On health risks of ambient PM in the Netherlands

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Netherlands Aerosol Programme

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ABSTRACT

Particulate Matter (PM) in the ambient air can lead to health effects and even to premature mortality. This result has been found in a score of epidemiological studies, but its cause is not yet clear. It is certain, however, that these effects are so serious and so extensive that further action is warranted. In the scientific literature ambient aerosols are known as PM, short for Particulate Matter. Depending on the diameter or size of the particles, they are termed PM_{10} (for particles with diameters of up to approximately 10 micrometres) or $PM_{2.5}$ (for those less than 2.5 micrometres in diameter). One micrometre is a thousandth of a millimetre. Humans inhale particles smaller than 10 micrometres, which end up deep in our airways.

Recent studies have presented well-founded assumptions concerning the biological mechanisms involved and the groups of people that are probably more susceptible to PM. Particulate Matter is a generic term for a complex mixture of large and small airborne particles. However, the causal factors within this complex mixture are difficult to disentangle and have not yet been identified. The second Section of this report looks at the different types of PM, their atmospheric behaviour and the methods of measuring them. The health effects associated with PM are also presented. Section 3 discusses the most recent epidemiological, toxicological and human clinical findings and their mutual relationships.

On the basis of epidemiological studies it has been estimated that in the Netherlands some 1,700 to 3,000 people per year die prematurely as a result of inhaling ambient PM. These figures reflect only the effects of acute exposure to air pollution. If the long-term effects of chronic exposure are taken into account, premature mortality could affect 10,000–15,000 people a year in the Netherlands. These last estimates for chronic exposure are more uncertain, because chronic effect studies are much fewer in number. The estimate of the chronic effects was based on foreign studies, which are not completely comparable with the Dutch situation.

Section 4 gives an overview of the most recent information relating to sources of PM and emissions in the Netherlands, while the last Section presents a critical evaluation of the current and future EU standards.
It is recommended that PM_{10} be retained as a standard for the time being, as it covers the effects of both fine and coarse particles. In view of the emerging evidence implicating fine particles in health effects, it is recommended that a standard for fine PM and/or a source-related fraction be developed as well.

Even with PM concentrations well below European Union (EU) standards, people's health will still be affected because no threshold has been found for the occurrence of health effects. PM is a complex mixture containing fractions that are to a greater or lesser extent health-relevant. This differentiation in potency has profound implications for an efficient and effective reduction of health impacts through PM emission abatement.

PM abatement can be justified by the precautionary principle. Further source- oriented actions could focus on reduction of the total PM_{10} aerosol mass or, first of all, on those PM fractions that are expected to be more health-relevant. This last option is preferred. These fractions are probably transport-related (diesel soot) and, more generally, combustion-related primary PM emissions. Abatement should therefore focus on these sources. In this respect, the abatement of uncontrolled shipping emissions has been identified as one of the more cost-effective control options. Abatement of other combustion sources such as industrial combustion, wood burning in fireplaces, and off-road machinery are also possible, but less cost-effective.

The European Union has decided on two standards for PM, a daily and an annual average value. The current EU standards for daily and annual average values are not equivalent, as was originally intended. In the Netherlands the following options are equivalent to the EU annual standard of 40 $\mu g/m^3$: a daily level of 50 $\mu g/m^3$ with 80 exceedances (while the EU allows 35 exceedances) or a daily level of 100 $\mu g/m^3$ with 7 permitted exceedances per year. For practical reasons a daily standard of 100 $\mu g/m^3$ is preferred. Although the EU has proposed two standards for PM, there are several arguments that only one standard would suffice – annual mean concentrations being the best choice. However, for reasons of communication to the public daily standards may be appropriate.

Compliance with the annual average EU standard seems feasible for PM_{10} in the Netherlands in 2005, although local exceedances at 'hot spots' cannot be ruled out. Compliance in 2010 with the indicative annual average EU standard of 20 $\mu g/m^3$ is not feasible, even at high cost. Expectations are that there will still be 36–40 exceedances per year of the EU daily standard of 50 $\mu g/m^3$ even after all planned abatement measures (Current Legislation of Emissions (CLE)) have been taken in 2010. Therefore, compliance with the current EU daily standards for 2005 and 2010 does not seem feasible in the Netherlands and adverse health effects will continue to occur.

SAMENVATTING

Fijn stof in de lucht kan leiden tot gezondheidsklachten en zelfs vroegtijdige sterfte. Dat blijkt uit een honderdtal epidemiologische studies. Hoe die effecten precies ontstaan is nog niet duidelijk. Vast staat echter dat de gezondheidseffecten door fijn stof zo ernstig en omvangrijk zijn dat nadere actie geboden is. In de wetenschappelijke literatuur staat fijn stof bekend als 'deeltjesvormige luchtverontreiniging' (Engels: Particulate Matter, ofwel PM.). Afhankelijk van de doorsnee van de stofdeeltjes wordt gesproken van PM_{10} (voor deeltjes met een doorsnee tot 10 micrometer) of $PM_{2,5}$ (doorsnee tot 2,5 micrometer). Een micrometer is een duizendste millimeter. Deeltjes kleiner dan 10 micrometer worden door mensen ingeademd en dringen door in de luchtwegen.

Dankzij recente studies zijn er gegronde vermoedens over de biologische mechanismen die in het spel zijn en welke groepen mensen waarschijnlijk gevoelig zijn voor blootstelling aan fijn stof. Maar aangezien 'fijn stof ' een verzamelnaam is voor een complex mengsel van allerhande grote en kleinere stofdeeltjes in de luchtverontreiniging blijft het lastig om oorzakelijke verbanden te ontrafelen. In hoofdstuk 2 van dit rapport komen de verschillende fijn stof deeltjes, hun onderlinge wisselwerking in de atmosfeer en de diverse meetmethoden aan bod. Ook wordt een overzicht gegeven van de gezondheidsklachten die fijn stof kan veroorzaken. In hoofdstuk 3 worden de nieuwste epidemiologische, toxicologische en medische inzichten in onderlinge samenhang besproken.

Op grond van epidemiologische studies wordt geschat dat in Nederland jaarlijks zo'n 1700 tot 3.000 mensen vroegtijdig overlijden door het inademen van fijn stof. En dan hebben we het alleen nog over de acute gevolgen van blootstelling aan luchtverontreiniging. Nemen we ook de lange-termijneffecten van chronische blootstelling aan fijn stof in beschouwing, dan zouden in Nederland mogelijk zelfs 10.000 tot 15.000 mensen jaarlijks vroegtijdig overlijden. De laatste schattingen zijn met meer onzekerheid omgeven, aangezien chronische effecten in minder studies gekwantificeerd zijn dan acute effecten. Bovendien is de berekening het resultaat van een vertaalslag van internationale onderzoeksresultaten naar de Nederlandse situatie en die is niet helemaal vergelijkbaar.

Hoofdstuk 4 van dit rapport geeft een overzicht van de meest recente informatie over bronnen en emissies van fijn stof in Nederland. Aansluitend wordt in hoofdstuk 5 de huidige en toekomstige Europese normstelling kritisch beoordeeld.
Aanbevolen wordt om voorlopig PM_{10} te blijven hanteren als Europese standaard voor luchtverontreiniging door grove èn fijnere stofdeeltjes. Daarnaast zou er voor het fijnste stof een aparte normstelling of een meer brongerichte normstelling ontwikkeld moeten worden omdat er steeds meer aanwijzingen komen dat kleinere stofdeeltjes de gezondheid bedreigen.

Overigens is nooit aangetoond dat de gezondheidseffecten pas boven een bepaalde drempelwaarde optreden. Zelfs van fijn stof concentraties ver onder de huidige Europese normen zijn gezondheidseffecten in de bevolking te verwachten. Fijn stof is een complex mengsel van allerlei fracties die meer of minder van belang zijn voor de gezondheid. Die verschillen in toxische potentie wegen zwaar mee bij een doeltreffend emissiebeleid.

Bestrijding van de uitstoot van fijn stof valt te rechtvaardigen vanuit het voorzorgbeginsel. Door verdere brongerichte maatregelen kan men de totale massa PM_{10} aërosol in de luchtverontreiniging terugdringen, of eerst die fracties aanpakken die vermoedelijk het meest relevant zijn voor de gezondheid. Waarschijnlijk behoren tot de relevante fracties het dieselroet uit de vervoerssector en fijn stof afkomstig van overige verbrandingsprocessen. Dergelijke bronnen verdienen prioriteit in het beleid voor uitstootbeperking van fijn stof. Bestrijding van de ongecontroleerde scheepvaartemissies blijkt bijzonder kosten-effectief. De aanpak van andere verbrandingsprocessen, zoals industriële verbranding, open haarden en mobiele werktuigen is ook mogelijk, maar minder kosteneffectief.

De EU heeft voor fijn stof twee normen vastgesteld, namelijk een dag- en een jaargemiddelde. Deze beide normen zijn niet gelijkwaardig, hoewel dat oorspronkelijk wel de bedoeling was. De Europese jaargemiddelde PM_{10} norm bedraagt 40 microgram fijn stof per kubieke meter lucht ($\mu g/m^3$). In Nederland kunnen we dat vertalen naar een dagelijkse norm van 50 $\mu g/m^3$ met 80 toegestane overschrijdingen per jaar (terwijl de EU-norm maar 35 overschrijdingen toestaat) of een dagelijkse norm van 100 $\mu g/m^3$ met 7 toegestane overschrijdingen per jaar. Om praktische redenen verdient die laatste norm de voorkeur. Overigens zijn er goede argumenten om maar één norm, en dan liefst een jaargemiddelde, te hanteren. Een daggemiddelde norm kan echter van pas komen bij publieksvoorlichting.

In 2005 lijkt de jaargemiddelde EU norm van 40 $\mu g/m^3$ voor fijn stof in Nederland in het algemeen haalbaar. Lokale overschrijdingen op 'hot spots' zijn echter niet uit te sluiten. In 2010 is de indicatieve jaargemiddelde waarde van 20 $\mu g/m^3$ in Nederland echter niet haalbaar, zelfs niet tegen hoge kosten. Zelfs als in 2010 alle voorgenomen stofbestrijdingsmaatregelen zijn uitgevoerd zullen vermoedelijk nog steeds 36 tot 40 maal per jaar daggemiddelde concentraties boven de 50 $\mu g/m^3$ voorkomen. De dagelijkse EU normen voor 2005 en voor 2010 lijken voor Nederland dan ook niet haalbaar en gezondheidseffecten zullen blijven bestaan.

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SUMMARISED CONCLUSIONS

Assessment of PM health risks

Epidemiological studies present worldwide evidence for particulate matter (PM) associated serious health effects in the general population, which may lead to hospital admissions and premature mortality. Dutch observations are in line with the international scientific literature. In spite of the ongoing scientific debate and prevailing uncertainties concerning the quantification of acute and chronic health effects, the overall conclusion is that PM-associated health effects are so extensive and serious that further action is warranted.

Epidemiological studies could not identify a threshold for exposure levels related to PM health effects. This precludes regular standard setting, with a No Observed Adverse Effect Level (NOAEL) and safety factors. It implies that for any PM standard a certain level of impact on health will have to be accepted.

Because there is no threshold, adverse health effects are less effectively avoided by reducing episodic high concentrations than by reducing annual average concentrations (which will reduce the magnitude of occasional peak concentrations as well).

Overall, health effects are consistently associated with PM_{10} and $PM_{2.5}$. (These terms refer to ambient particles with diameters of up to approximately 10 and 2.5 micrometre). These associations are found in spite of the local differences in air quality, sources and the proportion of the susceptible sub-population. However, there seems to be heterogeneity between locations within the various epidemiological time-series studies for PM-associated health effects. This heterogeneity is manifested in differences in the size of the effects and may probably be influenced by local ambient and population-related circumstances. As for the future, a gradually ageing population and an increasing proportion of asthmatics or people with circulatory problems will proportionally enlarge the potentially susceptible subpopulation.

Choice of PM indicators

Support is emerging for supplementing the current PM standard with other (smaller sized or source-related) indicators than PM_{10}. There is currently a lack of reliable information on ambient levels of these smaller sized or source-related fractions which is representative of the situation in the Netherlands. At the moment the available toxicological and epidemiological evidence is insufficient for regulating ultrafine (UF) particulate concentrations, though this is another field that needs more research as the potential health implications of UF may be considerable. It is recommended that PM_{10} be retained as a standard for the time being as it covers the effects of both fine and coarse particles. In view of the emerging evidence implicating fine particles in health effects, it is recommended that a standard for fine PM or a source-related fraction be developed as well.

