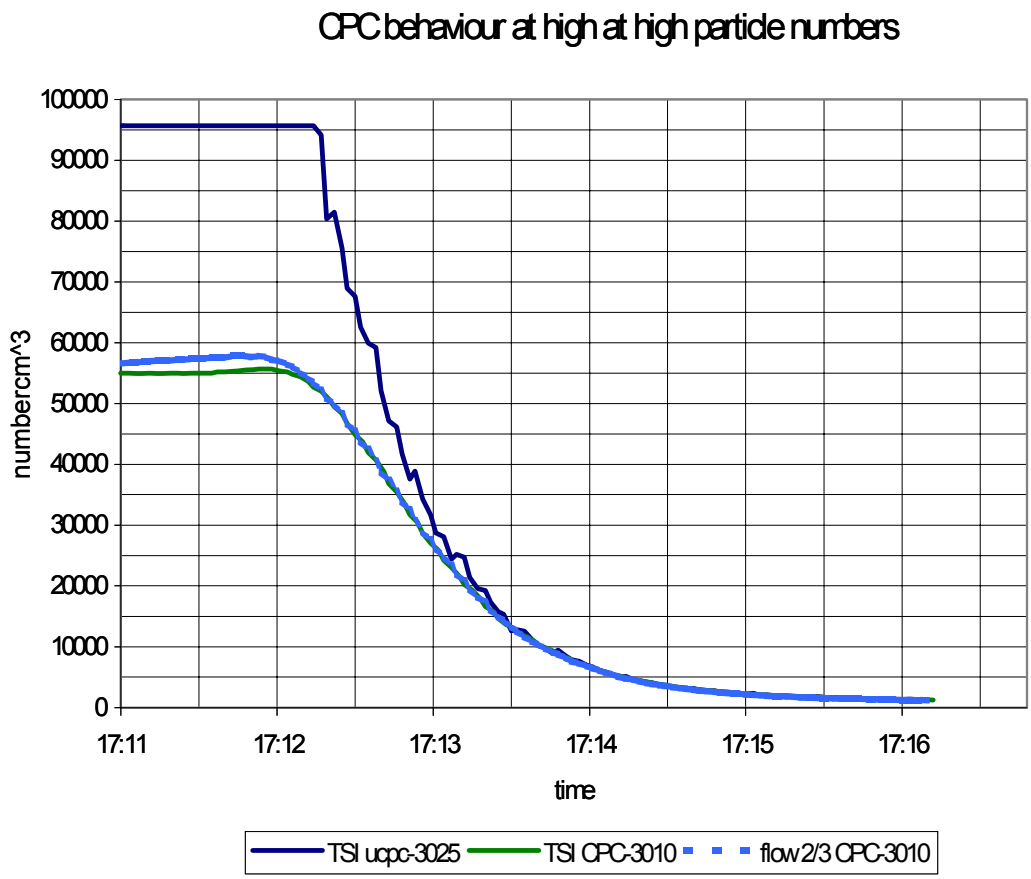

Figure 4.2 *Testing of the upper limits of particle number concentration for two types of CPC's: CPC-3025 (ultra-fine) and two CPC-3010's , one with nominal flow and one with reduced flow (2/3) where the CPC-3010 with reduced flow is corrected for flow.*

4.4 Results

In figure 4.2 results of the first test are shown. Indeed, a loss in efficiency occurs when measuring with the CPC-3010 number concentrations above 10^4 (due to coincidence losses). Saturation takes place at about $5.5 \cdot 10^4$ n/cm^3. However, by use of a correction polynomial derived by comparing with the CPC-3025, it is possible to enhance the CPC-3010 measuring range above its upper technical number limit (see figure 4.3).

The CPC-3025 changes its measuring technique above 10^3 n/cm^3 into the 'time of illuminescence' principle (i.e. measuring the time a particle resides in the laser beam), and above 10^4 n/cm^3 into the 'strength of illuminescence' principle (measuring the strength of stray-light reflected by the particles (or better, droplets) in the laser beam). This system therefore does not suffer from efficiency losses by coincidence. The upper limit for this type is around 10^6 n/cm^3.

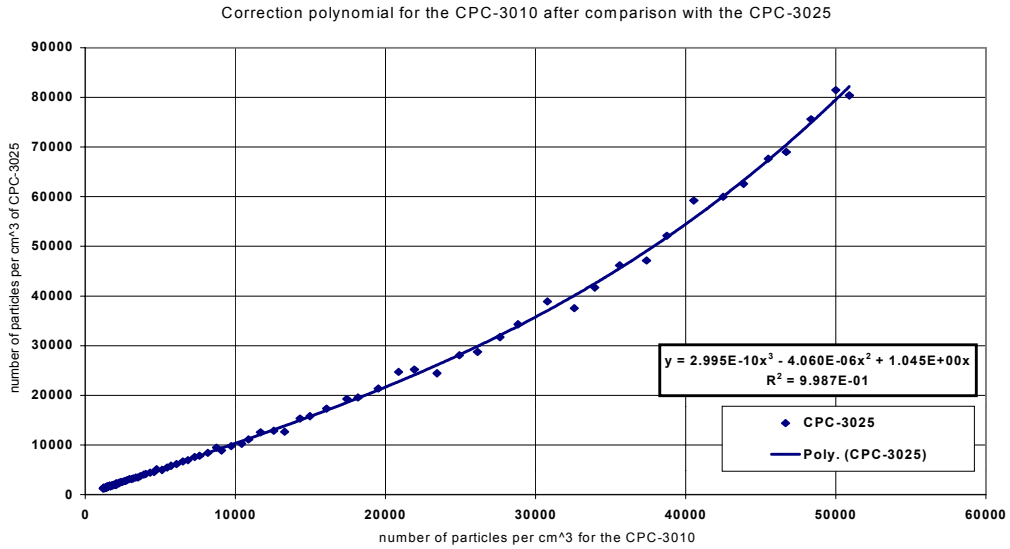

Figure 4.3 *Comparison of the CPC3010 with the CPC-3025 and resulting polynomial fit for correction.*

Knowing the difference in efficiency of the various CPC's one can take advantage of this in the interpretation of measurements of urban air at concentration levels below the upper limits of both instruments: an estimate of the number of very small particles can be made by subtracting the number measured with one CPC (i.e., the one with a higher low-diameter boundary, e.g. CPC-3010) from the number measured with another CPC (capable of counting smaller particles, e.g. CPC-3025).

Of course this estimate can become a reliable value if the lower size cut-off values (detection limits) have been determined. This requires a calibration with a calibration facility as described in Scheibel and Porstendörfer (1983) to be redesigned and build in the future. The result of such an exercise could be more reliable and rewarding than information drawn from the SMPS because of the large corrections needed in the applied data reduction of SMPS-measurements in this size range (see also comment in footnote 1). An example of a measurement with CPC's with different lower size detection limits is shown below (figure 4.4).

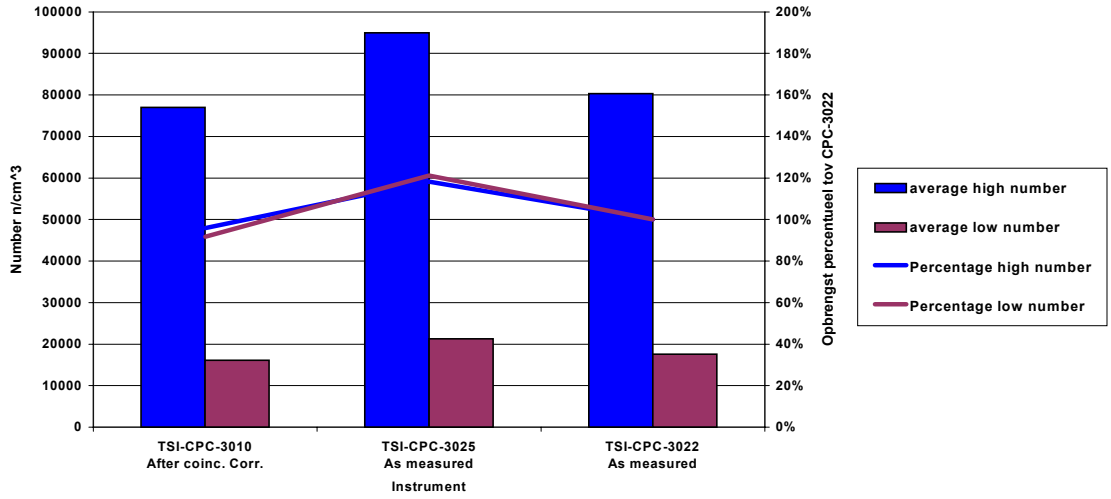

Figure 4.4 *Average number concentrations for two measurement periods in Amsterdam; CPC-3025 was higher than 10^6 - upper limit - during 75% of the "high number" period*

In this figure two cases are compared: measurements in an urban area with relatively high number concentrations and measurements in an urban area with relatively low number concentrations. As can be deducted from this figure, CPC-3025 measures on average 20 percent more particles than the CPC-3010 and CPC-3022 (last instrument taken as reference) in case of lower concentrations. This difference is due to the number of particles with diameters between 3 and 10 nm. This difference is even larger for the case with relatively high number concentrations for during these measurements the CPC-3025 remained during 75% of the time stable on its upper concentration limit of 10^6 n/cm^3 which means that the actual concentration during this period was higher but with an unknown amount. Even without this correction the difference between the CPC-3025 and the other CPC's was already 20 %. On estimate this percentage is probably closer to 30 or 40 % for this case.

4.5 Conclusions and recommendations

It is concluded that particle measurements in urban air are possible with the three Condensation Particle Counters in various experimental set-ups. The experimental set-up should depend on the concentrations to be expected. If the experimental site is not located in a street or not directly influenced by traffic - for instance in a park - all three CPC's may be used as stand-alone instruments. The varying low cut-off diameters may offer extra information on the amount of particles having diameters between 3 and 10 nm (or 20 nm).

However, an experimental set-up like the CPC-3022 stand-alone is to be preferred in the determination of urban air concentration levels. The other type of CPC's (3010 and 3025) can better be integrated into an SMPS-system because the extra information such a configuration gives on spectra and time-evaluation of spectra during the day. Another advantage of this set-up is that the total particle surface and total particle volume (or mass if density is known) can be calculated for the measuring range of the SMPS-system.

Reference

Scheibel, H.G. and J.Porstendörfer: J. Aerosol Sci. Vol 14 No.2 pp 113-126, 1983. Generation of monodisperse Ag- and NaCl-aerosols with particle diameters between 2 and 300 nm.

5. MEASUREMENTS OF PARTICLE NUMBER AND DENSITY WITH A MOVING UNIT IN THE CITY OF AMSTERDAM

5.1 Introduction

Mobile traffic sources are known to emit large quantities of ultra-fine particles, one of the physical properties believed to be (at least partly) responsible for the observed adverse health effects discussed in chapter 2. Because of the large spatial variability of traffic intensity within a city, the contribution of traffic to the number and mass concentration levels is expected to be varying largely depending to place and time. In this chapter, a preliminary analysis is presented of an experiment in which PM measurements have been performed by using a set of instruments installed on a moving vehicle. While driving through marine, rural as well as urban areas (Amsterdam and immediate surroundings) an impression of the spatial inhomogeneity can be made.

5.2 Experimental set-up

The measurements were performed with an ultra-fine condensation particle counter, CPC 3025 (TSI Inc.) and an optical particle spectrometer, LAS-X (PMS Inc. Modified with new electronics package from DMT Inc.). The CPC measures number concentration of particles with diameters larger than approximately 3 nm. The LAS-X measures concentration of particles with diameters between 0.1 and 10 μm in 15 size bins**.** The difference between the number concentration measured with the CPC and the summated number concentration measured with the LAS provides a measure of the number concentration of ultra-fine aerosol, particles smaller than 100 nm in diameter. The LAS-X actually provides number size distributions, which can be converted, using the average size of each size bin, to volume size distribution. The volume distribution, in turn, can be converted to the mass size distribution using the density of the particles, which usually has to be assumed.

To study the spatial variability of the aerosol number and volume concentrations in urban atmosphere, the CPC and the LAS-X were installed into a van. The instruments were placed inside the van's cabin, with the sampling inlets being at the roof of the cabin, facing the front of the vehicle (about 2.5 m above the ground). The instruments were sampling through approximately 1.5 meters of ¼" copper tubing. The sampling flow of the CPC was 1.5 l/min, while that of the LAS was 0.3 l/min. To allow for better spatial resolution of the measurements, the time resolution of the instruments was set to 2 s. The position of the vehicle was recorded with a GPS. Also written notes were taken of the situation on the road to help the interpretation of the measurements at a later moment.

5.3 Results

5.3.1 Number concentrations measured with CPC

In figure 5.1 the number of aerosols as measured along the ride is given, starting at Petten near the seacoast and ending in the Amsterdam area. Apparent is the gradual increase in numbers when approaching the urban environment of Amsterdam: during the ride in the coastal/rural area between Petten and Alkmaar the number concentrations were low (most of the time less than 10.000 n/cm^3); very low numbers were found in the immediate vicinity of the coast (less than 5.000 n/cm^3). The influence of the presence of urban agglomerations (Alkmaar,

Heemskerk, IJmuiden) becomes apparent when driving from Alkmaar to Amsterdam. Roughly, the lower limit in the number concentration is 10.000 n/cm^3 during this ride.

When entering the urban areas numbers rise rapidly up to 150.000 n/cm^3 and, at some locations, even attain extreme values around 10^6 n/cm^3. Overall, the highest numbers are observed when driving on the ring motorway (A10) around Amsterdam and in the centre. In the city itself the highest numbers are measured at busy road crossings characterised by accelerating and decelerating vehicles.

Figure 5.2 is the average result derived from two rides on the motorway ring A10. The findings of these two tours are pictured separately in figure 5.3a-b in order to obtain an idea about the variation in time. The traffic intensities at the western and southern 'axes' are the highest of all roads in Amsterdam. This is partly reflected in the measured number concentrations along the western axis of the ring motorway during both rides. Probably, the effect is enhanced by the prevailing SSW wind direction, which is almost along the western leg of the A10. The wind direction may also be responsible for the relatively low numbers found at the southern leg with the exception of the high values occurring near the junction with the A2-motorway.

As anticipated, very extreme number concentrations are measured in tunnels (Coentunnel, IJ-tunnel and Zeeburgertunnel, figures 5.2 and 5.3). While going through the tunnel it was repeatedly observed that the highest values (around $1.3 \cdot 10^6$ n/cm^3 in the Coentunnel) are measured in the second part of the tunnel. An explanation is that a relatively turbulent wind flow field is directed into the tunnel being generated by cars entering the tunnel mouth and bringing in clean(er) air from outside the tunnel. During the ride in the tunnel this influence diminishes. Another factor, probably more important is that car emissions in the second tunnel half are higher because more power is needed to overcome the uphill slope towards the end of the tunnel.

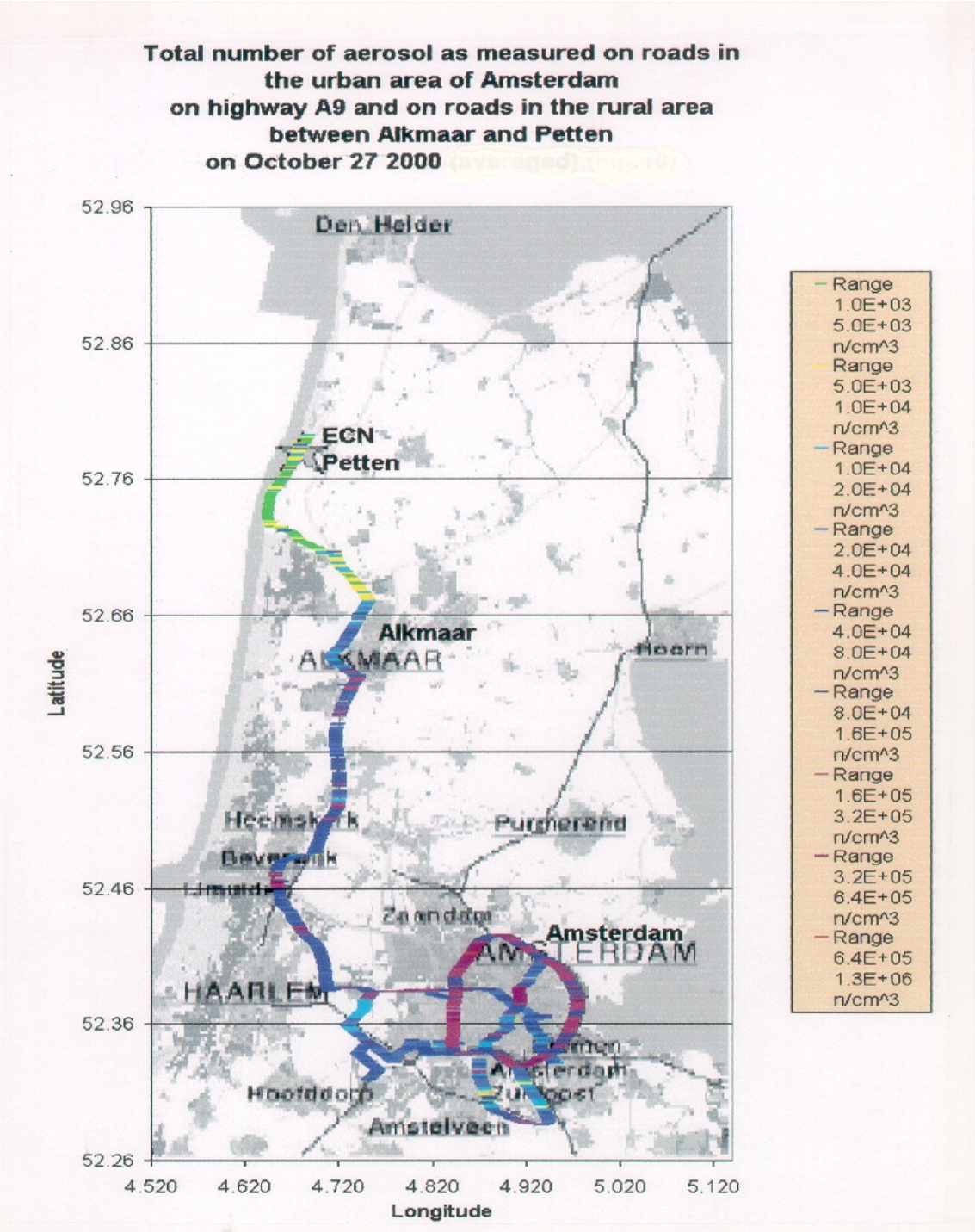

Figure 5.1 *Number of aerosols measured along the road from Petten to the city of Amsterdam*

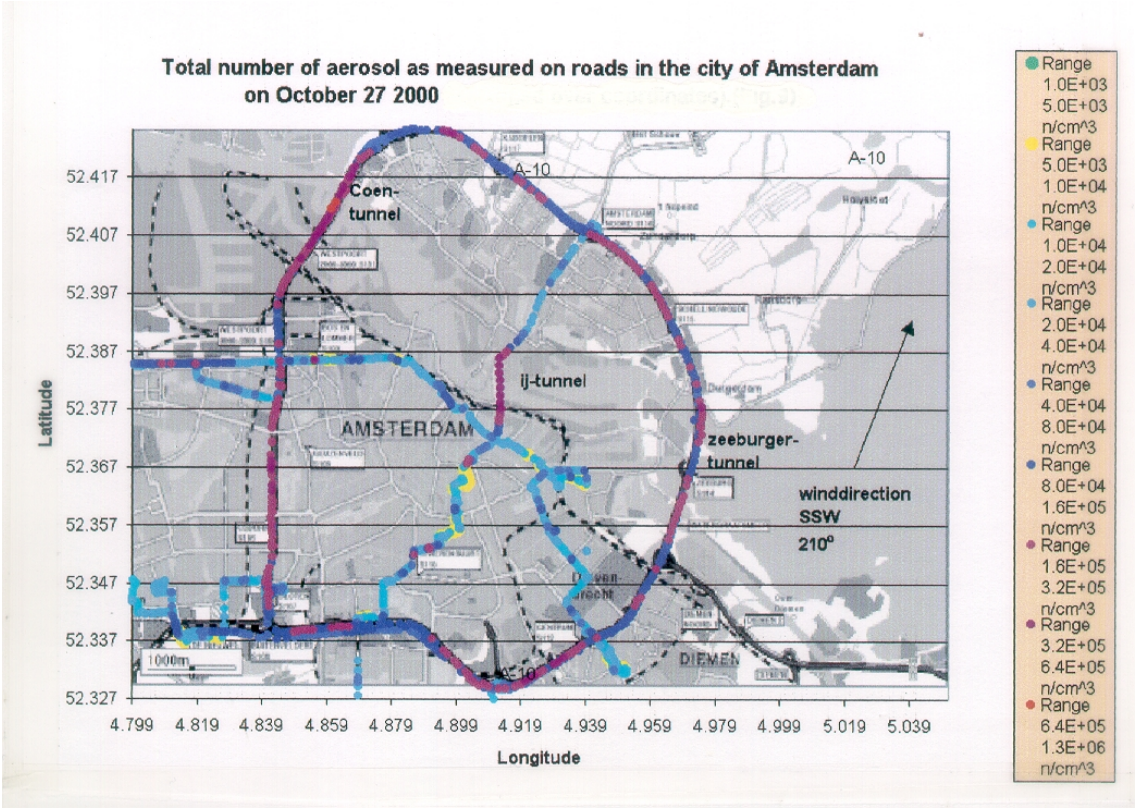

Figure 5.2 *Number of particles averaged over two tours on the A10 ring motorway of Amsterdam.*