Monitoring of PM

The levels of PM_{10} measured by a stationary site monitor seem to be representative of the personal exposure of the general public to ambient PM_{10}. Accurate measurement of PM is complicated. In the Netherlands a substantial fraction of the PM is ammonium nitrate, a salt that is in dynamic equilibrium with the gas phase of ammonium and nitrate. While PM is measured, the sample is heated to minimise interference from water. However, this volatises some of the PM, especially ammonium nitrate. The automatic PM_{10} monitoring network in the Netherlands therefore corrects for losses of semi-volatile material by using a factor of 1.3, as an approximation. The accuracy of PM measurements needs to be increased, as semi-volatile ammonium nitrate is a principal component of PM in the Netherlands. More information on the specific chemical composition and size distribution of PM representative for typical situations in the Netherlands is needed to test relevant hypotheses concerning health effects, source contributions and possible atmospheric influences.

Risk reduction with current PM emission control

Current policies will reduce emissions of PM_{10} by about 20% from 1998 to 2010. The fraction of PM_{10} that is combustion-related and suspected of being health-relevant will show an even larger reduction of 40%, based on projections of energy use, transportation developments and performance of new technology in real-world conditions. Dutch emissions of $PM_{2.5}$ will decrease by about 30%. Traffic is an important source of carbonaceous PM (which can be broken down into Elemental Carbon (EC) and Organic Carbon (OC), the mixture of which comprises diesel soot) and ultrafines, which are emitted at breathing height, close to a large part of the population in the Netherlands.

Whether a reduction in PM levels leads to a proportional reduction in health effects is still uncertain. PM is a complex mixture with fractions that are to a greater or lesser extent health-relevant. Changes in the composition of this mixture might change the health impact. So, the most cost-effective policy will be to reduce that part of PM that causes the health problems. Unfortunately, there are currently only suggestions for the causal fractions as they have not yet been identified.

These health-relevant fractions are probably transport-related (diesel soot) and, more generally, combustion-related primary PM emissions. Certain fractions of ambient PM probably do not cause significant health effects. These include particle-bound water and probably sea salt particles. A number of epidemiological studies suggest that the crustal fraction is less health-relevant than combustion-related fractions. Toxicological studies with pure ammonium sulphate and nitrate (Secondary Inorganic Aerosol (SIA)) have not established overt toxicity of these components, even at concentrations considerably above ambient levels. In contrast, epidemiological studies continue to find strong associations between adverse health effects and secondary aerosol components such as sulphates and nitrates. This divergence of results has not yet been resolved.

Table A. EU standards for PM_{10}.

	Phase1 1 January 2005	Phase2* 1 January 2010
Annual average	40 µg/m^3	20 µg/m^3
Daily average (24-hour)	50 µg/m^3	50 µg/m^3
Number of exceedances per year	35	7

*indicative value

The daily PM standard revisited

In 1999, the European Union promulgated PM standards for 2005 and 2010. The values for 2010 are indicative. This means that the values for 2010 become definitive after the evaluation in 2003. In this evaluation the experience of Member States in meeting the standards for 2005 will be taken into account, as well as the most recent scientific insights. The current EU PM standards can be found in table A:

The EU's original position paper envisaged promulgating two equivalent PM standards. However, the current EU annual average standard (40 µg/m^3) and daily standard (50 µg/m^3, 35 exceedances) for 2005 are not equivalent in the Netherlands.

With **80** permitted exceedances per year, a daily average of 50 µg/m^3 would be equivalent to an annual average of 40 µg/m^3, and a daily standard of 100 µg/m^3 PM_{10} with 7 exceedances. For practical reasons a standard with a value of 100 µg/m^3 and 7 exceedances is preferred to a value of 50 µg/m^3 that may be exceeded on 80 days. In general, the public is able to comprehend a standard with a small number of exceedances better. Although the EU has proposed two standards for PM, there are several arguments that only one standard would suffice – annual mean concentrations being the best choice. However, for reasons of communication to the public, daily standards may be appropriate. Whether or not two averaging times are needed for an EU PM standard is a policy decision.

Dutch compliance with air quality standards with current control policy

Annually averaged values in the Netherlands obtained through modelling are consistent with measurements of PM_{10} here. Compliance with the annual average value of 40 µg/m^3 seems feasible for PM_{10} in the Netherlands, although local exceedances at 'hot spots' cannot be ruled out. However, compliance with the daily average value of 50 µg/m^3 with 35 permitted exceedances is probably not feasible in 2005. Because of the relatively large contribution of foreign PM in a small country like the Netherlands, combined with our substantial natural background levels caused by sea salt, crustal and other natural material, a daily level of 50 µg/m^3 will easily be exceeded. Expectations are that there will still be 36–40 exceedances per year of the EU daily standard of 50 µg/m^3 even after all planned abatement measures (Current Legislation of Emissions (CLE)) have been taken in 2010. Compliance with the indicative annual average value of 20 µg/m^3 for PM_{10} and with the indicative daily average value of 50 µg/m^3 with

7 permitted exceedances in 2010 is not possible in the Netherlands. If abatement measures are implemented in neighbouring countries (as one might expect they will), the number of exceedances will decrease.

The prospect of additional PM abatement

The ultimate potential for reducing primary PM_{10} emissions (on top of currently agreed measures, CLE) could be 60% in the Netherlands. This abatement package is called 'MFR_{ult}': ultimate Maximum Feasible Reduction. The cost of achieving the 'MFR_{ult}' is about 6000 million euro per year. The 'MFR_{ult}' reduction of 60% in primary PM_{10} emissions in the Netherlands will result in a 1.1 $\mu g/m^3$ lower PM_{10} concentration averaged over the country. An emission reduction up to a cost-efficiency of 55 euro/kg PM_{10} will lead to a reduction by a quarter (abatement package: '$2010_{quart\ red}$'). This can be achieved at a cost of 210 million euro per year and will result on average in a 0.3 $\mu g/m^3$ lower PM_{10} concentration. From the absolute value of the PM_{10} levels one can conclude that, averaged on a national level, these reductions seem fairly small. Locally, however, higher reductions in PM_{10} levels of 1 to 5.5 $\mu g/m^3$ are modelled in the '$2010_{quart\ red}$' abatement package. The maximum reductions will be achieved in Rotterdam, which is densely populated. It is interesting to note that the measures directed at *transport* in the '$2010_{quart\ red}$' abatement package focus on the shipping sector only. When concentrating on probably more health-relevant fractions of PM, like traffic-related diesel soot, modelled reductions are relatively higher even. The presented abatement packages ('$2010_{quart\ red}$' and 'MFR_{ult}') correspond to a decrease of 20% and 50% respectively in average traffic-related diesel soot concentration levels of Dutch origin. These effects would increase even further if similar reduction technologies were to be applied to traffic in foreign countries also.

Supplementary PM abatement can be based on the precautionary principle. Further source-oriented actions could focus on the more cost-effective reduction of the total PM_{10} aerosol mass, or could first of all focus on those PM fractions that are expected to be more health-relevant. This last option is preferred. These fractions are probably transport-related (diesel soot) and, more generally, combustion-related primary PM emissions. In this respect, the abatement of uncontrolled shipping emissions has been identified as one of the more cost-effective control options. The abatement of other combustion-related sources such as industrial combustion, wood burning in fireplaces, and off-road machinery is also possible, but is less cost-effective. Additionally, climate change mitigation strategies may reduce combustion-related PM emissions.

Residual risk with improved PM air quality

A substantial part of the PM_{10} levels in the Netherlands cannot be influenced by policy measures, as natural sources are responsible for their ambient concentrations. Because future abatement measures will further reduce the anthropogenic fraction, the contribution of the natural fraction will increase proportionally. More insight into the chemical composition (specific tracers) and contribution of different sources to the currently 'non-modelled' and generally natural part of PM_{10} is necessary to find out how much of the current PM levels may eventually be influenced by abatement measures.

PM air quality will improve in the future. Despite the air quality, it could be conjectured that the health impact associated with PM will nevertheless become more pronounced. In the Netherlands the gradual ageing of the population and other demographic developments could lead to a more than proportionate rise in the susceptible sub-groups. However speculative the previous remark, continuing vigilance seems required for this only partially understood problem of PM.

SAMENVATTENDE CONCLUSIES

Beoordeling van gezondheidsrisico's van fijn stof

Epidemiologische studies uit de hele wereld wijzen op een verband tussen fijn stof (Eng.: *Particulate Matter* of *PM*) en ernstige gezondheidsklachten, die tot ziekenhuisopname en vroegtijdige sterfte kunnen leiden. Dat beeld wordt bevestigd door Nederlands onderzoek. Over ernst en omvang van de acute en chronische gezondheidseffecten woedt nog een wetenschappelijk debat. Er zijn nog veel onzekerheden. Vast staat echter dat de gezondheidseffecten door fijn stof zo ernstig en omvangrijk zijn dat nadere actie geboden is. In epidemiologische studies is geen drempelwaarde aangetoond waaronder géén gezondheidseffecten meer met fijn stof in verband gebracht kunnen worden. Er is dan ook geen klassieke grenswaarde of normstelling met een zogenoemde "No Observed Adverse Effect Level (NOAEL)" voor fijn stof. Welke normstelling men ook kiest, de bijbehorende gezondheidseffecten in de bevolking zullen nooit helemaal uit te sluiten zijn.

Omdat er voor gezondheidsklachten door fijn stof, geen drempelwaarde bestaat, zijn dergelijke gezondheidseffecten doeltreffender te verminderen door de jaargemiddelde concentraties fijn stof te verlagen dan door incidentele piekconcentraties te bestrijden. Bovendien zal het verlagen van de jaargemiddelde concentraties fijn stof ook tot vermindering van de incidentele piekbelastingen leiden.

Wereldwijd kunnen de gezondheidseffecten van fijn stof worden gekoppeld aan PM_{10} en $PM_{2.5.}$ (Dat zijn stofdeeltjes met een diameter tot ongeveer 10 respectievelijk 2,5 micrometer). Dit verband wordt steeds opnieuw gevonden ondanks lokale verschillen in luchtkwaliteit, wisselende bronnen en een wisselend aandeel van gevoelige groepen in de bevolking. Toch komen er in de diverse epidemiologische studies ook verschillen tussen lokaties aan het licht. De omvang van de gezondheidseffecten is vermoedelijk afhankelijk van lokale omgevingsfactoren of bevolkingsomstandigheden. In de toekomst zal een groter deel van de bevolking extra gevoelig zijn voor fijn stof. Dat komt door de toenemende vergrijzing en door het stijgende aantal astmatici en mensen met hart- en vaatstoornissen.

Keuze van indicatoren voor PM

Naast de huidige Europese PM_{10} normstelling voor fijn stof groeit de behoefte aan normstellingen, gericht op fijnere deeltjes of fracties afkomstig uit specifieke bronnen.
Over de Nederlandse situatie ontbreekt echter voldoende betrouwbare informatie. Aanbevolen wordt om voorlopig PM_{10} als normstelling voor grove èn fijnere stofdeeltjes te handhaven. Daarnaast zou er voor fijnere stofdeeltjes een aparte normstelling of een meer brongerichte normstelling ontwikkeld moeten worden omdat er steeds meer aanwijzingen komen dat kleinere stofdeeltjes de gezondheid kunnen schaden.
De huidige toxicologische en epidemiologische informatie is onvoldoende om regelgeving op te stellen voor de ultrafijne (UF) deeltjes, die kleiner zijn dan 0,1 micrometer. Een flink deel van die ultrafijne deeltjes is afkomstig van het verkeer. Op dit gebied is

meer onderzoek nodig, want de gezondheidsschade door ultrafijne deeltjes zou aanzienlijk kunnen zijn.