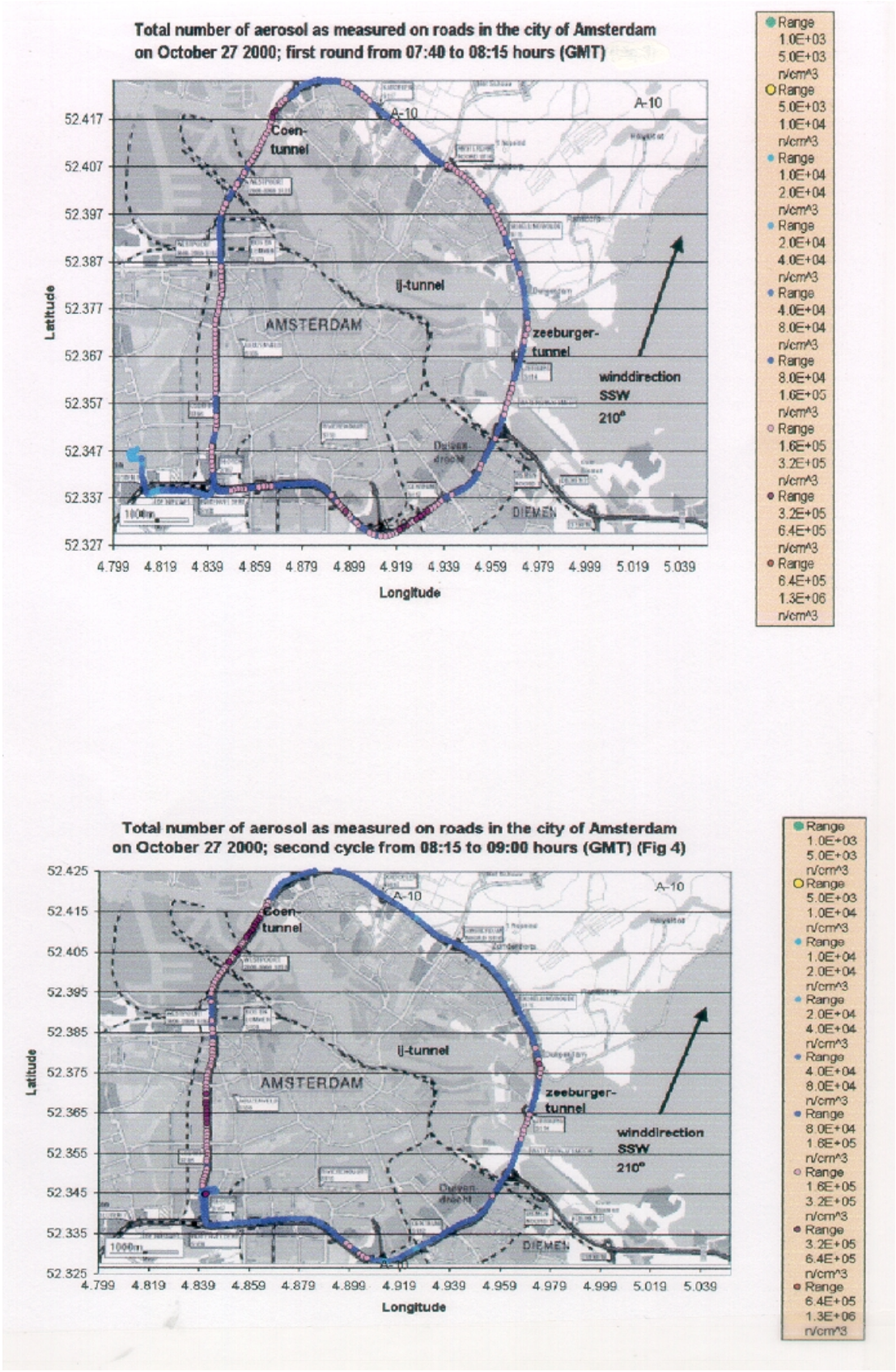

Figure 5.3 Number of aerosols measured during the first (a) and second (b) tour on the A10 ring motorway in Amsterdam.

Finally, the averaged atmospheric number distribution is shown in figure 5.4. To discern the presence of different surroundings, the data set, i.e. the measurements along the traversed road, is divided into four sections: urban (city of Amsterdam including the contribution of motorways and tunnels), urban-influenced ('suburban'), rural (between Alkmaar and Schoorl) and coastal (Petten-Schoorl). For each section the particle size-distribution has been calculated. Note that the particle diameter is shown on a logarithmic scale. The result itself is also given on a logarithmic scale in order to display the wide range in mass for different particle sizes.

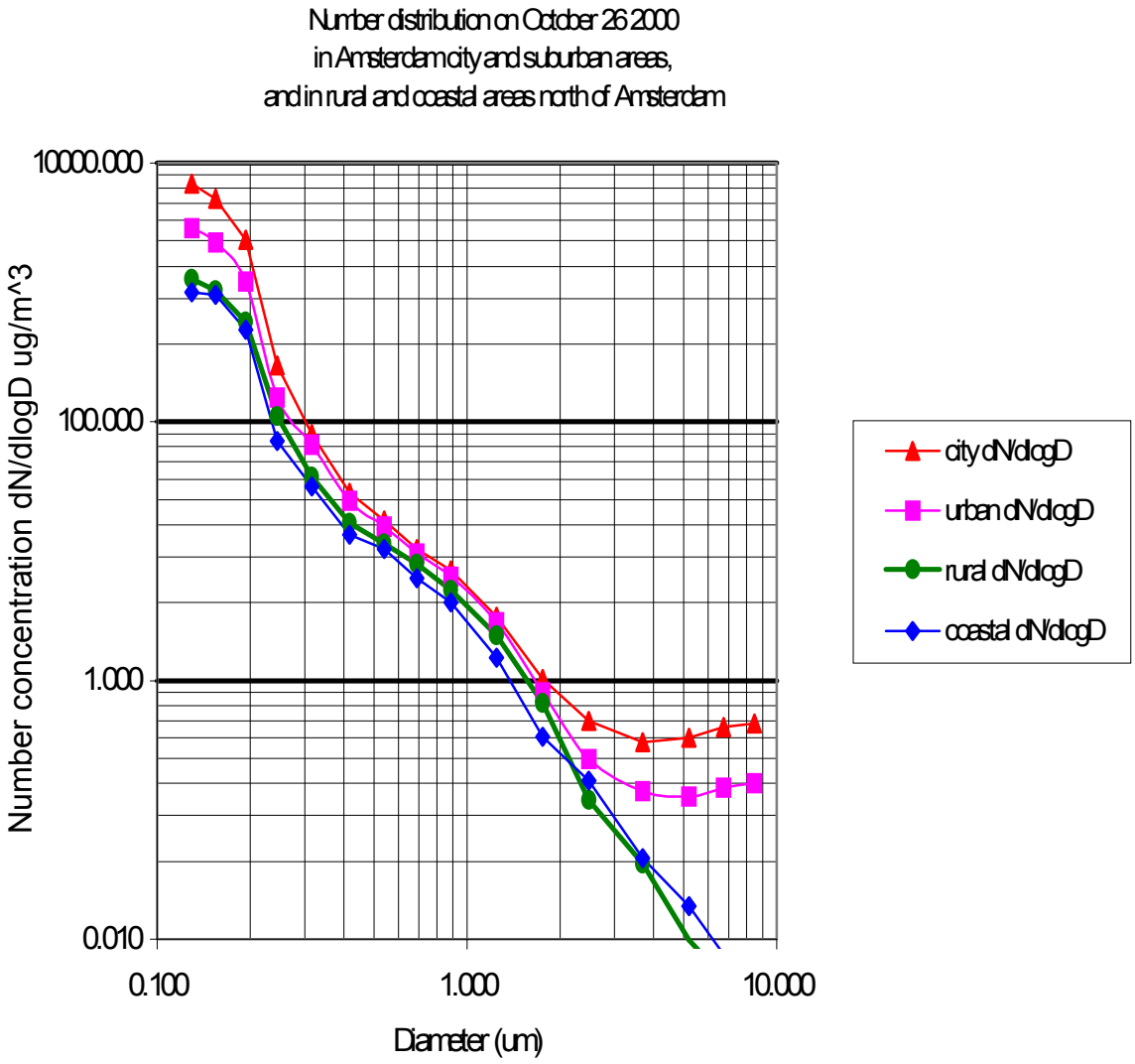

Figure 5.4 *Average number of particles as a function of particle diameter for the four sections.*

The distributions indicate that by far most of the particles are small (below 1 µm, no matter where they are measured. For all particle sizes, numbers are systematically higher in Amsterdam and in suburban quarters. The largest differences in number are observed for particles with diameters less than 0.4 µm. Also, the number of heavy particles is higher in cities and suburban areas.

5.3.2 Mass concentrations measured with an optical particle sizer (LAS-X)

The mass distribution with respect to size as derived from the entire ride on October, 26th is given in figure 5.5[6]. Again, the division in various specific sections has been made. The figure indicates that, while most of the particles in urbanised areas are small (<1µm, see figure 5.4), most of the particle mass is determined by the particles having a diameter larger than 2 µm. This effect is almost absent for particle masses measured in rural and coastal areas.