Meten van PM

De PM_{10} niveaus in de buitenlucht die op een vast meetpunt worden gemeten, blijken in de praktijk ook representatief te zijn voor de persoonlijke blootstelling van het algemene publiek aan PM_{10}. Het nauwkeurig meten van fijn stof is echter lastig. In Nederland bestaat een flink deel van het fijn stof uit ammoniumnitraat, een zout dat in een dynamisch evenwicht verkeert met de gasfase van zowel nitraat als ammonium. Tijdens de metingen van het fijn stof in de stofmonitor worden de luchtmonsters verwarmd om storing door water te minimaliseren. Daarbij vervluchtigt echter een deel van het monster, vooral ammoniumnitraat. Het huidige automatische meetnet in Nederland hanteert daarom een factor van 1,3 om te corrigeren voor de verliezen van het semi-vluchtige deel van het fijn stof. Het is wenselijk dat de precisie van deze PM metingen vergroot wordt. Ook is meer informatie nodig over de specifieke chemische samenstelling en deeltjesgrootteverdeling voor representatieve situaties in Nederland. Daarmee kunnen hypotheses over gezondheidseffecten, bronbijdragen en mogelijke atmosferische invloeden worden getoetst.

Risicoreductie en de bestrijding van emissies

Door het al vastgelegde fijn stof beleid gaan de emissies van PM_{10} tussen 1998 en 2010 in Nederland met zo'n 20 procent omlaag. De fijn stof fractie die verbranding gerelateerd is zal zelfs met 40 procent afnemen, en de $PM_{2.5}$ emissies met 30 procent. Verkeer is een belangrijke bron van ultrafijne deeltjes en koolstofhoudend fijn stof. Dieselroet is een mengsel van elementaire en organische koolstof. Verkeersemissies komen op leefniveau in woonwijken terecht en dicht bij belangrijke bevolkingsconcentraties.

Of een vermindering van fijn stof concentraties ook tot een evenredige vermindering van de gezondheidseffecten zal leiden is nog onzeker. Fijn stof is immers een complex mengsel, waarin sommige fracties meer gezondheidsrelevant zijn dan andere. Veranderingen in de samenstelling van dit mengsel kunnen van invloed zijn op de omvang en aard van de gezondheidseffecten. De meest kosten-effectieve maatregelen richten zich met name op de meest toxische fracties in het fijn stof. Helaas weten we momenteel nog niet met voldoende zekerheid welke fracties dat zijn. Waarschijnlijk behoren tot de voor de gezondheid relevante fracties het dieselroet uit de vervoerssector en fijn stof afkomstig van overige verbrandingsprocessen. Bepaalde fracties fijn stof in de buitenlucht, zoals zeezoutdeeltjes of het water in de stofdeeltjes, veroorzaken waarschijnlijk geen gezondheidseffecten. Een aantal epidemiologische studies doet vermoeden dat bodemstofdeeltjes minder relevant zijn voor de gezondheid dan stofdeeltjes afkomstig van verbrandingsprocessen. Toxicologische studies met zuiver ammoniumsulfaat en -nitraat (secundaire anorganisch aërosol) wijzen niet op een hoge toxische potentie van deze beide componenten, zelfs niet bij aanmerkelijk hogere concentraties dan in de buitenlucht. Wèl wordt in epidemiologische studies keer op keer een samenhang gevonden tussen gezondheidsklachten en aanwezigheid van sulfaat en nitraat als bestanddelen van secundaire anorganisch aërosol. Deze tegenstrijdigheid valt nog niet te verklaren.

Tabel A. EU normen voor fijn stof (PM10)

	Fase 1 1 Januari 2005	Fase2* 1 Januari 2010
Jaargemiddelde	40 µg/m^3	20 µg/m^3
Daggemiddelde (24-uur)	50 µg/m^3	50 µg/m^3
Aantal overschrijdingen per jaar	35	7

*indicatieve waarde

Europese normstelling opnieuw bekeken

In 1999 heeft de Europese Unie de fijn stof normstelling voor 2005 en 2010 vastgesteld. De normstelling voor 2010 betreft een zogenoemde indicatieve waarde. Dat wil zeggen dat deze normen pas definitief worden vastgesteld na een evaluatie in 2003 van de ervaringen die in de diverse lidstaten zijn opgedaan met de normstelling van 2005. Bovendien zal rekening worden gehouden met de nieuwste wetenschappelijke inzichten. Deze EU normen zien er uit zoals in tabel a is weergegeven.

In het document dat oorspronkelijk ten grondslag lag aan de EU normstelling werd aangekondigd dat beide EU fijn stof normen (daggemiddeld en jaargemiddeld) gelijkwaardig ofwel equivalent zouden zijn. In de praktijk blijkt echter dat in Nederland de huidige EU jaargemiddelde PM_{10} norm voor 2005 van 40 µg/m^3 niet equivalent is aan de daggemiddelde norm van 50 µg/m^3 met 35 toegestane overschrijdingen per jaar. Pas met **80** toegestane overschrijdingen per jaar zou een daggemiddelde norm van 50 µg/m^3 equivalent zijn aan de jaargemiddelde norm van 40 µg/m^3. Datzelfde geldt voor een daggemiddelde norm van 100 µg/m^3 met 7 overschrijdingen.
Om praktische redenen wordt de voorkeur gegeven aan een daggemiddelde norm van 100 µg/m^3 met 7 overschrijdingen boven een daggemiddelde norm van 50 µg/m^3 met 80 overschrijdingen. In het algemeen zal ook het publiek een hogere norm met een geringer aantal overschrijdingen beter kunnen begrijpen.
Hoewel de EU twee normen heeft voorgesteld, is er een aantal argumenten waarom voor fijn stof één norm toch voldoende is. De jaargemiddelde norm is dan de beste keuze. Om redenen van risicocommunicatie kan een daggemiddelde norm toch nuttig zijn. Aan beleidsmakers de keuze of er twee EU normen nodig zijn voor fijn stof.

Voldoet Nederland aan de luchtkwaliteitsnormen bij het huidige beleid?

De jaargemiddelde fijn stof concentraties die we in Nederland modelleren zijn consistent met onze metingen. We verwachten dat het voldoen aan een jaargemiddelde PM_{10} norm van 40 µg/m^3 in Nederland in 2005 wel haalbaar zal zijn, hoewel plaatselijke overschrijdingen op een aantal met name verkeersbelaste ‘hot spots’ niet uit te sluiten zijn. Daarentegen is het niet waarschijnlijk dat we in 2005 overal de daggemiddelde norm van 50 µg/m^3 met 35 toegestane overschrijdingen halen. Vooral in zo’n klein land als Nederland met een relatief grote buitenlandse bijdrage en een relatief hoge achtergrondsbelasting door zeezout wordt een daggemiddeld niveau van 50 µg/m^3 gemakkelijk overschreden.

De verwachting is dat zelfs als alle overeengekomen bestrijdingsmaatregelen in 2010 zullen zijn uitgevoerd volgens het '*current legislation scenario*' (CLE) er in Nederland toch nog 36 tot 40 overschrijdingen van de EU daggemiddelde norm van 50 µg/m^3 zullen zijn. De indicatieve EU normen voor 2010 van 20 µg/m^3 als jaargemiddelde en een daggemiddelde norm van 50 µg/m^3 met 7 overschrijdingen zijn dan ook voor Nederland geen van beide haalbaar.

Perspectief voor een verdere vermindering van fijn stof

Uiteindelijk is het technisch mogelijk om de Nederlandse PM emissies met nog eens 60 procent extra te verminderen (bovenop de al voorgenomen maatregelen volgens het *current legislation scenario*). Dit maatregelenpakket wordt "MFR_{ult}" genoemd: de ultieme Maximaal bereikbare Reductie. Dit maatregelenpakket kost jaarlijks ongeveer 6 miljard Euro. Gemiddeld over Nederland zal een extra reductie van de fijn stof emissies met 60 procent leiden tot een 1,1 µg/m^3 lagere jaargemiddelde concentratie aan PM_{10}.

Daarnaast is nog een ander maatregelenpakket doorgerekend, dat aanmerkelijk goedkoper uitpakt. Hierbij wordt de emissiereductie beperkt tot maatregelen met een marginale kosten-efficiëntie van 55 Euro per bespaarde kg $PM_{10.}$ Dit maatregelenpakket, waarbij de Nederlandse emissies met ongeveer 25 procent ofwel een kwart afnemen, wordt "$2010_{quart\ red}$" genoemd. Het pakket kost jaarlijks 210 miljoen Euro. Gemiddeld over Nederland zal een reductie van de fijn stof emissies met 25 procent leiden tot een 0.3 µg/m^3 lager jaargemiddelde concentratie aan PM_{10}.

Dat lijkt maar een kleine vermindering van de huidige concentraties fijn stof in Nederland. Plaatselijk worden echter forsere reducties verwacht. Zo leidt het pakket "$2010_{quart\ red}$" plaatselijk tot verminderingen van 1 tot 5,5 µg/m^3. De maximale reducties kan men bij dit pakket verwachten in het dichtbevolkte Rotterdam. Interessant is ook dat de op het verkeer gerichte maatregelen in het pakket "$2010_{quart\ red}$" alleen betrekking hebben op de scheepvaart. Als we ons concentreren op de waarschijnlijk meer verkeersgerelateerde fracties van fijn stof, zoals dieselroet, dan zijn de gemodelleerde verminderingen zelfs relatief belangrijker. De al genoemde pakketten "$2010_{quart\ red}$" en "MFR_{ult}" leiden tot een afname van het Nederlandse dieselroet van respectievelijk 20 procent en 50 procent. Als ook in het buitenland vergelijkbare maatregelen worden getroffen, wordt de aanpak nog effectiever.

Aanvullende fijn stof bestrijding kan gebaseerd worden op het 'voorzorgbeginsel'. Bij verdere brongerichte acties kan men zich richten op het zo kosten-efficiënt mogelijk terugdringen van ofwel de totale PM_{10} massa ofwel op de waarschijnlijk meer gezondheidsrelevante fractie daarvan. Aan de laatste optie wordt de voorkeur gegeven. Deze fracties zijn waarschijnlijk verkeersgerelateerd dieselroet of meer in het algemeen verbrandinggerelateerde PM emissies. Daarom is het bestrijden van de nu nog vrijwel onbestreden scheepvaart emissies een bij uitstek kosten-effectieve optie. Het bestrijden van andere verbrandinggerelateerde bronnen zoals industriële verbranding, stoken van openhaarden en mobiele werktuigen is ook mogelijk, maar minder kosten-effectief. Ook aanvullende maatregelen in het kader van het klimaatbeleid kunnen de verbrandinggerelateerde fijn stof emissies helpen terugdringen.

Welke risico's blijven nog over bij een verbeterde luchtkwaliteit?

Een aanzienlijk deel van de PM_{10} niveaus in Nederland is niet door beleidsmaatregelen te beïnvloeden, aangezien het afkomstig is van natuurlijke bronnen. Naarmate de door mensen veroorzaakte emissies verder worden teruggedrongen, stijgt het aandeel van de natuurlijke bronnen. Er is meer inzicht nodig in de chemische samenstelling en in de bijdragen van de diverse bronnen aan de merendeels natuurlijke en tot nog toe meestal niet gemodelleerde fijn stof fracties in de lucht. Daaruit valt af te leiden in hoeverre het fijn stof probleem uiteindelijk door milieumaatregelen kan worden aangepakt.

Ook al wordt het fijn stof probleem aangepakt, de bijbehorende gezondheidsklachten zullen niet van de agenda verdwijnen, integendeel. In Nederland worden mensen steeds ouder en wellicht leiden ook andere demografische ontwikkelingen tot een meer dan evenredige toename van extra gevoelige bevolkingsgroepen. Hoe dat uitpakt is de vraag, maar bij een nog zo slecht begrepen fenomeen als fijn stof blijft voortdurende waakzaamheid geboden.

1 INTRODUCTION

In 1998, a proposal was made for a daughter directive for *inter alia* PM_{10} based on the European Union (EU) Framework Directive on Ambient Air Quality. In 1999 the following standards were promulgated, c.f. Table 1.

This PM_{10} standard is to be evaluated in 2003 and brought into line with new scientific developments in knowledge about the effects of PM_{10} on health and the environment. In addition, the practical experience of member states in applying the standards, as well as the feasibility of meeting the standards, are to be considered.

The EU PM_{10} directive was based on a position paper from 1997, which discussed four different aspects of the field of PM risk assessment. The first of these is the pollutant description (PM_{10}). New research suggests that there are currently other descriptors like finer PM, ultra fine (UF) or source-related PM that also need to be considered for purposes of standard setting. The second aspect is that of the averaging time. Risk estimates based on recent measurements in the Netherlands suggest that either standard would lead to similar risk estimates. Monitoring of PM is the third aspect. In the position paper and the accompanying documents the need for the use of a correction factor was indicated because the semi-volatile fraction of PM is only partially measured in the currently used automatic measuring devices. Compliance assessment is difficult in these circumstances. The fourth aspect is that of cost. For this last, but certainly not least, aspect, more information has again become available since 1997. The EU has decided that a new position paper is warranted in 2003 and is working on it.

The full report accompanying this executive summary has been prepared for a number of Dutch Ministries in the context of the Netherlands Aerosol Programme to facilitate a Dutch position in the evaluation process of the EU PM directive in 2003. The Netherlands Aerosol Programme was instigated at the request of three Ministries, that of Housing, Spatial Planning and the Environment, that of Transport, Public Works and Water Management and that of Economic Affairs. It is being conducted jointly by the Netherlands Institute of Public Health and the Environment (RIVM, Bilthoven), the National Organisation for Applied Scientific Research (TNO, Apeldoorn), the Energy Research

Table 1. EU standards for PM10.

	Phase1 1 January 2005	Phase2* 1 January 2010
Annual average	40 µg/m^3	20 µg/m^3
Daily average (24-hour)	50 µg/m^3	50 µg/m^3
Number of exceedances per year	35	7

*indicative value

Foundation (ECN, Petten) and the Institute for Risk Assessment Studies (IRAS, Utrecht).

In July 2001, the Netherlands Aerosol Programme distributed a discussion document on health risks of particulate matter in ambient air. This document, also known as the 'orange document', was discussed *inter alia* at a speciality workshop held on 6 September 2001, following the annual ISEE conference, which took place that year in Garmisch Partenkirchen, Germany. Some 30 experts from Europe and the US attended this speciality workshop and discussed the ideas presented in the discussion document. A second formal opportunity for international discussion arose during a two-day workshop in June 2002 with some 20 experts from the Netherlands and US-EPA in Research Triangle Park (North Carolina) in the United States. Judging by the discussions that followed, including those outside the regular workshops, the 'orange document' fulfilled its purpose well. The feedback we received helped us improve the original document.

However, on a number of issues the scientific evidence available regarding the role of PM is as yet indecisive. Weighing the current evidence, experts sometimes arrive at different conclusions. This report, therefore, by no means contains definitive answers. The whole process we went through has resulted in the common ground presented here. This position will constitute a major element in the Dutch contribution to evaluation of the EU daughter directive on ambient PM in 2003.

This executive summary consists of four parts:

- For those not familiar with the basic concepts of PM and its health effects Section 2 presents a brief explanation in order to facilitate understanding of the rest of this document and the full report.

- Section 3 presents a summary of the most recent information on the health effects of PM, in which the available epidemiological, toxicological and human clinical information will be treated jointly.

- In Section 4 the information currently available on PM air quality in the Netherlands is presented. This section also devotes attention to sources and emissions of PM and precursors of secondary inorganic aerosols (SIA).

  The different standards for PM and other policy options are presented in Section 5. This section also contains information on the ambient PM levels that may be expected in 2005 and 2010. Points of scientific debate will be presented to indicate which different options these views produce for later abatement measures.

Further details can be found in the accompanying full report, which is available on CD-ROM (RIVM report # 650010 032) and can be requested by e-mail from (E.Buringh@rivm.nl) or can be downloaded from the Internet (www.rivm.nl.) by going to publications and the pdf-file of report number 650010 032.

2 SOME BASIC CONCEPTS

In this section of the executive summary highly abridged information is provided on some basic concepts of PM. A more thorough treatment can be found in the full report, which also gives the appropriate literature references. Those readers who are already familiar with ambient PM and its associated health effects could skip this section and continue reading from Section 3, where the results of the Netherlands Aerosol Programme are presented.

2.1 Basic concepts concerning Air Quality

2.1.1 Particulate Matter

Particulate Matter (PM) or particles with an **aerodynamic diameter** of 10 µm or less are called inhalable. This is the fraction that is naturally inhaled by humans, while larger particles are less likely to enter the human respiratory tract. To mimic human respiration and correctly estimate the inhaled dose, ambient PM is sampled through a size-selective inlet with a 50% efficiency cut-off at 10 µm aerodynamic diameter. This PM fraction is called PM_{10}. In the alveolar region (deep in the lungs) peak deposition for particles above 0.1 µm occurs near a diameter of 4 µm. Particles that are smaller still are categorised as $PM_{2.5}$ (50% cut-off 2.5 µm), PM_1 or even $PM_{0.1}$.

From a human health point of view, PM generally larger than 10 µm in diameter and forming a large part of TSP (**Total Suspended Particulates**) is probably of less concern, because we do not inhale much of it. However, studies of traffic-related air pollution in conjunction with cedar pollen (> 10 µm) in Japan suggest that increased rhinitis can be a result of exposure to larger particles. From the point of view of nuisance dust, large particles or TSP can be a problem. Information on the PSD **(Particle Size Distribution)** is essential to understand the potential health effects of PM. The deposition of PM in the respiratory tract and lungs (inhaled dose) varies with particle size. Apart from its size distribution, the chemical speciation and bioavailabilty of PM is also information that is needed to understand the health effects of PM. For an understanding of current health-related PM problems, three size ranges are in general distinguished.

The smallest particles are the **ultra fine particles** or ultra fines (UF), which are less than 0.1 µm in diameter. Although this fraction makes hardly any contribution to the total mass of PM_{10}, it dominates the number of particles in the air. Ultra fine particles coagulate quite rapidly with the fine particles in the atmosphere and largely end up in the accumulation mode (0.1 µm < PM <1.0 µm) of these sub-micron particles. The next size class is the **fine fraction,** smaller than 2.5 µm (but bigger than 0.1 µm), and the largest is the **coarse fraction** of PM_{10} in the range between 2.5 and 10 µm. The (reactive) surface area of the fine and ultra fine fraction is also much larger than that of the coarse mode fraction. Recently, some researchers have also used a size cut-off of 1 µm (PM_1). This is a cut-off

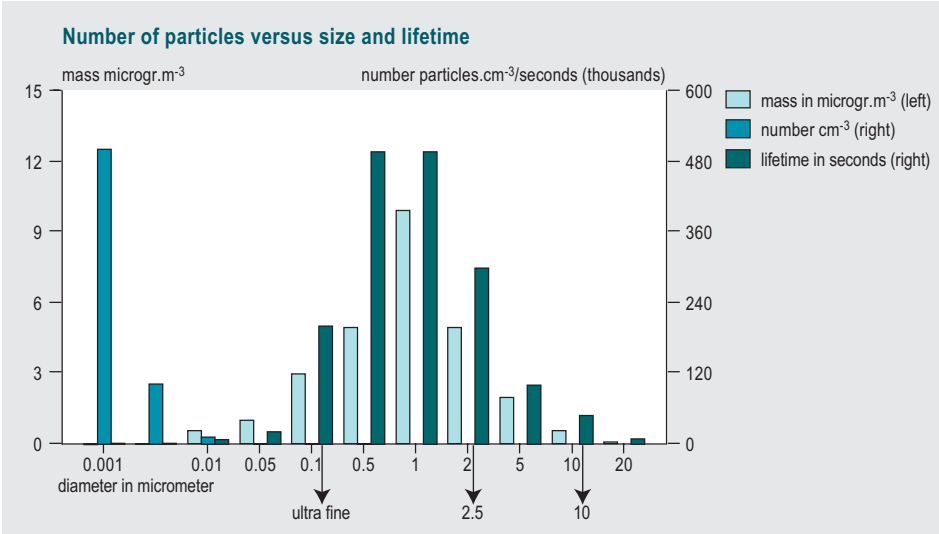

Figure 1. Particle mass (in µg/m^3), numbers (in N /cm^3) and life time (in seconds).

that best discriminates between the crustal, which is generally larger than 1 µm, and the non-crustal fraction, which for a large part is smaller than 1 µm in diameter. However, this does not imply that anthropogenic PM is not part of the fraction above 1 µm.

Figure 1 presents the particle mass and numbers versus diameters and residence time or lifetime of the particles. The peak in numbers of particles is with the ultra fines, which have a lifetime of less than 20,000 seconds or 6 hours. The peak in mass is more with particles around 1 µm, which also have longer lifetimes of 6 days.

Sometimes other particle measures are also reported, and as they will be presented in this report, too, they need to be mentioned here. In the past, BS was used to indicate the **Black Smoke** concentration, based on the reflectance of filters. These BS particles are mostly smaller than a few µm and do not necessarily relate to the total particle mass. The original calibration curve for BS (OECD method) is based on the measured concentrations of PM in the United Kingdom (UK) in the 1950s. The ambient mix of PM has changed, so the original calibration curve has lost its meaning. The current reflectance measurement of BS filters has been found to correlate highly with elemental carbon and is therefore a useful surrogate for primary traffic-related emissions.

2.1.2 Sources and composition of PM

PM has both a **primary** component, which is emitted directly by sources such as traffic and industry, or indirectly as wind-blown soil particles and sea spray, and a **secondary** component which is formed in the atmosphere by chemical reactions of gases, most notably sulphur dioxide, oxides of nitrogen, ammonia and volatile organic compounds.

PM can be emitted, and exist in the atmosphere, in a wide range of particle sizes. Both primary and secondary PM may be the result of **natural** or man-made (**anthropogenic**) sources. Heterogeneous chemical reactions of gases with ambient PM in the atmosphere can lead to new and sometimes highly reactive components. A high ambient relative humidity can also cause PM to exist in a droplet form beside a more solid particulate form. In the Netherlands these high relative humidity conditions are often met.

Nearly all UF particles are formed during high temperature **combustion processes** by either mobile sources or fossil fuel-based power production. However, in specific circumstances UF may be formed by natural processes, e.g. at coastal sites. Fine particles mainly result from atmospheric reactions of PM or atmospheric reactions involving the gaseous precursors (NO_x, SO_2, NH_3) leading to secondary PM. Primary fine PM emitted by diesel engines is of a carbonaceous nature and consists of elemental and organic carbon. It is also known as DEP (Diesel Exhaust Particulate) or “diesel soot”. Coarse particles relate more to primary emissions by mechanical processes or the handling of dusty materials. The re-suspension of crustal material by turbulence caused by traffic and wind-blown soil particles also result in rather coarse particles. However, there is no strict relation between **size distribution** and **sources** of emissions, as in the Netherlands, for example, natural emissions of sea salt in PM_{10} contain approximately equal amounts of fine and coarse particles, when coarse is defined with a 50% cut-off at 2.5 µm. However, when a cut-off of 1 µm is used sea salt, with a mass median diameter in the 1-2.5 µm range, is classified largely as coarse.

Apart from its size range and emission sources PM can also be characterised by its **chemical composition**. The chemical composition of PM in ambient air depends on the contribution made by both anthropogenic and natural sources. The former includes primary emissions from industry, power production and traffic, and secondary emissions of gaseous precursors. The natural sources mainly consist of primary emissions of sea salt, wind-driven soil dust and secondary organic particulate matter. The chemical composition of PM varies in accordance with these different emission sources. One can distinguish the carbonaceous part of PM, consisting of elemental carbon (EC) and organic particulate matter (OC), secondary PM (the ammonium salts (NH_4^+) of nitrates (NO_3^-) and sulphates (SO_4^{--})), also called **SIA (Secondary Inorganic Aerosols),** and natural PM (sea salt, crustal material and secondary organic particulate matter from natural emissions of precursors). The generic term SOA (Secondary Organic Aerosol) can comprise aerosols formed from natural as well as man-made emissions.
Elemental carbon and ammonium salts are chemically well defined, while organic PM, sea salt and crustal material consist of mixtures of chlorides, oxides of metals and silicates and a wide range of organic compounds. Of specific toxicological interest are the so-called transition metals. Besides their chemical composition as such, the bioavailability of the various components is probably also very important for their possible health effects. However, as a consequence of PM sampling methodologies the exact composition of a single particle is largely unknown. For example, the surface of a sulphate particle may very well be covered with transition metal, or crustal particles may be covered with very small carbonaceous material.