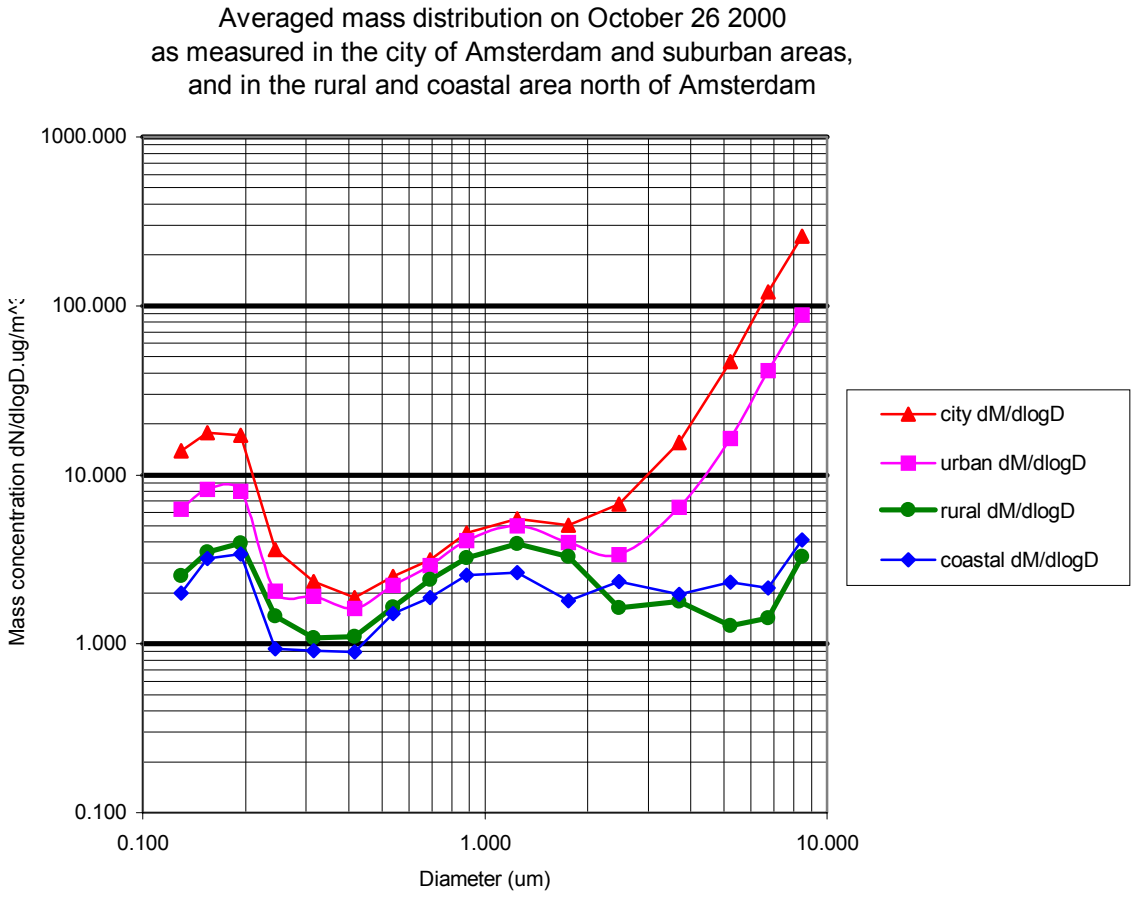

Figure 5.5 *Average particle mass distribution as a function of particle diameter for the four sections during the ride from Petten to Alkmaar on October, 26th, 2000.*

The findings in figure 5.5 can be explored somewhat further by dividing particle mass in the typical mass fractions PM_{10}-$PM_{2.5}$ (PM_{10} minus $PM_{2.5}$), $PM_{2.5}$ - PM_1, and PM_1 (containing all particles that are smaller than 1 µm) for the entire ride; these can be seen in figs. 5.6a-c. In particular, it is interesting to identify those sections where the various mass fractions obtain a maximum value.

[6] The mass per measured size was obtained by multiplying the calculated volume per size with the assumed average density of 1.75 kg/m^3 of the particles. For particles originating from traffic the density might be somewhat lower (towards 1.4 kg/m^3); long-range transport is dominated by by ammonium sulphate and ammonium nitrate having densities of 1.73 and 1.77 kg/m^3, respectively.

The distribution of the fraction PM_{10} - $PM_{2.5}$ is rather uniform; mass concentration values are well between 0 and 40 $\mu g/m^3$ most of the time. Only at urbanised areas like Alkmaar and the inner city of Amsterdam, motorway junctions, and in tunnels the contribution to this mass fraction is substantially higher with levels well above 1000 $\mu g/m^3$.

The variation in concentration for the mass fraction $PM_{2.5}$ - PM_1 is fairly limited: most of the time the measured concentrations are less than 5 ug/m^3. The maxima are attained at restricted locations, i.e. immediately before and after a motorway junction, in a tunnel and at the very centre of Amsterdam.

The variation of the smallest particle mass fraction (PM_1) is more apparent: levels above 20 $\mu g/m^3$ are measured on the A9 motorway, the busy motorway between Haarlem and Amsterdam, the Amsterdam city and in tunnels, in contrast with the two mass fractions above.

6. DISCUSSION

The inhalation of fine dust, whether or not in combination with other air-polluting components, causes damage to human health. Epidemiological research shows that in the Netherlands about 1.000 deaths each year occur due to exposure of fine particulate matter in air (Hoek et al., 1997). Although this number is affected by uncertainty, some of the dying itself in relation to PM is beyond doubt. Furthermore, an increase in hospital admission, asthmatic diseases and lung inflammation during periods with high concentrations of PM has been observed. It is not to be expected that this problem will become less important in future as the traffic, which is assumed to be the main cause still increases in volume. For the EU region the financial burden that is inflicted by PM on human health and, by consequence, the public health care has been estimated on milliards of guilders.

Current European legislation only regulates the mass of PM in ambient air. PM concentrations inside and outside cities are fairly equal (40 $\mu g/m^3$). In recent literature there are indications that other properties of PM (or combination of those properties) causes the damage, such as number and size of particles, kind of surface and chemical composition (heavy metals, carbon). Due to these observation the local Health Administration in Amsterdam has decided to measure, in addition to mass, the number of particles in city air. For decision makers in large cities there is yet no urgent incentive to take measures to improve their local air quality, although the Air Directive on PM is regularly exceeded. The current EU limits will be rephrased in 2003. To this purpose one should look for a parameter (instead of mass) that better represents the risk of occurrence of adverse health effects. From all possible sources, traffic/transport is the one considered as the most important in urban agglomerations. The consequences of an introduction of such a new indicator in the future transport policy of a large city, e.g. the stimulation of clean transport means, should be determined in an early stage.

In first order this research project is directed on the development of accurate measurement of PM concentrations. The improvement of the achievement of the technical instrumentation and the execution of field experiments, in which the improved apparatus is used, are two elements to serve this purpose. The increase in knowledge, publication of results as well as the initiation and expansion of (existing) co-operation with universities and scientific institutes is another major element in this work.

The second goal is to create two relationships that are both necessary for a convincing introduction of alternative transport namely the one between PM (or certain parts of PM) and transport (or certain elements in it) and between PM (or certain components in it) and health effects.

A new directive with respect to PM is expected to have consequences in an urban traffic policy. Due to this development the introduction of clean transport means obtains a better perspective for the near future. The current knowledge with respect to the PM problem and its consequences and, as a result, the absence of a belief in the necessity to design and to implement stronger legislation among policy makers today should then be improved. Especially when it can proven that the adverse health effects are caused by the combustion in cars, buses etc. a convincing argument is there for the introduction of electrical fuel cell (hybrid) cars or trucks.

The introduction of clean transport means in cities might reduce traffic-related health effects. However, the following obstacles exist:

a) In the Netherlands the mass of PM measured in large cities is hardly higher than in rural regions. This is the policy argument against clean but more expensive alternative transport means.

b) The precise mechanisms after inhalation of PM resulting in health problems are not known yet. As a consequence, the current EU Air Directive on PM is debated in the prelude towards 2003 when this directive is reconsidered. This has lead to an abiding attitude.
c) The knowledge of the urgency of the PM problem and of alternative transport mean is fairly limited.
d) Damage and costs due to conventional transport on one hand and the profit of the introduction of alternative transport on the other hand in a large city like Amsterdam are not known yet.

In this report the outcome is described of the work during the last year. The items in this work can be divided into four areas of interest: health, measurements, instrumentation and local policy. In summary, the interesting results for the year 2000 were:

Health
An inventory the findings in literature with concern to the relationships between the negative effects on health and the physical and chemical characteristics of PM in urban air was made. The study reveals that physical properties like surface, solubility and possibly numbers may be important indicators for the observed health effects as well. Chemical compounds that might play a role are metals, soot, PAHSs, endotoxines and acids. The foremost cause for health effects in urban is defined as: the increased presence of toxic, insoluble components (soot, PAHs and metals) that are particularly part of the smaller particles (diameter less than 1 μm) emitted by traffic. The outcome of this study was also meant to be a leading guide in the fulfilment and co-ordination for the remaining items in this project.

Experimental campaigns
Due to the findings summarised above, a study was started to investigate the toxicity of air influenced by emissions of traffic. Measurements of upwind and downwind concentrations along a motorway were compared for differences in chemical composition, size, mass and numbers. The most important but still qualitative finding is that in air influenced by vehicular emissions the toxicity is higher than in air that is not influenced by traffic. The same question will be tried on basis of measurements in Amsterdam in 2001 but than with the further developed MODI-SJAC-IC system.
As a preparation to this, measurements have been executed in 2000 with a mobile unit in order to find those locations where emissions due to traffic can be discerned best from the local urban background levels. It was found that the highest numbers and mass are observed at certain crossings, especially those with traffic lights, on the ring motorway around Amsterdam and in the IJ- and Coentunnel.

Instrumentation
This part is directed on the judgement of existing measurement instruments and further development of measurement techniques with respect to accuracy and automation of apparatus developed within ECN that can be used for measurements in an urban environment. In 1999 the possibilities and impossibilities of the TEOM have been described after tests on mass concentrations of stable and volatile material; this year the Condensation Particle Counter (CPC), an instrument for measuring the number of particles, further explored. A comparative study revealed that the one of the three available types (CPC-3022) is the best option for measurements of PM in urban air. This is because the measurement range of this instrument corresponds most with the size spectrum of particles emitted by traffic. The other two types can better be integrated with an SMPS system.

Policy
The acquired experimental results will serve as input in 2001 and especially in 2002 in discussions with policy makers and also, if possible, as motivation for the introduction of electrically driven cars in urban areas. In order to find out what the general opinion is today is in

co-operation with the unit Policy Studies determined how the various governmental bodies (Ministry, regional, local) handle the problem of PM in concrete situations and how they want to solve this. The most important findings is that the level of knowledge is (still) relatively low, that there is a growing need for information, and that one considers alternative transport as a solution for the long-term. One sees no possibility to accelerate a large-scale implementation of alternative transport by means of policy measures.