Heterogeneous atmospheric reactions of ambient PM with gases can in the right circumstances result in highly **reactive products** like radicals and peroxides. Due to their high reactivity these components are hard to measure, particularly using conventional particle sampling techniques. Other lesser known factors are particle charge and radiation. These considerations need to be taken into account when crude particle measures like mass concentrations of PM_{10} or $PM_{2.5}$ are compared with health effects. It is important not to overlook these reactive products as it might be quite possible that some unmeasured component (from a currently unknown source) correlating highly with PM or SIA is causing the statistically observed associations between PM and health effects.

Part of the PM in the Netherlands consists of **semi-volatile material**. Conditions during the sampling and measurement of PM will influence the quantitative and qualitative outcome. The dynamic atmospheric behaviour of particles is important for both size and mass of ambient PM. The dynamic equilibrium of semi-volatile compounds with the gas phase influences transformation processes and can change particle mass and composition considerably. The ammonium nitrate concentrations in PM_{10} in the Dutch situation are influenced to a large extent by the local ambient ammonia concentrations because of the dynamic equilibrium with the gas phase. Due to heating during automatic PM measurement the semi-volatile material, or some of it, is lost. Therefore, PM_{10} measurements in the Netherlands are corrected by a factor of 1.3 to compensate for the losses. This factor of 1.3 has been established experimentally and has been quantified correctly for large geographical scales and long-term averages. Locally it may, however, differ considerably on a day-to-day basis due to the varying composition of the semi-volatile fraction of the aerosol.

2.1.3 Transport of PM

The atmospheric **residence times** and hence ranges of travel of these different size fractions vary considerably. They range from more than 60 hours for sub-micron particles larger than 0.1 µm in diameter to less than two hours for the size class above 20 µm. UF particles, with diameters of less than 0.1 µm, generally have much shorter residence times (a couple of hours or less) than the sub-micron particles. Residence times are also equivalent to a mean **transport distance**. A residence time of 60 hours ($0.1 < PM < 1$ µm) is equivalent to a transport distance of more than a thousand kilometres for average western European conditions. For a two-hour residence time ($PM > 20$ µm) the mean transport distance is only 35 km. This leads to the conclusion that for different PSD and their sources the scale of the PM problems may vary from local to more or less continental. PM concentrations in a small country like the Netherlands will be influenced by a combination of local and, for the larger part, foreign emissions. For particles smaller than 0.1 µm, the ultra fines, other time scales and transport distances are relevant. UF react quickly with other (larger) particles and concentrations are generally elevated up to a couple of hundred metres from major roads. More information can be found in the full report.

2.1.4 Monitoring of PM

Two tools are generally used (in combination) to characterise ambient air pollution. The first is monitoring and the second is modelling. Accurate routine **monitoring** of PM is complicated. Due to the dynamic equilibrium of moisture in the atmosphere and the aerosol phase and the semi-volatile part of PM, which is also in a different dynamic equilibrium with its gas phase, accurate measurement of PM is, to use an understatement, quite difficult. Generally, the problems with semi-volatiles play a role in filter methods as well as automatic monitoring devices. In the Netherlands an instrument (**FAG**) is currently being used which is based on the principle of beta-attenuation and actually uses a correction factor of 1.3 to compensate for losses. Other countries (like the UK) use a tapered element oscillating microbalance (**TEOM**), which probably needs a larger correction factor because of the more extensive evaporative loss associated with the higher temperature in the instrument.
Apart from the aerosol mass mentioned above, other necessary measurements for the characterisation of PM are its size distribution (PSD) and chemical composition (preferably also in the various size classes). For the desired size cut, a size selective head (e.g. 10 µm, 4 µm, 2.5 µm or 1 µm) can be employed before the measurement device. Averaging time is also an important aspect of PM, as part of the PM may be in the form of short-lived radicals or other reactive constituents.

Ambient air pollution is generally measured at a **stationary background site**. In the Netherlands there are regional and urban sites, supplemented by street sites that are more traffic-oriented. In the Netherlands a single stationary site appears to be quite representative of a much larger area and not just of its immediate surroundings. This can be concluded from the high correlation of daily PM_{10} data for the monitoring stations in the network that covers the country. However, people tend to live indoors in the Netherlands instead of outdoors. The PM concentrations indoors appear generally to be a fair reflection of the ambient PM levels outdoors. Research using **personal** monitors, i.e. instruments carried by individuals, has indicated that a central site monitor adequately represents personal exposure to ambient PM. This is why it is possible to establish a relationship between ambient PM levels and health effects in a population that spends the greater part of its time indoors. It also means that the population is exposed to PM that is generated outdoors, so it is a public concern. In addition to PM of outdoor origin, human beings are exposed to PM of indoor origin such as tobacco smoke, PM associated with certain occupations and mechanically generated, coarse PM that is encountered everywhere where people move around. Such exposures are usually not correlated with exposures to PM of outdoor origin, so that they do not confound the association between outdoor PM and health effects.

The frequency of occurrence of any ambient air pollutant can be described by a log-normal distribution curve. This means that the distribution of the concentrations is described by two parameters: the **geometric mean** (GM) and the **geometric standard deviation** (GSD). There is a certain mathematical relationship between the **arithmetic mean** (AM) or normal (yearly) average and the GM and GSD. This is described in the

full report. The yearly average (AM) from the EU standard can be transformed into a GM and GSD and so a **90-percentile** or a **98-percentile** can be calculated for the expected distribution of concentrations. A 98-percentile is the concentration that is exceeded on 2% of the days or 7 days per year. When the number of exceedances is 35 it virtually coincides with a 90-percentile of the distribution. Both of these percentiles or numbers of exceedances can be found in the EU daily standards.

2.1.5 Modelling of PM

For **PM modelling** models with long-term (years) or short-term (hours to daily) meteorology are available. Long-term models generally use the actual meteorology of a specific year or the 10-year average meteorology, whilst short-term models use the actual daily meteorological data. In the Netherlands, RIVM uses the OPS dispersion model to calculate yearly averages. Both types of model calculate the PM concentrations based on input parameters like direct PM **emissions** and the emission of precursor gases of secondary aerosols. Yearly average emissions can now be presented for the Netherlands and for Europe with quite some reliability, but accurate and actual daily emissions are still problematic. As a rule of thumb, a diffusive emission of approximately 20 ktonnes of PM_{10} in the Netherlands results in a yearly average regional concentration of around 1 µg/m^3. Urban concentrations or those close to point sources can be considerably higher. Of course, it is always essential to compare the calculations of the models with actual measurements.

In Figure 2, two pie charts show the distribution of Dutch PM emissions of PM_{10} and $PM_{2.5}$ over different **source sectors**. This figure shows, for instance, that the relative contribution for $PM_{2.5}$ made by the transport sector is greater than its relative contribution to PM_{10}.

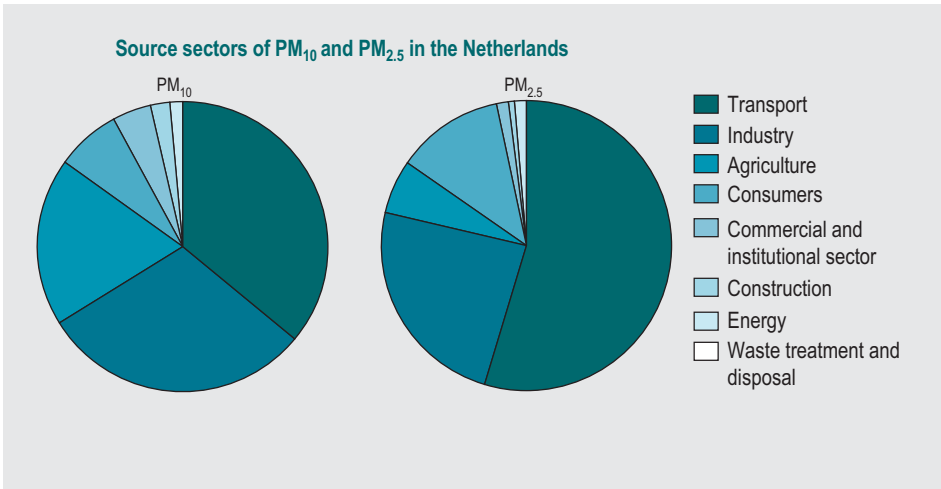

Figure 2. PM_{10} emissions and $PM_{2.5}$ emissions in 1998 by source sector in the Netherlands.

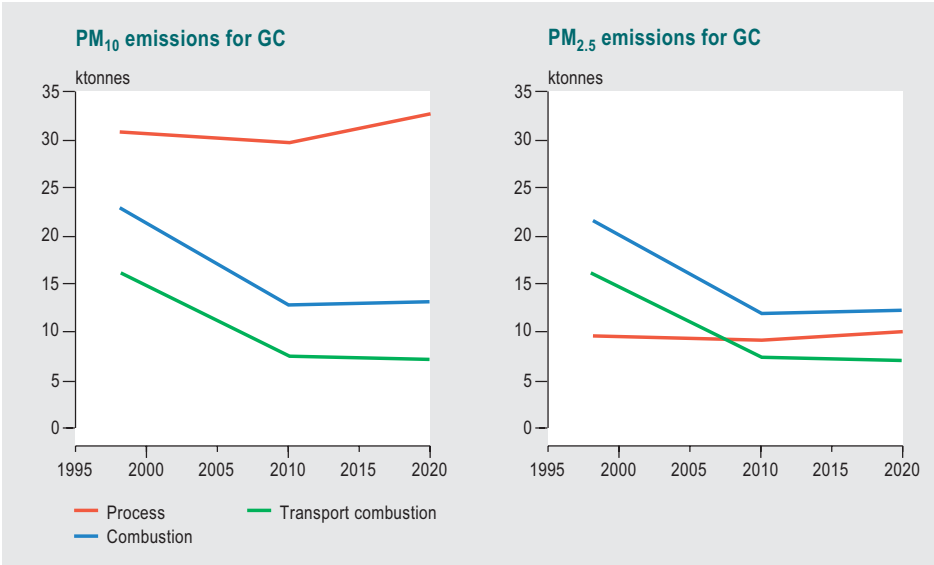

Figure 3. Dutch combustion and process emissions of PM_{10} and $PM_{2.5}$ in ktonnes for the GC scenario

For the modelling of future concentrations, different economic **scenarios** have been developed to estimate the emissions of primary PM and of precursor gases for SIA. These scenarios answer to names like **Global Competition** (GC) or **European Co-ordination** (EC) and are described in more detail in the full report, where the appropriate literature references can also be found. Within the scenarios the influence of different packages of abatement measures can be calculated to present a number of options, including their price tags and the expected impact on PM concentrations. The current legislation emission scenario is presented as a base case under the name **2010-CLE**.

Figure 3 presents the development of future primary **PM emissions** in the Netherlands in a number of years. It shows, for instance, that the process-related emissions of PM are expected to remain more or less similar in size between 1995 and 2020, while the possibly more health relevant transport- and combustion-related emissions will decrease considerably in the Netherlands in the near future.

2.2 Basic concepts concerning Health Effects

2.2.1 Internal dose of PM

In general, epidemiological studies associate ambient concentrations with health effects, assuming a relationship between ambient concentrations (as a surrogate for exposure) and the delivered internal dose. However, the internal dose of PM is highly dependent on the particle diameters, personal behaviour and individual variation in respiratory

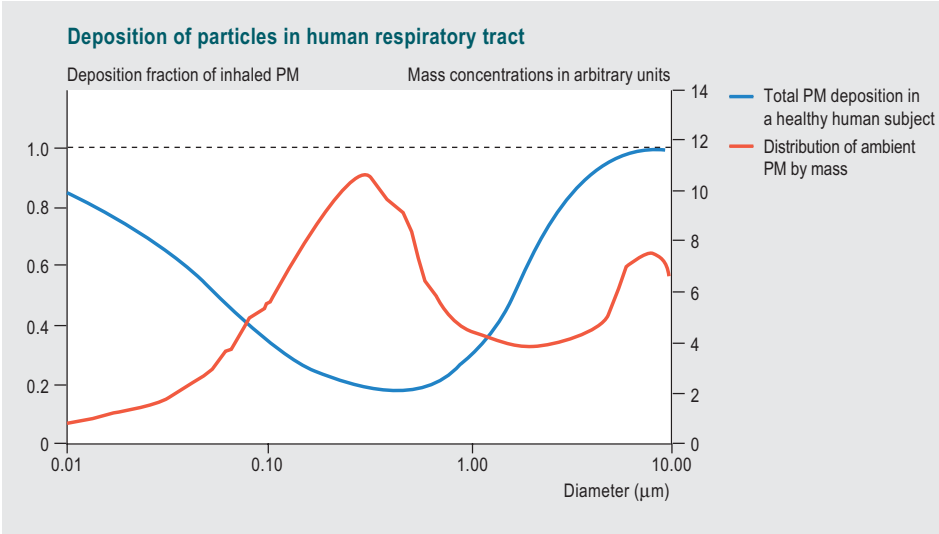

Figure 4. Deposition of particles in the human respiratory tract as well as an average ambient particle mass and number distribution as function of the diameter

tract morphology. The biological effects of ambient PM_{10} particles may be dependent on the dose at critical target sites and organs. Estimating or measuring this (deposited) dose of particles is called **dosimetry** and it forms the link between the external exposure concentration (mostly expressed as mass) and the effective dose at the target site. Health effects of PM_{10} and its constituting fractions in the airways and lungs may also depend on the specific dose metric, usually expressed as particle mass, surface or number per unit (total lung, surface area, lung branch or lobe, c.f. Figure 4).

Large individual variations exist in regional deposition due to the differences in lung function, age and morphology. In general, PM deposition is significantly larger for children (in the age range 0-15 years) than for adults. Exercise increases total PM deposition, and a marked shift occurs in the location it is deposited (head, trachea, lungs). Also, both coarse and ultra fine particles can be very efficiently removed from the air stream in the nasal area. This can have profound implications if the recently developed hypothesis that particles can be transported through the olfactory nerves towards the brain is correct. Pulmonary diseases can also result in a significant increase in local PM doses, in particular in the conducting airways. Altogether this implies that certain conditions may be present in which one person can receive a substantially higher (local) dose than another in the same air quality conditions. Although this a plausible explanation for the PM-associated health effects, population dose estimates should provide an answer to the question of whether using doses rather than PM concentrations for a central site increases the RR for health outcomes.

A special field of exposure characterisation is that of the dosimetry of ambient PM in the human body. The **deposited dose** along the human **respiratory tract**, starting in the

nose and ending deep in the lungs, differs for the various particle sizes. The larger particles and the ultra fines are almost completely deposited when they are inhaled. The fraction least deposited is that between approximately 0.1 and 1 µm in diameter, but it is this mode specifically that accumulates in the ambient air. This means that an average person breathing 10 m^3 per day and exposed to a given PM level with an average size distribution will receive a deposited dose that is considerably less than the inhaled amount of PM. Approximately half of the inhaled PM will be deposited in the body and the other half will be exhaled again; for further details see the full report. However, as shown by these models as well, people with lung diseases may receive a considerably higher internal or even local PM dose compared with a healthy subject. The effects of age (infants versus adults) and changes in breathing pattern (rest versus heavy exercise) can also be studied using these models. Another advantage of dosimetry models is the option to extrapolate a human exposure to an experimental animal and vice versa. Ultimately, the actual received PM dose for a given population can be estimated based on air quality data and activity pattern databases.

2.2.2 Epidemiological research on health effects of PM

Over the last decade a large number of epidemiological studies have been published on the association between ambient PM exposure and possible health effects. Reported **health outcomes** are pulmonary function decrements, respiratory symptoms, hospital and emergency department admissions and mortality. More recently studies were published that focused on birth defects and general practitioner visits in relation to PM exposure. In general, a distinction can be drawn between two types of health outcome: ***mortality*** and ***morbidity*** *(including hospital admissions, lung function decrements and symptoms)*.

The obvious **route of exposure** of air pollution is through the respiratory tract, and it is biologically more plausible that air-pollution-related health effects will cause deaths from, for instance, **respirable causes, pneumonia, chronic obstructive pulmonary disease (COPD) or cardiovascular diseases** than from unrelated causes of death such as digestive diseases. This type of pattern is also what we see in the different epidemiological time-series studies. As the number of deaths for respiratory diseases and for cardiovascular diseases is lower than that for all-cause mortality, the ensuing RR per specific increase in PM should be higher for those specific causes of death than that for all-cause mortality. Also, this is a general pattern we find in epidemiological studies. When the association between PM_{10} and a presumed unrelated cause of hospital admissions (digestive diseases) was studied in the Netherlands, this unrelated cause had no association with air pollution, whilst the respiratory and cardiovascular causes did have an association with ambient air pollution. Such qualitative information gives confidence in the resulting picture of PM-associated health effects. More recently, other routes of internal exposure have been postulated, like deposition of particles in the nasal area and translocation through the olfactory nerves into the brain. This also increases the likelihood for systemic effects, including heart failure.

Most of the recently published studies assessed the relationship between excess *daily mortality* (acute mortality) and exposure to PM (expressed as PM_{10}, $PM_{2.5}$, $PM_{10-2.5}$, TSP, Black Smoke, CoH) in human populations. In these studies a **time-series analysis** is performed on the relationship between daily mortality counts in a population (city, county, country) and daily variations in air pollution levels. Mostly, a specific time period is taken into account between the correlation of air pollution data and health effects. This time is called the **lag** time. Lag 0 means that air pollution and health effects on the same day are studied. Lag 1 means a study of air pollution on one day with health effects on the next day, etc. Although the first publications in the early 90s showed differences in the techniques for statistical analysis, most of the recently published studies show general correspondence in the analytical approach. The analysis most applied is the Generalised Additive Model (GAM) Poisson regression with adjustments for seasonal cycles, long-term trend, temperature, day of the week. In a few more recently published studies an adjustment was also made for dew point and barometric pressure in addition to the aforementioned co-variates. Outcomes of the models are Relative Risk (RR) estimates, expressing the excess daily mortality (total mortality or cause-specific mortality) per magnitude increase in PM (per 50 $\mu g/m^3$, or per 100 $\mu g/m^3$, or per interquartile range, etc.). In the published literature, with only few exceptions, generally consistent mortality-relative risks have been reported, although heterogeneity (differences in the amount of effects) is reported between different study locations within studies. This could be interpreted as pointing to local factors like the PM composition, sources or, possibly, the proportion of the susceptible sub-populations, which may influence the extent of the health effects.

There are far fewer published studies on the mortality effects of *long-term* exposure to ambient PM than on the mortality effects of acute exposure. The current number of studies published, so-called **cohort studies**, is five. The total number of publications in scientific journals about these five cohort studies is, of course, much larger, but essentially all conclusions are based on the information provided by just five cohort studies. In a prospective cohort study, individual persons are followed over time and the occurrence of disease or death is related to the air pollution exposure during their lifetime, or parts of it. The advantage of cohort studies over time-series studies is that information is gathered on the individual level (smoking, occupational exposure, socio-economic status, food intake etc.), while time-series studies make use of existing databases on mortality statistics with no additional information on the individuals who died. On the other hand, time-series studies are relatively easy to perform because of the existence of the databases on mortality and statistics, while prospective cohort studies are time-consuming and labour-intensive, which makes them relatively expensive.
Three of the five cohort studies were population-based, while the other two looked into the relationship between long-term exposure to air pollution and mortality in particular populations. The US studies in the general population especially have been very thoroughly re-analysed and have generally been corroborated. Overall, the results showed statistically significant relationships between long-term exposures to PM and/or sulphates and excess mortality.

But cohort studies are essential when we want to make a statement concerning, for instance, the extent of **life shortening** involved in these associations with air pollution. For the moment we can say there is an association between the daily variation in PM and mortality, but it is impossible to state on the basis of the time-series studies how much earlier this mortality occurs. The advancing of death might as a minimum have been the lag period in question or it might have been much longer if that person had not passed away on that specific day; he or she may have lived for another year or more. In the amount of life shortening and therefore in actual public health consequences, this makes quite a difference. Unfortunately, we cannot retrieve such information adequately from time-series studies, although recent work on 'harvesting' has shown that the amount of life shortening in the time-series studies is more than a few months. In order to estimate the full extent of life shortening we need cohort studies. For the question of the public health impact of PM-associated health effects some information on the amount of life shortening would be most welcome indeed. Published estimates suggest that the amount of life shortening is in the order of 1-2 years for realistic exposure contrasts.

Another factor for which cohort studies with their individual data are more suitable than time-series studies is the question of finding and characterising **susceptible subgroups.** As we currently think that not everyone is equally susceptible, we need to find out who is less and who is more susceptible and how this can be influenced. The more susceptible part of the population will probably be persons with existing cardiovascular or pulmonary diseases for whom the (possibly little bit of) extra stress caused by air pollution leads to mortality, but we do not know this exactly. We also do not know the answer to an even more difficult question: whether there is a gradually shifting susceptibility of the population as a whole. In the Western world the number of people with asthma or diabetes, for example, has increased considerably when viewed over a period of decades.

A measure for the health effects that is being used frequently in the full report is the epidemiological concept of **Relative Risk** or RR. By definition a RR is the ratio of the risk of disease in an exposed cohort over a defined time interval compared to the risk of disease in an unexposed cohort over this same interval. Because everybody is exposed to ambient air pollution to some extent, in environmental epidemiology this RR is quite often expressed as a relative increase in mortality, e.g. for a specific increase in PM concentrations. It is important to realise that the Relative Risk concept can be applied not only to cohort studies which compare groups of subjects that experience different exposures because they live in clean or polluted environments; it also applies to time series studies in which the mortality within a population is compared between clean and polluted days.
As an example, a RR of 1.034 per 100 $\mu g/m^3$ increase in PM_{10} for all-cause mortality as recently found in a time-series study in the Netherlands indicates that for a 100 $\mu g/m^3$ rise in the daily average PM_{10} concentrations the risk of mortality by any cause will be augmented by 3.4% on the polluted day compared to the mortality experienced in the same population on a clean day. If this same relative risk had been expressed per

50 µg/m^3 of PM_{10} increase, its numerical value would have been 1.017 instead of the now presented 1.034. It is therefore important to note the amount of PM increase when comparing RR values. Although such RRs may seem "small", they are repeated on every polluted day, and when applied to a large population, the annual number of deaths that can be attributed to such exposures can be quite large.
A RR of 1.000 indicates that the risk in an exposed cohort (or on a polluted day) is not different from the risk in an unexposed cohort (or on a clean day). The **95% confidence interval (CI)** of the RR indicates the precision with which the RR was estimated. Generally, large studies using valid methods produce smaller CI's than small studies or studies using inadequate methods. When the 95% CI contains 1.000, this indicates that the RR presented is not statistically significant for the chosen level of confidence. In the above-presented example of the RR of all-cause mortality for the Dutch population associated with PM_{10}, the 95% confidence interval lies between 1.023 to 1.044. Other particle measures, e.g. $PM_{2.5}$, PM_1, TSP or BS could also be taken instead of $PM_{10,}$ of course. It is also possible to calculate a RR for a specific chemical fraction of PM, e.g. sulphate or nitrate. Grouping different chemical components together by a principal components analysis allows us to explore relations between health effects and the principal sources of grouped components. Sometimes a RR is also expressed in interquartile ranges instead of a certain amount of PM. This interquartile range indicates the difference between the PM concentrations at the 25th and the 75th percentile of the distribution – i.e., the distribution of daily average concentrations when applied to a time series study, or the distribution of long-term average concentrations to which different cohorts are exposed.

2.2.3 Mechanisms of PM-related health effects

A limitation of epidemiological studies is that they, for obvious ethical reasons, have limited possibilities for producing experimental evidence; most of the conclusions are based on observational studies. Toxicology and human clinical studies complement epidemiology, by providing experimental evidence and by investigation of biological mechanism for the health effects. The strengths and limitations of toxicology mirror those of epidemiology, and it is obvious that combination of both lines of investigation provides a better insight in effects of air pollution than either scientific discipline can provide in isolation.

One approach for identifying biological mechanisms is to break down the complex PM mixture into smaller fractions based on the physical, chemical or biological characteristics of PM. **Physical characteristics** are further divided into size (coarse, fine, ultra fine), surface area, number, charge or radiation of particles. **Chemical classification** is roughly done between organic and inorganic or between a water- soluble or non-soluble fraction, and often also by specific groups such as transition metals, salts, hydrocarbons, crustal-like material, etc. **Biological characterisation** is based on contents of moulds, fungi, bacteria (or components produced by bacteria, e.g. endotoxins) pollen, etc. Apart from knowing what is causing the health effects, it is also important to know how these

effects are induced in order to link the epidemiological observations with an underlying **biological mechanism**.