APPENDIX

FIJN STOF BELEID EN ALTERNATIEF VERVOER IN NEDERLAND[7]

1.1 Verkeer en luchtkwaliteit

Er zijn verschillende factoren die de luchtkwaliteit bij verkeerssituaties bepalen en die dus oorzaak van knelpunten kunnen zijn (VROM, 1998):

- Verkeersintensiteit. Een van de belangrijkste factoren is het aantal verkeersbewegingen per uur. Hoe drukker een verkeerssituatie is, des te groter is het risico van een knelpunt.
- Doorstroming. De snelheid en continuïteit waarmee het verkeer doorstroomt is eveneens van grote invloed op de luchtkwaliteit. Verkeersopstoppingen kunnen snel leiden tot een knelpunt.
- Fractie vrachtverkeer (inclusief bussen). In een Nederlandse straat bestaat gemiddeld 8% van het verkeer uit vrachtverkeer en bussen. Vrachtverkeer en bussen hebben een grotere negatieve invloed, vooral ook wat betreft fijn stof, op de luchtkwaliteit.
- Wegtype. Van belang hierbij zijn de breedte van de weg, het al dan niet aanwezig zijn van een middenberm, de afstand tussen trottoir en wegas en de hoogte van de bebouwing langs de weg.
- Verversing van lucht. Dit betreft de factoren die van invloed zijn op de verversing van de lucht boven wegen. Omdat het hier in veel gevallen (overhangende) bomen betreft, wordt dit ook wel de bomenfactor genoemd. Bij overhangende bomen kan een tunnel ontstaan, waardoor de luchtverversing belemmerd wordt.
- Parkeerbewegingen. Het aantal parkeerbewegingen in een straat is ook van invloed op de luchtkwaliteit.

1.2 Belangrijke factoren bij beleid

De kosteneffectiviteit van beleidsmaatregelen ten aanzien van het verbeteren van de luchtkwaliteit wordt bepaald door:
- de relatie tussen de kosten van de maatregelen en de effecten op de luchtkwaliteit;
- de effecten van de maatregelen op andere beleidsterreinen, bijvoorbeeld geluid en verkeersveiligheid;
- de allocatie van de kosten over de verschillende beleidsterreinen, oftewel: Wie neemt welk deel voor zijn rekening?

Er zijn verschillende factoren die de haalbaarheid van beleidsmaatregelen kunnen beïnvloeden:
Bestuurlijk draagvlak;
- draagvlak;
- Termijn waarop effecten kunnen worden waargenomen;
- Financieringsmogelijkheden;
- Externe randvoorwaarden die de slaagkansen kunnen beperken (bijv. groeiende mobiliteit, benzineprijzen, tarieven openbaar vervoer).

[7] Deze bijlage is een samenvatting van de belangrijkste resultaten van een in samenwerking met Beleidsstudies uitgevoerd onderzoek. Een uitgebreid rapport (ECN-I--99-001) is verkrijgbaar bij de auteur (A. Groot).

1.3 Verkeersmaatregelen ter verbetering van de luchtkwaliteit

Het Centrum voor energiebesparing en schone technologie (CE) heeft in 2000 een studie uitgevoerd waarin de milieubaten, de kosten en de haalbaarheid van verkeersmaatregelen zijn onderzocht, die kunnen leiden tot een verbetering van de stedelijke luchtkwaliteit (Metz et al., 2000). De resultaten voor een aantal van de onderzochte verkeersmaatregelen staan weergegeven in Tabel A.1.

Tabel A.1 *Haalbaarheid, overheidsinitiatief en effect ten opzichte van de referentiesituatie op de imissie van PM_{10} in stadsstraten van verschillende aanvullende verkeersmaatregelen Bron: Metz et al., 2000*

Verkeersmaatregel	Effect op PM_{10} immissie in stadsstraten t.o.v. de referentiesituatie	Haalbaar-heid	Verdeling initiatief over overheden:		
			Rijk	Prov.	Gem.
Alle stadsbussen op LPG	2,2%	0/+		•	•
Alle streekbussen op LPG	1,1%	0/+		•	•
Roetfilters op alle stadsbussen	2,1%	+		•	•
Roetfilters op alle streekbussen	1,1%	+		•	•
Alle stadsbussen op CNG (aardgas)	2,2%	0		•	•
Alle streekbussen op CNG	1,1%	0		•	•
Stimuleren van elektrische bromfietsen	0,2%	0/+	•		
Verplichten katalysator bromfietsen	0,1%	0/+	•		
Verbetering doorstroming verkeer	0,8%	0/+			•
Aftoppen van de spits	0,2%	-	•		
Bundelen goederenstromen in de stad	0,4%	0			•
Alle bestelwagens op LPG	5,6%	-	•		
Accijnsverhoging brandstoffen	1,4%	-	•		
Accijnsverhoging brandstoffen (extra)	3,0%	-	•		
Kilometerheffing op personenauto's	0,4%	-	•		
Dieselpersonenauto's op LPG	2,5%	0/-	•		
Vrachtwagens op LPG in de stad	1,1%	-			•
Roetfilter op vrachtauto's	3,8%	+	•		

Uit de tabel kan worden geconcludeerd dat het plaatsen van roetfilters op bussen en vrachtwagens, wat betreft de combinatie effect en haalbaarheid, de beste maatregel is.

Volgens het CE bedraagt de PM_{10} immissie in gemiddelde stadsstraten in de referentiesituatie ca. 53 $\mu g/m^3$ in 1995 en ca. 37 $\mu g/m^3$ in 2010. De verwachte norm voor 2010 is 20$\mu g/m^3$, dus is er nog 46% extra reductie vereist ten opzichte van de referentiesituatie. Uit alle onderzochte maatregelen heeft het CE een pakket samengesteld waarmee het maximale reducerende effect kan worden behaald. Dit effect bedraagt voor PM_{10} ca. 16%. Hiermee wordt geconcludeerd dat de norm voor 2010 voor PM_{10} met de onderzochte verkeersmaatregelen **niet** gehaald kan worden. Het aandeel verkeer in de totale stedelijke PM_{10} immissie wordt geschat op 20%. Daarmee concludeert het CE dat 80% van deze 20% kan worden gereduceerd met de onderzochte maatregelen.

1.4 Alternatief vervoer

Alternatief vervoer kan in twee soorten worden ingedeeld. In de eerste plaats kan er gedacht worden aan een alternatieve invulling van het huidige vervoersysteem, in de tweede plaats aan een geheel nieuw vervoersysteem. Voorbeelden van geheel nieuwe vervoersystemen zijn een ondergronds logistiek systeem of een bovengronds automatisch vervoersysteem voor containers (Combiroad) (CCT, 1996, 1997, 1999). Uit verschillende studies is gebleken dat grootschalige invoering van dergelijke vervoersystemen op korte en middellange termijn niet waarschijnlijk is (IPOT, 2000).

Twee alternatieve aandrijfsystemen zijn hybride aandrijving en door brandstofcellen gevoede elektrische aandrijving. Bij hybride aandrijving gaat het om een combinatie van een elektromotor en een verbrandingsmotor. Er zijn verschillende configuraties mogelijk in de wijze waarop de beide motoren gekoppeld zijn. In principe is het mogelijk om met een hybride aangedreven voertuig bepaalde afstanden, bijvoorbeeld binnen stedelijk gebied, emissievrij te rijden.

Hybride aandrijving voor vrachtwagens is alleen zinvol voor goederendistributie in stedelijk gebied. Dit betreft slechts een fractie van het totale vrachtverkeer in Nederland. Bij langeafstandsvrachtvervoer draait de dieselmotor het grootste deel van de tijd op vollast en in een optimaal werkpunt en is dan moeilijk te overtreffen door een hybride systeem (Smokers et al., 1997). Er zijn in enkele Nederlandse steden al proefprojecten uitgevoerd met hybride distributievrachtwagens, onder andere in Veenendaal en Groningen. Er zijn echter nog geen resultaten bekend met betrekking tot een eventuele verbetering van de stedelijke luchtkwaliteit. Op termijn zou grootschalige invoer van hybride distributietrucks in stedelijk gebied mogelijk moeten zijn. Een voorwaarde is dan wel het aanleggen van grote distributiecentra aan de rand van steden, waar al het vrachtvervoer langs gaat. Dit soort ontwikkelingen moeten worden gestimuleerd en zo nodig geïnitieerd door de gemeentes. Een maatregel ter bevordering van het gebruik van hybride distributietrucks is vrijstelling van venstertijden voor deze trucks. Grootschalige inzet van hybride distributietrucks in stedelijk gebied zal in principe voor een verlaging van de fijn stof emissies zorgen. Hoe groot deze verlaging zal zijn en wat het effect zal zijn op de stedelijke fijn stof concentraties is moeilijk te zeggen. Verdergaand onderzoek zou deze vragen mogelijk kunnen beantwoorden.

Brandstofceltechnologie wordt veelal gezien als de energietechnologie voor de toekomst. Bij het proces in de brandstofcel komen geen schadelijke emissies vrij en de brandstof (waterstof) kan op duurzame wijze, met behulp van hernieuwbare energiebronnen, worden geproduceerd. Er wordt op grote schaal gewerkt aan de ontwikkeling van brandstofcelvoertuigen. Grote producenten, zoals Daimler-Chrysler, verwachten binnen vijf jaar een personenauto op brandstofcellen in serieproductie te hebben. Het probleem bij de invoering van brandstofcelvoertuigen is de brandstof. Om de meeste energiebesparing en emissiereductie te bewerkstelligen, kunnen de voertuigen het beste waterstof als brandstof gebruiken. Dit zou echter een geheel nieuwe waterstofinfrastructuur vergen en dat is binnen afzienbare tijd niet te realiseren. De voertuigen kunnen ook vloeibare brandstoffen, zoals methanol of zelfs gewone benzine, gebruiken. Deze worden dan in een reformer in het voertuig omgezet in waterstof. Hierbij is de energiebesparing en de emissiereductie ten opzichte van een gewone verbrandingsmotor echter veel geringer.

In het geval van vrachtwagens is het de vraag of brandstofceltechnologie binnen 10 tot 15 jaar zal kunnen concurreren op energie en emissies met de steeds efficiëntere en schonere dieseltechnologie. Daarnaast zal het nog langer duren tot brandstoftechnologie ook wat betreft kosteneffectiviteit concurrerend zal zijn met dieseltechnologie (Kolke, 1999; Smokers et al., 1997; Bos en Van den Bosch, 1999). Volgens Bos en Van den Bosch (1999) is tegen het jaar 2010 reductie door de inzet van brandstofcelsystemen in vrachtwagens en bussen realiseerbaar

tegen reductiekosten die nog iets hoger liggen dan de reductiekosten op basis van verbeterde dieseltechnologie.

In bussen zijn de kansen voor de implementatie van brandstofcellen op korte termijn het beste. Redenen hiervoor zijn dat bussen gemakkelijk met waterstoftanks kunnen worden uitgerust en dat ze op een centraal punt getankt kunnen worden. Hierdoor is het eenvoudiger om met waterstof als brandstof te werken, waarmee optimale milieuwinst gehaald wordt. Brandstofcelvoertuigen zorgen voor een forse verlaging van de fijn stof emissies. Hoe groot deze reductie is ten opzichte van verbeterde dieselvoertuigen is echter moeilijk te zeggen, evenals wat precies het effect zal zijn op de fijn stof concentraties. De komende tien tot vijftien jaar zullen brandstofcelvoertuigen in ieder geval nog niet op grote schaal bijdragen aan het oplossen van de fijn stof problematiek (bij onveranderd beleid).

1.5 Verantwoordelijkheden verschillende overheden

Voor fijn stof is nog niet duidelijk hoe de verantwoordelijkheden precies komen te liggen. Vooralsnog lijkt het erop dat het Rijk in geval van fijn stof in ieder geval de komende jaren het voortouw zal gaan nemen. Dit in tegenstelling tot de andere luchtverontreinigende stoffen, waarvoor de verantwoordelijkheid steeds meer gedecentraliseerd wordt. Een en ander zal worden vastgelegd in de AMvB inzake fijn stof, die uiterlijk op 19 juli 2001 in werking treedt.

Vooral gemeenten boven 40.000 inwoners hebben te maken met luchtkwaliteitknelpunten bij verkeerssituaties. Gemeentes zijn verantwoordelijk voor de volgende zaken:

- Bewaking van de luchtkwaliteit bij verkeerssituaties;
- Opnemen van luchtkwaliteit in het gemeentelijk milieubeleidsplan;
- Rapportageplicht aan de provincie, eens per vier jaar, bij (bijna) overschrijdingen is jaarlijkse rapportage verplicht. Rapportage geldt alleen voor de daartoe verplichte stoffen CO, NO_x en Benzeen en geschiedt middels standaard rapportageformulieren of verkeersmilieukaarten met een toelichting gericht op lokale knelpunten;
- Opstellen van een meerjarenplan van aanpak luchtkwaliteit verkeerssituaties.
- Gemeentes zijn verantwoordelijk voor het aanleggen en onderhouden van het gemeentelijke wegennet.

Provincies zijn verantwoordelijk voor de volgende zaken:

- De vergunningverlening aan en de bewaking van de luchtkwaliteit rond inrichtingen;
- Het toetsen van gemeentelijke bestemmingsplannen;
- Rapportageplicht aan de Rijksoverheid (VROM) inzake de provinciale luchtkwaliteit, opgebouwd uit de gemeentelijke rapportages;
- Provincies zijn verantwoordelijk voor het aanleggen en onderhouden van het provinciale wegennet.

De Rijksoverheid is verantwoordelijk voor de volgende zaken:

- De landelijke achtergrondniveaus van luchtverontreinigende stoffen;
- Het ‘Schone auto’-beleid. Het merendeel van het wegverkeer is niet gebonden aan een bepaalde regio of stad en valt dus onder de verantwoordelijkheid van het Rijk.
- Rapportage aan de EU inzake de landelijke luchtkwaliteit, opgebouwd uit de verzamelde provinciale rapportages.
- Het Rijk is verantwoordelijk voor het aanleggen en onderhouden van het rijkswegennet.

2. RESULTATEN INTERVIEWS

In dit hoofdstuk worden de resultaten van een aantal afgenomen interviews puntsgewijs beschreven. Er zijn uitgebreide interviews afgenomen met het ministerie van VROM, een drietal provincies, een viertal grote gemeentes en met het GGGD in Amsterdam.

2.1 Ministerie van Volkshuisvesting, Ruimtelijke Ordening en Milieubeheer

2.1.1 Status fijn stof beleid

Fijn stof staat bij VROM de laatste tijd meer in de belangstelling dan vroeger. De belangrijkste oorzaken hiervoor zijn de EU-normstelling en de alarmerende onderzoeksresultaten over de gezondheidseffecten. Er is een herziening van de AMvB Luchtkwaliteit in voorbereiding waarin de EU-normen geïmplementeerd worden. Hiervoor is ook een rondje langs andere landen gemaakt. Nederland loopt hier echter voor op andere landen. De normen voor fijn stof moeten uiterlijk 19 juli 2001 van kracht worden en zijn zodanig scherp dat ze een flinke impuls hebben gegeven aan het nationale luchtkwaliteitsbeleid.

2.1.2 Knelpunt

Van belang voor scherpere normen is de aantoonbaarheid van de noodzaak daartoe. Met andere woorden, er moet voldoende wetenschappelijke kennis en informatie beschikbaar zijn. Hierop wordt later nog even ingegaan. Een voorbeeld van het kennisprobleem is het vaststellen van de indicatieve fijn stof grenswaarde voor 2010 op 20 $\mu g/m^3$. De indicatieve grenswaarde is nu nog niet aan de orde, want zo'n waarde wordt nog niet als een wettelijke grenswaarde vastgelegd. Het vormt een wenselijk lange termijn doel, waar internationaal - en wetenschappelijk, qua onderbouwing - nog veel discussie over bestaat. In de geplande Europese evaluatie in 2003 over deze stofnorm zal op basis van alle nieuwe kennis en inzichten moeten worden beoordeeld of deze norm nog steeds juist is of dat een alternatieve grenswaarde moet worden voorgesteld. Het signaal dat nu van deze indicatieve grenswaarde uitgaat, is dat het bereiken van de norm voor 2005 (40 $\mu g/m^3$, jaargemiddeld) nog niet genoeg is. Het is echter wel duidelijk dat de haalbaarheid van een jaargemiddelde norm voor PM_{10} van 20 $\mu g/m^3$ in 2010 in diverse EU-landen twijfelachtig is. Welke waarde dan eventueel als alternatief kan worden gekozen, is nu nog niet te zeggen.

2.1.3 Maatregelen en instrumenten

Veel maatregelen die gemeentes vanuit andere motivatie (geluidsoverlast, parkeerbeleid) nemen hebben indirect ook effecten op de luchtkwaliteit. Volgens VROM is het zaak om hier zoveel mogelijk van te profiteren, vooral wanneer directe oplossingen niet makkelijk voor handen zijn. Meelifteffecten kunnen al voor een redelijke bijdrage in de verbetering van de luchtkwaliteit zorgen zonder dat er gericht actie is ondernomen.

Om op lange termijn tot een structurele oplossing van het fijn stof probleem in Nederland te komen is een Europese aanpak vereist. Dit vanwege de grote bijdrage van het buitenland in de Nederlandse fijn stofconcentraties.

2.1.4 Alternatief vervoer

Het NMP4 gaat zich minder richten op technologische verbeteringen van huidige technieken omdat is gebleken dat op de lange termijn geen echte oplossingen voor de klimaat- en verzuringproblemen oplevert. Daarom wordt er door VROM voor de lange termijn gemikt op heel nieuwe technologieën zoals elektrisch vervoer met brandstofcellen. Voor de korte tot middellange termijn ziet VROM geen grote rol weggelegd voor elektrisch vervoer, in die zin dat het weinig zal bijdragen aan het verbeteren van de luchtkwaliteit. VROM is wel van mening dat lagere overheden hun eigen wagenpark zo milieuvriendelijk mogelijk moeten invullen, dus

eventueel met alternatieve vervoerswijzen. Naast het geringe effect op de luchtkwaliteit heeft dit vooral waarde als voorbeeldfunctie en signaal naar de burger.

2.1.5 Relatie met andere overheden

Volgens VROM liggen de verantwoordelijkheden voor luchtkwaliteit, ook in geval van serieuze knelpunten, in de eerste plaats bij gemeentes en provincies zelf. Wanneer die het zelf niet kunnen oplossen kunnen ze bij VROM aan de bel trekken, maar dan moeten ze ook zelf met een plan komen. VROM beoordeelt dit dan en kijkt of er middelen zijn om dit plan uit te voeren. Het initiatief ligt dus bij de lagere overheden. VROM houdt zich zelf alleen met het landelijke en internationale plaatje bezig en niet met specifieke zaken. VROM streeft dus naar een gedeeltelijke decentralisatie van het luchtkwaliteitsbeleid. Voor fijn stof wordt vooralsnog echter een andere aanpak gevolgd. VROM is van mening dat het voortouw in het fijn stofbeleid, vanwege de vele onzekerheden en knelpunten, vooralsnog bij het Rijk ligt. Onderlinge afstemming met de andere overheden wordt hierbij wel noodzakelijk geacht.

Voor wat betreft de grenswaarden voor fijn stof worden de waarden overgenomen die in de Europese dochterrichtlijn zijn vastgelegd. Gezien de overschrijding op vrij grote schaal die op dit moment nog plaats vindt van de norm waarin in 2005 dient te worden voldaan, in combinatie met de hoge achtergrondconcentratie en een overheersende bijdrage uit het buitenland (t.o.v. de NL-bijdrage) wordt het weinig zinvol geacht bij de gemeentelijke en provinciale overheden het initiatief te leggen om plannen te ontwikkelen voor oplossingen voor die situaties waar de norm overschreden wordt.

2.2 Provincies

2.2.1 Status fijn stof beleid

Over het algemeen staat luchtkwaliteit duidelijk op de provinciale politieke agenda, zij het niet zeer hoog. Een uitzondering vormt de provincie Zuid Holland. Dit wordt mede veroorzaakt door de specifieke situatie in Zuid Holland, namelijk de aanwezigheid van het Rijnmondgebied. De aandacht voor luchtkwaliteit in provincies is overigens de laatste jaren groeiende. In het luchtkwaliteitsbeleid wordt fijn stof in de meeste gevallen nog niet als afzonderlijk aandachtspunt meegenomen. Enkele provincies doen, vooruitlopend op de herziening van de AMvB Luchtkwaliteit, al wel wat onderzoeken om te kijken hoe fijn stof in het beleid meegenomen zou moeten worden. De aandacht voor fijn stof leeft in enkele provincies al langere tijd, men doet er beleidsmatig echter nog weinig tot niets mee. In veel provincies wordt fijn stof pas een beleidsonderwerp wanneer de nieuwe AMvB ingaat. Veel provincies nemen derhalve een afwachtende houding aan.