In some epidemiological cohort studies, groups with a higher socio-economic status (SES) did not die prematurely from PM. This does not indicate that a higher degree of education protects against the effects of PM but that probably something in the life style of people with a higher SES gave them better protection compared to people with a lower SES, or that something in the life style of the lower SES groups made them more susceptible. It is very relevant to discover what this is, in order to protect the general population.

A number of other biological mechanisms of action are currently being studied, some of which do not necessarily need to be preceded by pulmonary effects, e.g. neurogenic inflammation or blood coagulability.

Since research into inhalation exposures to PM of human volunteers and experimental animals (***in vivo* research**) is limited, both ethically and cost-wise, alternative techniques have been applied in recent toxicological research into PM.

The simplest way of studying toxicological effects of PM, using cultured cells or organs, is called ***in vitro* research**. These systems have been shown to be very sensitive. PM collected on a filter or directly in a solution can be used to expose these cells or organs. However, the type of exposure is not very realistic and results from these studies are difficult to extrapolate to the human situation. On the other hand, it can be a very useful tool to study mechanisms of PM or to compare PM collected in different places (for instance, with heavy or light traffic) or at different times (episodes).

A second alternative is to use suspensions of collected PM or model components for intranasal or **intratracheal instillation** in animals and human subjects. A solution of the pollutant is introduced direct into the body in a laboratory. This technique is useful for sorting out the dose-effect relationship (or relative toxicity) of PM. However, the manipulation of PM in instillation studies also directly changes original form and particle size distribution (PSD). Also, the manner of administration of the collected PM in instillation studies is completely different from the manner of administration (and hence, the dose) received during normal breathing and should at least be considered as a very short-term high exposure. Nevertheless, these studies have proven to be useful in understanding the contributing factors and confirming some of the epidemiological findings.

Extensive human clinical and experimental animal studies are ongoing to characterise the toxicity of PM. In the case of animal studies, the presumed susceptible part of the human population is mimicked by inducing specific cardiopulmonary diseases or focusing on senescent animals. For instance, animal research has been done in the laboratory using an asthma model, pulmonary hypertension, pulmonary inflammation and systemic hypertension. A drawback with these **disease models in animals** is that they are

not completely equivalent to the human disease status. They are, as the name says, only a model. A second issue is that you never really know if the laboratory animal you are testing is representative of the reactions of a human being, or if you are looking at endpoints that are sensitive enough and representative for the human endpoints. Or the timing of exposure and observation may not have been right in the animal studies.

Most of the toxicological studies are driven by hypotheses on specific fractions of PM_{10}: acidic aerosols, UF, organic fractions, mixtures of particles and gaseous compounds, transition metals and particle charge. Often, a single constituent is tested, while the ambient PM is a complex mixture which may have undergone different atmospheric reactions or can result in interactive effects. Negative results in toxicological studies are nearly as difficult to interpret as positive findings.

So nowadays, special technologies have been developed which only concentrate the PM without affecting the rest of the air pollution mix. With these concentrators it is possible to increase PM levels in the exposure atmospheres up to 30-80 times. Specific size ranges (coarse, fine, ultra fine) can be selected for study. Using these **concentrators** more or less guarantees that laboratory animals and human volunteers are exposed to the actual ambient PM mix, but in a more concentrated form so health effects may be easier to detect.

Figure 5 presents a simplified scheme for possible mechanisms of health effects associated with PM which can be used to form an idea of the complexity of the PM enigma. The different concepts presented in Figure 5 and their relationships are treated in more depth in the full report, as space limits a more extensive description in this executive summary.

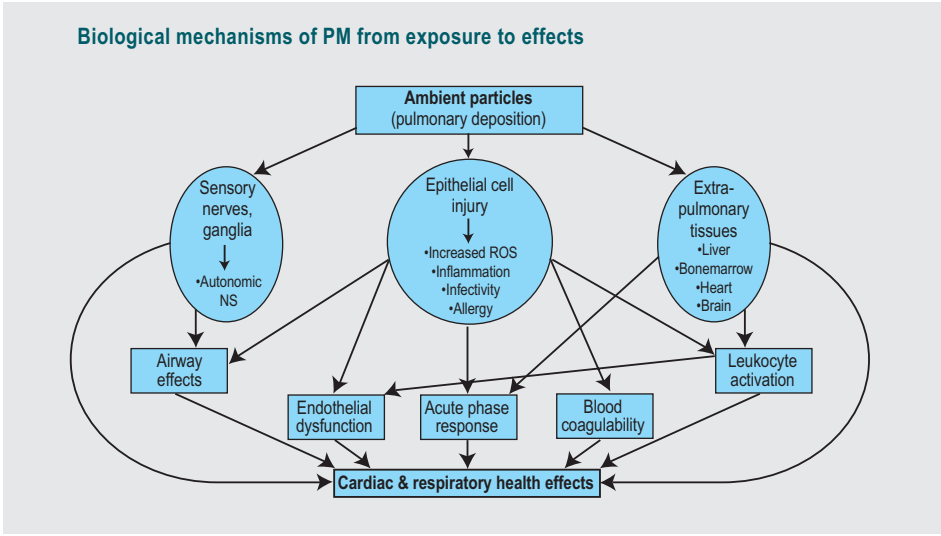

Figure 5. Possible mechanisms from exposure of PM to effects.

3 HEALTH EFFECTS ASSOCIATED WITH PM

3.1 Assessment and quantification of health effects

Epidemiological studies present worldwide evidence for particulate matter (PM) associated health effects in the general population, as has been described in Section 2 on basic concepts and in the full report. The current time-series studies have led to more than a hundred replications worldwide of the associations between ambient air pollution and health effects. The criteria for judging causality for environmental epidemiology, based on the work of Hill, can be considered to have been largely met for health effects of ambient PM. PM is not only associated with mortality, but also with less serious health effects indicated by hospital admissions or medicine use. PM appears to be associated more with diseases involving the respiratory tract or cardiovascular system, as one would expect it to be. Individuals are not all equally susceptible to PM-associated health effects.

Elderly people with existing respiratory or cardiovascular diseases seem to be at greater risk. There is increasing coherence between epidemiology and toxicology as far as the mechanisms are concerned; there is a growing body of toxicological data providing some degree of biological plausibility of the different mechanisms for epidemiological health outcomes, but there is also still conflicting evidence – as one would expect given the complexity of the problem.

Table 2 presents the results of the most recent 7-year time-series analysis of PM-associated premature mortality in the Netherlands. As stated previously in Section 2, other health effects that occur, such as hospital admissions, can also be described by means of a RR. However, in this executive summary premature mortality is used as an indicator of other health effects that are not reported here in detail. The reader is reminded that when mortality is reported, this is only the 'tip of the iceberg' and that many other less than fatal health effects in the population are implicated.

Table 2. Relative Risk (RR) for mortality at lag 1 for PM_{10} and different causes of death in the Netherlands per 100 µg/m^3 increase in PM_{10} (1992-1998).

	RR	95%	CI
Total mortality	**1.034**	1.023	1.044
Respiratory mortality	**1.115**	1.079	1.151
COPD mortality	**1.105**	1.057	1.154
Pneumonia mortality	**1.115**	1.059	1.173
Cardiovascular mortality	**1.024**	1.008	1.041

Estimated using the strictest (new) convergence criteria

Epidemiological studies in the Netherlands present evidence for PM-associated health effects. These Dutch observations are in line with the international scientific literature.

The health effects in the Dutch population associated with ambient particulate matter (PM) are large when based on the numbers of people involved and serious when their nature is taken into account. On the basis of the relative risk (RR) from the epidemiological time-series studies in the Netherlands presented in Table 2 it is estimated that approximately 1,700 premature deaths per year are associated with particulate matter in the ambient air. When the average results of Europe-wide and US time-series studies are used for a similar extrapolation instead of the Dutch mortality studies, a figure of 3,000 premature deaths is calculated for the Netherlands. Both of these estimates are for the acute effects of air pollution. If the results of two of the four US cohort studies could be quantitatively extrapolated to the Netherlands, even higher figures of 10,000 to 15,000 deaths per year would emerge for long-term exposure to air pollution. The uncertainty of the quantification increases from acute to chronic effects, due also to a lack of fully representative data for the situation in the Netherlands.

The quantification of chronic PM exposure in relation to health effects is a major challenge. Some researchers, weighing all the available evidence, conclude that the US results can be transferred quantitatively to the Netherlands. Others currently urge greater caution, pointing to the conflicting results in some of the US studies and a different pollution mix for the US and the Netherlands. More research is needed to increase the confidence in the type of quantitative transfer presented in the previous paragraph.

Based on the time-series studies, approximately 1% of mortality in the Netherlands is associated with acute exposure to PM_{10}. When data from the chronic studies performed in the USA are transferred to the Netherlands this figure is nearly an order of magnitude higher. In spite of the ongoing scientific debate and prevailing uncertainties concerning the quantification of acute and chronic health effects, the overall conclusion is that PM-associated health effects are so extensive and serious that further action is warranted.

One of the most consistent results of the different epidemiological studies is the fact that there does not seem to be a threshold in the concentration below which no effects of air pollution on health occur. Therefore no standard can be derived in which there is a ‘safe’, no-effect situation for the population. It also indicates that regular toxicological standard setting for non-carcinogens with a No Observed Adverse Effect Level (NOAEL) and use of ‘safety’ or, better, ‘uncertainty’ factors is not possible for PM. For this reason, the choice of a quantitative standard for PM is currently shifting from the scientific to the policy domain, which must decide what level of risk still has to be accepted. Science can inform the decision making process by providing accurate exposure-response relationships, by providing sound estimates of the costs involved in various pollution abatement strategies.

PM is associated with serious health effects, for which there does not appear to be a threshold. This precludes regular standard setting with an NOAEL and the use of safety factors. It implies that for any PM standard a certain level of impact on health in the population will have to be accepted.

A further similarity in the different studies is that the reported concentration-response curves are almost linear, although they seem to level off at higher concentrations. Concentration–response curves certainly do not rise steeply at higher PM concentration levels. The actual form of the concentration-response curve indicates that over a year, more of the health effects occur on days with average concentrations than during episodes with high concentrations – simply because there are many more days with 'average' concentrations than days with episodic, high concentrations. This differs from the historical situation with episodes in the past, when large numbers of victims were associated with the high concentrations during and immediately after these episodes. Preventing episodes is probably less effective in reducing total mortality and morbidity than bringing down the average levels of PM, because combatting episodes does not necessarily reduce long-term average concentrations, whereas reducing long-term averages necessarily also reduces episodic high concentrations.

The shape of the concentration-response curves indicates that the numbers of cases of PM-associated health effects at average concentrations contribute more to the total risk than those during episodes do. Lowering yearly average values of PM is probably more effective than concentrating solely on the prevention of episodes.

Coherence can be found in the fact that study-averaged values of the RR of time-series in the USA (NMMAPS) and Europe (APHEA2) are numerically quite similar. This overall similarity of the RR at the same time also includes dissimilarity, as geographical differences in RR within the studies are approximately a factor of three, and local influences may lead to differences between the various studies. These differences are covered by the term heterogeneity within the different studies. A considerable part of the existing variation between study areas is probably influenced by local circumstances such as the age distribution within the exposed populations, the sources contributing most to the PM etc. The fact that heterogeneity exists is important because this provides clues for the critical causal factors and hence for policy measures.

There seems to be heterogeneity (differences in the size of effects probably influenced by local circumstances) between locations within the various epidemiological time-series studies for PM-associated health effects.

As mentioned previously, mortality has been associated in more than a hundred time-series studies with PM or with air pollution in general. Usually, at least when they were studied, associations appear not only with PM but also with the ambient levels of gases such as O_3, SO_2, CO and NO_2. In two-pollutant models with PM, these associations with a number of gases generally remain significant when the two-pollution models correct for the PM

effects. This is usually explained by concluding that the gases have an independent effect, indicating that it is necessary to look at the whole ambient mixture instead. Statistically, it is very difficult to tease these different effects apart, as the daily concentrations of the various components of air pollution (apart from ozone) all have a high mutual correlation. The driving forces for the daily levels are the prevailing meteorological conditions, because the magnitude of the emissions from the different sources in the Netherlands is relatively continuous whereas daily differences in meteorology can be substantial.
An additional study in the US has shown that the ambient gaseous concentrations at a central site correlate with the personal $PM_{2.5}$ exposure concentrations but not with the personal gaseous exposure concentrations. This provides evidence that the ambient gaseous concentrations at a central site act as surrogates for personal $PM_{2.5}$. This might also be the case for other PM size fractions, like PM_{10}, but these data were not measured. So in this executive summary and the accompanying full report we will concentrate on PM and not on air pollution in general.