De belangrijkste motivatie voor het fijn stof beleid voor provincies is het gezondheidseffect van fijn stof. De Europese richtlijn en de komende AMvB worden echter ook in alle gevallen genoemd, enerzijds als motivatie, anderzijds als middel. Er is zodoende een duidelijke tweedeling waar te nemen inzake het provinciale fijn stof beleid. Er zijn een aantal provincies die een voortrekkersrol vervullen en zich de gezondheidseffecten van fijn stof zodanig aantrekken dat ze ondanks het gebrek aan ondersteunende wetgeving toch al langere tijd proberen fijn stof beleid te maken. Bij de meeste andere provincies wordt de eerder vermelde afwachtende houding ingenomen en gaat men pas fijn stof beleid maken op het moment dat duidelijkheid ontstaat over de inhoud van de komende AMvB.

Beleidsmaatregelen worden over het algemeen gekozen op basis van hun rendement op de gezondheidseffecten, alsmede hun kosteneffectiviteit. Kosteneffectiviteit speelt vooral een rol bij de vergunningverlening voor grote inrichtingen. Een derde zeer belangrijk punt bij het kiezen van beleidsmaatregelen is draagvlak. Bij provincies gaat het dan voornamelijk om bestuurlijk draagvlak, maar daarbij speelt het maatschappelijke draagvlak indirect natuurlijk een rol, evenals kennis over het onderwerp.

2.2.2 Knelpunten

Als belangrijkste problemen voor provincies worden genoemd de beperkte wettelijke mogelijkheden om luchtkwaliteit aan te pakken, de beperkte kennis inzake de fijn stof problematiek en de niet optimale informatiestromen tussen overheden onderling en overheden en de onderzoekswereld. Veel provincies, vooral de meer vooruitstrevende, hebben behoefte aan meer wettelijke mogelijkheden om beleid op gebied van luchtkwaliteit te voeren. Het is voor provincies zeer moeilijk om bedrijven aan te pakken wanneer deze aan alle wettelijke regels voldoen, terwijl het maatschappelijk wenselijk kan zijn om extra maatregelen te nemen.

De problematiek rond fijn stof is nog relatief onbekend. Waar moet het beleid op worden gericht: massa, deeltjesgrootte of deeltjessamenstelling? Een lagere overheid kan bovendien weinig uitrichten tegen de achtergrondconcentratie van fijn stof. Hoe ver moet een provincie gaan met maatregelen met betrekking tot fijn stof wanneer bekend is dat het effect van fijn stof in binnenlucht in geval van roken een factor 400 hoger is dan dat van fijn stof door verkeer? Aan de andere kant is er geen concentratiedrempel van fijn stof bekend waaronder geen gezondheidseffecten meer optreden. Omdat het kennisniveau omtrent fijn stof zo laag ligt is het voor provincies moeilijk om een richting te kiezen. Om deze reden wachten veel provincies met eigen initiatieven inzake fijn stof beleid totdat VROM een landelijke koers heeft uitgezet.

2.2.3 Maatregelen en instrumenten

Een aantal provincies zijn voornemens een toetsingskader te ontwikkelen voor de toekenning van vergunningen en het toetsen van bestemmingsplannen. Hierin zou luchtkwaliteit, waaronder fijn stof, dan ook opgenomen moeten worden. Het effect van advies en sturing naar gemeentes valt of staat met de welwillendheid van gemeentes om hieraan gehoor te geven. Over het algemeen treden hier weinig problemen op. Bovendien hebben provincies vaak nog wat financiële middelen achter de hand waarmee de sturing gestimuleerd kan worden, in de zin dat gemeentes (extra) geld kunnen krijgen wanneer ze in de lijn van het provinciaal beleid handelen.

2.2.4 Alternatief vervoer

Provincies kunnen weinig doen aan stimuleren of promoten van alternatief vervoer. Hiervoor hebben provincies te weinig invloed op verkeer en vervoer en bovendien hebben provincies nauwelijks een eigen wagenpark waarmee een voorbeeldfunctie verricht zou kunnen worden. Over het algemeen heerst het idee dat er van grootschalige invoer van elektrisch vervoer op korte tot middellange termijn weinig te verwachten is. Een veel genoemde opmerking is dat de huidige dieselmotoren nog steeds veel schoner worden en dat dieseltechnologie, vooral in het vrachtvervoer, behoorlijk superieur is. De fijn stof problematiek wordt vooral veroorzaakt door het vrachtverkeer dus ten aanzien van fijn stof wordt van elektrisch vervoer niet veel verwacht. Op lange termijn ziet men wel mogelijkheden voor brandstofceltechnologie, maar ook hier geldt dat implementatie juist bij het vrachtverkeer het meest moeilijk is.

2.2.5 Relatie met andere overheden

Volgens een aantal provincies zou VROM een meer coördinerende rol op zich moeten nemen dan nu het geval is. VROM zou de spin in het web moeten zijn tussen de EU enerzijds en de lagere overheden anderzijds. Op dit moment is VROM echter bezig om het luchtkwaliteitsbeleid steeds meer te decentraliseren. Voor fijn stof zal de decentralisatie, vanwege de vele onzekerheden en hoge achtergrondconcentraties in Nederland, echter nog even op zich laten wachten. De vooruitstrevende provincies vinden dat ze zelf veel verantwoordelijkheid zouden moeten hebben inzake het fijn stof beleid. Nauw overleg met gemeentes is hierbij onontbeerlijk, zeker om regionale knelpunten goed in beeld te brengen. Samen met gemeentes zouden zo probleemgerichte oplossingen moeten worden gezocht. Een belangrijke rol voor provincies hierbij is er zorg voor te dragen dat er geen afwenteling van de problemen plaatsvindt van de ene gemeente naar de andere.

Op dit moment is het overleg en de samenwerking met gemeentes in veel gevallen niet zo intensief als idealiter voor het bovenstaande nodig is. De intenties zijn er meestal wel, in de praktijk komt er echter nog te weinig van. Het gebeurt nog veel dat de verplichte rapportage het enige contact is tussen gemeentes en provincies op het gebied van luchtkwaliteit.

Wanneer er door de gemeentes knelpunten worden gerapporteerd, nemen provincies dat in de huidige situatie meestal voor kennisgeving aan. Terugkoppeling vindt nog nauwelijks plaats, mede doordat daar wettelijk weinig mogelijkheden voor provincies liggen. Bovendien gaan provincies er vanuit dat gemeentes zelf doen wat ze kunnen om knelpunten op te lossen. Het eventueel opleggen van sanctiemaatregelen wanneer knelpunten niet opgelost worden lijkt de provincies dan ook niet zinvol. Zeker in geval van fijn stof heerst sterk de gedachte dat de nieuwe grenswaarden zeker overschreden gaan worden en dat gemeentes daar vanwege de hoge achtergrondconcentraties weinig aan kunnen doen.

2.3 Gemeentes

2.3.1 Status fijn stof beleid

Bij de meeste grote gemeentes staat luchtkwaliteit wel in de belangstelling, maar wordt er nog niet veel direct, structureel beleid op gevoerd. Meestal wordt het meegenomen in het verkeers- of ruimtelijke ordeningsbeleid of worden er ad hoc beslissingen genomen. Dit laatste is meestal het geval bij acute knelpunten die vanuit de bevolking worden aangedragen. De meeste gemeentes rapporteren volgens de wet aan de provincie en hebben een gemeentelijke verkeersmilieukaart, waar luchtkwaliteit in is meegenomen.

Er is over het algemeen vooral aandacht voor de rapportageplichtige stoffen, waartoe fijn stof niet behoort. Fijn stof is bij veel gemeenten dan ook pas een issue wanneer de AMvB voor fijn stof ingaat. Ze nemen wat betreft beleid dan ook een afwachtende houding aan. Er zijn echter ook enkele vooruitstrevende gemeentes, waar fijn stof al wel zeer belangrijk wordt gevonden. Ook deze gemeentes zijn echter gedwongen te wachten op het ingaan van de komende AMvB, voordat ze fijn stof beleid kunnen gaan uitvoeren. Ze zijn, in tegenstelling tot de afwachtende gemeentes, meestal wel nu al bezig met beleidsoriëntatie ten aanzien van fijn stof.

Het luchtkwaliteitsbeleid in gemeentes wordt in de meeste gevallen gemotiveerd door de regelgeving enerzijds en door de bevolking anderzijds. Ook de politieke “kleur” van de gemeenteraad en het enthousiasme van milieuambtenaren kunnen van invloed zijn op de mate van belangstelling voor luchtkwaliteit binnen gemeentes.

Naast de AMvB spelen ook de gezondheidseffecten een grote rol als motivatie voor fijn stof beleid. Het verminderen van deze effecten wordt over het algemeen als belangrijkste speerpunt gezien voor fijn stof beleid, belangrijker dan het halen van de grenswaarde. De algemene gedachte is vaak dat overschrijdingen onvermijdelijk zijn, maar dat de bevolking hier zo weinig mogelijk last van moet hebben. Een overschrijding in de middenberm van een drukke doorgangsweg zou dus gedoogd kunnen worden, maar een overschrijding in een drukke woonwijk moet worden aangepakt.
In de meeste gemeentes is verkeer de belangrijkste bron van fijn stof, daar zal het meeste beleid dan ook op gericht gaan worden.

In een beperkt aantal gevallen is luchtkwaliteitsbeleid gebaseerd op klachten die rechtstreeks vanuit de bevolking zijn aangedragen. Het gaat dan meestal om specifieke, vaak acute, knelpunten, waar over het algemeen ad hoc beleid op wordt gevoerd.

2.3.2 Knelpunten

Het belangrijkste knelpunt voor gemeentes is het feit dat ze nauwelijks iets kunnen doen aan hun lokale fijn stof concentratie, vanwege de hoge achtergrondconcentratie. Dit feit brengt twee

problemen met zich mee. Enerzijds is het voor de bevolking moeilijk te begrijpen dat een stof met bewezen schadelijke gezondheidseffecten door hun gemeente zo moeilijk aangepakt kan worden. Anderzijds is het voor de betrokken milieuambtenaar moeilijk om bestuurlijk draagvlak te krijgen voor beleidsmaatregelen waarvan het maar de vraag is in hoeverre ze bijdragen aan het oplossen van overschrijdingen. Over het algemeen leeft bij de betreffende milieuambtenaren wel het idee dat de gemeente wat betreft fijn stof moet doen wat zij kan, mede als signaal van welwillendheid naar de burgers. Andere betrokken afdelingen, zoals verkeer en ruimtelijke ordening hebben hier echter nogal eens moeite mee, zeker als de totaal te besteden financiële middelen beperkt zijn.

2.3.3 Maatregelen en instrumenten

In veel gevallen is verkeersbeleid en ruimtelijke ordeningsbeleid gekoppeld, bijvoorbeeld bij het inrichten van nieuwe woonwijken, het aanleggen van parkeergelegenheden en het herinrichten van stadscentra. Veel gebruikte maatregelen zijn het autoluw maken van stadscentra, een verkeerscirculatieplan en het aanleggen van grote centrale parkeerplaatsen in combinatie met hoge parkeertarieven in het centrum en goed geregeld vervoer van en naar de parkeerplaatsen.

De mogelijkheden binnen de vergunningverlening zijn beperkt door de wettelijke regelingen. Een gemeente kan niet meer eisen dan in de regels staat. Gemeentes hebben via de vergunningverlening wel invloed op de plaats waar bedrijven zich vestigen en kunnen op die manier dus zorgen dat ‘vervuilende’ bedrijven niet in dicht bewoonde gebieden komen. Voor fijn stof gaat het dan vooral om op- en overslagbedrijven of andere bedrijven die veel vrachtverkeer genereren.

Bovenstaande maatregelen worden overigens maar in zeer beperkte mate expliciet genomen vanuit de motivatie om de luchtkwaliteit te verbeteren. Zolang de AMvB nog niet van kracht is, speelt fijn stof als afzonderlijke component bij het nemen van dergelijke maatregelen helemaal nauwelijks een rol. In de meeste gevallen worden maatregelen in de eerste plaats genomen om het verkeer te reguleren. Zaken als verkeersveiligheid, geluidshinder en verkeersdrukte worden vaak belangrijker gevonden dan luchtkwaliteit. Effecten op de luchtkwaliteit spelen wel een rol, maar geen beslissende. In veel gevallen moet de luchtkwaliteit het hebben van de meelifteffecten, die overigens fors kunnen zijn. Fijn stof moet het op dit moment alleen maar hebben van meelifteffecten.

2.3.4 Alternatief vervoer

De meeste grote gemeentes hebben een redelijk groot wagenpark in hun beheer of onder hun invloed, zoals dat van de gemeentelijke diensten, de vuilophaaldienst en het stedelijk openbaar vervoer. Bij de meeste gemeentes heeft men de intentie om deze wagenparken zo milieuvriendelijk mogelijk in te vullen, eventueel met behulp van alternatieve aandrijftechnieken. In de praktijk wordt er echter, vooral voor de zwaardere voertuigen, in de meeste gevallen gekozen voor schone conventionele technieken zoals schone dieselvoertuigen met roetfilter of LPG-aangedreven voertuigen. Voor de kleinere voertuigen, voor bijvoorbeeld de plantsoendienst, wordt wel in meer gevallen gekozen voor elektrisch aangedreven voertuigen. Dit wordt in de eerste instantie gedaan vanuit milieuoogpunt, maar ook vanuit het oogpunt van het geven van het goede voorbeeld naar de burgers.

Op de lange termijn zien de meeste gemeentes wel een grotere rol weggelegd voor alternatief vervoer. Vooral de brandstofceltechnologie wordt daarbij veel genoemd. Een aantal gemeentes heeft al projecten lopen met hybride stadsbussen en hybride distributievrachtauto’s. In Amsterdam gaan binnenkort een aantal brandstofcelbussen ingezet worden in het stedelijk openbaar vervoer. Grootschalige invoer van dergelijke technieken zullen volgens de meeste gemeentes echter zeker nog 15 jaar op zich laten wachten.

2.3.5 Relatie met andere overheden

Gemeentes hebben in de eerste instantie te maken met de provincie. Een aantal vooruitstrevende gemeentes die participeren in landelijke werk- of stuurgroepen heeft in dat kader rechtstreeks contact met VROM, maar in het dagelijkse werk is dat contact er in principe niet.

De contacten tussen gemeentes en provincies zijn over het algemeen goed. Op het gebied van luchtkwaliteit wordt het initiatief meestal aan de gemeentes overgelaten. Zoals al eerder vermeld doen provincies in de meeste gevallen niet aan terugkoppeling naar gemeentes op basis van eventuele gerapporteerde knelpunten. Wanneer gemeentes echter expliciet bij de provincie aankloppen vanwege een knelpunt is deze meestal wel bereid om (financiële) medewerking te verlenen.

De meeste gemeentes voelen zich sterk verantwoordelijk voor de luchtkwaliteit in hun gemeente en zijn bereid om zelf beleidsinitiatieven te ontplooien. Nauw overleg met de provincie wordt hierbij zeer belangrijk gevonden. De decentralisatie van het luchtkwaliteitsbeleid die door VROM in gang is gezet wordt door de meeste gemeentes positief beoordeeld, mits VROM zorg blijft dragen voor het aangeven van duidelijke grote lijnen. Ter invulling hiervan worden specifiek op gemeentes gerichte publicaties zoals de *Wegwijzer verkeerssituaties* ten zeerste toegejuicht.

3. CONCLUSIES/AANBEVELINGEN

- Het kennisniveau omtrent fijn stof is bij de verschillende overheden dusdanig laag dat het zeer moeilijk is om op korte termijn een structureel en effectgericht fijn stof beleid te (gaan) voeren. Dit betekent dat enerzijds meer fijn stof onderzoek nodig is en anderzijds meer doelgerichte kennisverspreiding, bijvoorbeeld in de vorm van een periodieke publicatie waarin onderzoeksinstituten en hogere overheden gezamenlijk de nieuwste inzichten bespreken.
- Gemeentes hebben voor complexe beleidsonderwerpen behoefte aan handreikingen van VROM of de provincie, met daarin informatie en voorbeelden over signaleren, analyseren en oplossen van concrete (beleids-)knelpunten. Voor gemeentes zijn dit soort handreikingen nuttig ter beperking van uitgebreid eigen onderzoek en om een kader voor beleid te scheppen. Voor VROM en de provincies zijn dit soort handreikingen effectieve instrumenten om gemeentes te sturen in de door hen gewenste richting terwijl de uiteindelijke verantwoordelijkheid toch bij de gemeentes ligt.
- In principe wil VROM wat betreft luchtkwaliteitsbeleid een verdergaande decentralisatie doorvoeren. Het is nog onzeker of dit in geval van fijn stof op korte termijn zal gebeuren. De redenen hiervoor zijn de vele onzekerheden waarmee de fijn stof problematiek nog omgeven is, het feit dat de norm op grote schaal overschreden wordt, de hoge achtergrondconcentratie en de grote bijdrage van het buitenland in de Nederlandse fijn stofconcentratie. VROM is derhalve van mening dat het voortouw in het fijn stofbeleid vooralsnog bij het Rijk ligt. Onderlinge afstemming met de andere overheden wordt hierbij wel noodzakelijk geacht.
- Een aantal vooruitstrevende provincies en gemeentes zijn vooruitlopend op de AMvB inzake fijn stof zelf al bezig met het uitstippelen van fijn stof beleid. De rest van de provincies en gemeentes neemt een meer afwachtende houding aan inzake fijn stof en zal pas na het ingaan van de AMvB fijn stof beleid ontwikkelen.
- Verbeteren van de luchtkwaliteit vormt meestal niet het hoofddoel van gemeentelijke beleidsmaatregelen in het verkeer. Regulatie van verkeersstromen en het terugdringen van geluidsoverlast zijn nogal eens belangrijker. Maatregelen in dat kader hebben echter in de meeste gevallen ook gunstige effecten op de luchtkwaliteit.
- Het ministerie van VROM zal niet veel eerder dan 19 juli 2001 duidelijkheid geven over hoe het fijn stof beleid landelijk ingevuld gaat worden. Uiterlijk op die datum moet de nieuwe EU-richtlijn inzake fijn stof in de Nederlandse wetgeving geïmplementeerd zijn.
- Voor een structurele lange termijnoplossing van het fijn stof probleem is een Europese aanpak vereist. Alleen dan kunnen de landelijke achtergrondconcentraties omlaag worden gebracht. Locale overheden zouden dit als vrijbrief aan kunnen grijpen om zich niet aan de regels te hoeven houden.
- De meeste geïnterviewde beleidsmedewerkers voor luchtkwaliteit verwachten dat de door de EU ingestelde indicatieve PM_{10}-richtlijn voor 2010 (20 $\mu g/m^3$) niet zal worden gehaald. Uit onderzoek van het CE blijkt dat met verkeersmaatregelen 16 van de voor de indicatieve richtlijn benodigde 46% PM_{10}-reductie kan worden gerealiseerd. Het gaat hierbij om maatregelen die onder andere aangrijpen op voertuigtechnische aspecten, de brandstofmatrix, de afwikkeling van verkeer, de modal split, de kosten, de ruimtelijke ordening en de vermindering van het aantal kilometers.
  Opmerkelijk is dat aan het niet halen van wettelijke grenswaarden voor luchtverontreinigende stoffen of het niet oplossen van gerapporteerde knelpunten zijn geen consequenties of sancties verbonden. Dit geldt voor alle overheden.
- Er wordt met betrekking tot luchtkwaliteitsbeleid en voor zover van toepassing tot fijn stof beleid bij provincies en gemeentes maar weinig volgens de ideale beleidscyclus gewerkt. De intentie tot het expliciet werken volgens beleidscycli is wel aanwezig, maar in de praktijk worden veel beslissingen ad hoc genomen. De belangrijkste redenen hiervoor zijn tijdgebrek en het gebrek aan financiële middelen.

- Alternatief vrachtvervoer wordt door de verschillende overheden gezien als lange termijn oplossing. Voor de korte termijn wordt meer gezien in schone dieseltechnologie, eventueel in combinatie met roetfilters. Voor stadsbussen zou de implementatie van alternatieve aandrijvingen sneller kunnen gaan, maar ook dit zal naar verwachting niet voor 2010 grootschalig gebeuren. De verschillende overheden zien nauwelijks mogelijkheden om de grootschalige implementatie van alternatief vervoer met behulp van beleidsmaatregelen te versnellen.

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