Ambient gaseous concentrations at a central site correlate with the PM exposure concentrations measured using a personal monitor, which are a measure of the exposure of the individual, but not with the personally measured gaseous exposure concentrations.

A recent Dutch time-series study in the city of Amsterdam showed that people living very close to major roads (with approximately twice the primary traffic contribution compared to an urban background) had a higher RR for all-cause mortality than those living in the average urban background in Amsterdam. However, on a larger geographical scale - at a national level instead of close to major roads - another recent Dutch time-series study indicated that the RR for all-cause mortality in the four major cities of the Netherlands is not different from the RR in the more regional part of the rest of the Netherlands. Average PM_{10} levels are almost equal for both situations, whilst the primary PM contributed by traffic is almost doubled in the urban background compared to the regional background situation in the Netherlands. If traffic had been the sole source of PM-related health effects, this would have been revealed by a higher RR in the major cities. However, concentrations measured at regional background stations are likely to underestimate exposure of the population, which also outside the four major cities lives primarily in urbanised or semi-urbanised areas with traffic exposure. The difference between the two studies may therefore not be all that great.
Principal-component analyses of different fractions of PM in the US point towards traffic as a source of the health-relevant fraction of PM. Other Dutch and foreign studies also provide evidence that health effects are related to the distance from a major road or to traffic density, and to heavy-duty traffic more specifically. The results of a Dutch cohort study clearly point to more health effects at addresses near major roads.

Traffic seems to be a major source of health-relevant PM.

Recently, ultra fines (UF) have come into the picture as a possible cause of PM- associated health effects. A substantial part of UF is emitted by traffic. Toxicological experiments

have indicated that ultra fine particles could produce serious health effects in laboratory animals. The UF are smaller than 0.1 µm in diameter and mainly produced by combustion sources. Ongoing studies in Los Angeles suggest that ultra fines are more potent compared to fine or coarse mode PM and that the potency could also be related to the distance from a highway. In the Netherlands ambient levels of UF and PM_{10} do not correlate, so if in future Dutch effect research the UF fraction were also to be associated with health effects, this would possibly point to a different mechanism than that for PM_{10} health effects.

The available toxicological and epidemiological evidence is less complete for ultra fines (UF), though this also is a field which needs more research as the potential health implications of UF may be considerable.

Sometimes a contradiction is seen in the fact that numerous epidemiological studies all point to ambient air pollution playing a part in causing health effects, but that toxicology has difficulty in confirming this evidence by finding a plausible mechanism for the associated health effects at the current concentrations. Around the middle of the last century there were a number of environmental disasters involving high concentrations of PM (as well as other pollution) which led to large numbers of deaths. Exposures more comparable to the current situation were observed during two recent events reported in peer-reviewed literature: Utah Valley, with the closure for over a year in the winter of 1986-1987 of a steel mill causing heavy pollution and the reunification of Germany in 1989, after which a great number of polluting industries were shut down and air pollution decreased. In both situations lower levels of ambient pollution have resulted in a subsequent decrease in population health effects. These situations cannot, however, be generalised to the ambient PM situation in the Netherlands, as the principal sources, composition and levels of PM are partly different.

In the past, the abatement of high PM levels in a number of specific situations has subsequently led to a decline in health effects in the population.

3.2 Risk reduction and differentiation of PM

In order to link current knowledge on PM to the influence of PM abatement requires the analysis of a more extensive PM time-series. To demonstrate how policy measures have changed ambient PM levels and to assess how this may have influenced PM- associated health effects, we need to look at historical levels of PM. When looking at a longer period of time, we find that levels of PM and other (gaseous) air pollution have decreased considerably in Western Europe. There is information on historical pollution levels in the UK spanning a large period of time. For the Netherlands the levels of BS have more than halved over the quarter of a century between the 1960s and the 1980s (see Figure 6). This decline is largely due to environmental abatement measures taken by industry and authorities during this period and coincides with a shift from oil and coal for domestic heating in the Netherlands to the use of natural gas. Decreases in the 50-percentiles

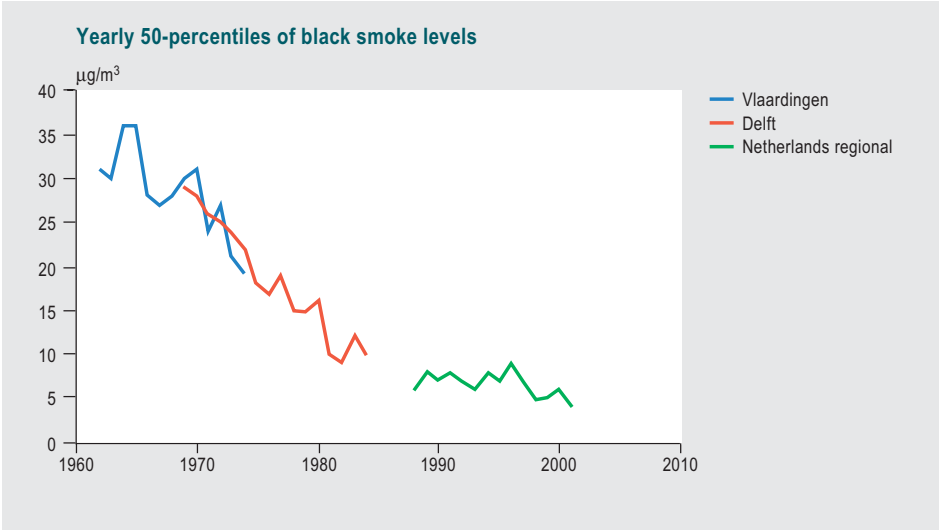

Figure 6. Yearly 50-percentiles of black smoke (BS) levels in µg/m^3 in Vlaardingen and Delft 1962 -1984 and Netherlands Regional average of 10 sites 1988 –2001

of BS at the regional measuring sites over the past decade have become more gradual, similar to the decreases in average PM_{10} levels during that period presented in Figure 6.

There are two key questions connecting ambient aerosol levels and PM risk assessment.

1. Will lower levels of PM lead to a reduced environmental risk for the Dutch population in general?

2. Does every fraction of PM (whether chemical, physical or source-related) contribute proportionally to the PM risk assessment or are there specific sources or fractions that have a greater or lesser relevance to health effects than others?

There is insufficient scientific evidence available on current environmental problems at ambient concentrations in the Netherlands to answer the first question. The previously presented examples of Utah Valley and former Eastern Germany indicate that in those situations, which were different from prevailing Dutch levels, the answer can be affirmative.

At first sight it seems likely that a further decrease in PM levels will lead to a reduction in health risks. However, PM is a complex mixture with more and with less health-relevant fractions. Changes in the composition of this mixture might change its health impact.

Although there seems to be a difference in potency of PM fractions based on toxicological and human clinical testing of the pure substances, it remains unclear whether the

toxic effects of these fractions in the real PM mixture are similar. Testing of individual pure fractions will not solve the problem of some as yet unknown synergistic effects of a mixture of, as such, innocuous substances. Based on our experience the possible occurrence of a mechanism of synergism can never be ruled out scientifically. An alternative explanation might be chemical interactions that alter the composition of ambient PM in such a way that the highly toxic and reactive compounds formed might not be found after sampling (e.g. active radicals). As such, epidemiological observations about the effects of ambient air pollution will be difficult to reconcile with toxicological information on different pure fractions of the ambient PM mixture.

The evidence concerning sulphate-induced health effects at ambient levels provided by epidemiology and toxicology is paradoxical and may have implications for abatement measures. There is strong evidence from epidemiological studies pointing to a role of sulphates (part of the SIA and originating from SO_2) in relation to health effects. Large fractions of Dutch ambient particles are ammonium nitrate and sulphate, together with sodium chloride. However, in the full report the toxicological and human clinical studies presented in more than 150 publications supporting a causal role of sulphate and other components of SIA have been evaluated. No significant adverse health effects have been observed for the pure components of these substances following exposures which are an order of magnitude higher than ambient levels in the Netherlands. In those studies in which effects have been observed, these effects have to be attributed to the acidity in sensitive subjects (asthmatics). In addition, ambient PM nitrates and sulphates are soluble in water, and normal concentrations in body fluids in the lung tissue are at least an order of magnitude higher than those that could be achieved from the absorbed dose by way of inhalation. The current toxicological and human clinical evidence does not support the epidemiological observation that sulphate is a causal factor at current concentrations in the ambient air in the Netherlands. It therefore remains unclear whether or not a reduction in sulphate (as well as in nitrate) concentrations in ambient air will result in a similar reduction in health effects in the general population. The recent results of the Dutch 7-year time-series study even suggests that lower average levels of sulphates do not necessarily lead to lower health effects in the Dutch population.

A number of US epidemiological studies indicated that days with high levels of crustal material did not necessarily coincide with more health effects in populations. This result ties in nicely with a principal-component analysis of PM_{10}, which showed that in the US the crustal fractions do not seem to be associated with health effects. However, some other US studies in the arid areas of the country show that the coarse fraction of PM_{10}, which is predominantly crustal, *is* associated with health effects. However, results of this kind from semi-arid areas are probably not applicable to the more temperate situation in the Netherlands. For years crystalline crustal material (e.g. quartz dust) has been used as a standard dust and a positive control to elicit health effects in some biological studies. Non-crystalline crustal material is probably less health-relevant.

PM has to be seen as a complex mixture of fractions with greater and with lesser health relevance. The most efficient and cost-effective reduction of health effects will be achieved by reducing the most toxic part of PM. Although significant progress has been made over the past few years, there are currently only suggestions for the causal fractions.

When the health effects of various fractions of PM differ, the question arises of whether it can be easily categorised by size to simplify abatement measures. At first sight, such a separation by size might seem to be a convenient way of dividing the ambient PM mixture into a less and a more health-relevant fraction. Though crustal material is concentrated predominantly in the coarse size fraction and approximately half the sea salt is in this fraction, such a crude separation on size only is not advisable. Toxicological studies in the Netherlands and elsewhere have indicated that the coarse fraction of PM (with diameters between 2.5 and 10 µm) certainly provokes toxicological responses in laboratory animals and in *in vitro* studies.

Not including the coarse part of PM_{10} in a health-oriented standard may result in a relevant fraction being missed.

In epidemiological research finer particles seem to be more relevant for health effects than coarser particles. Whether the various indicators compare differently with regard to the causation of health effects is a question that cannot yet be answered.
The sometimes reported associations between UF and health effects warrant further research in order to arrive at a clearer picture. In the Netherlands the correlation between UF and other PM metrics seems poor, which may suggest that an additional separate effect of UF may exist, with possibly other mechanisms of action. However, current toxicological, human-clinical and epidemiological information on UF is insufficient to base a standard on.

Nevertheless, arguments are emerging that standards in the smaller ranges of PM, but not as small as UF, might be useful. A number of arguments support a $PM_{2.5}$ standard, as there are numerous scientific papers in which associations between $PM_{2.5}$ and health effects have been reported. Arguments of applicability call more for putting the future cut-off at 1 µm, because the crustal fraction will then be eliminated from the PM mixture. For southern European countries affected by Sahara dust this would be an argument to consider. However, less information is available on PM_1 at present.

In the future other PM indicators will probably be appropriate to supplement or to replace the current PM standard. It is recommended to develop a size or source related standard for fine, accumulation mode particles using a cut-off in the 1 – 2.5 µm range in view of the large body of evidence that has accumulated concerning adverse health effects of fine PM. Such a standard should supplement rather than replace a PM_{10} standard in view of the indications of adverse effects of the coarse PM fraction. It is, therefore, recommended that PM_{10} also be retained as a standard for the time being. Research is needed to establish whether in

addition, other standards need to be developed for instance for the ultra fine particle fraction.

Apart from crustal material and SIA, the toxicological database on PM health effects is not yet at a stage where it can provide clear answers regarding causal factors, whether using a physical (surface area, charge, radiation), chemical (e.g. transition metals, organic substances), or biological (viruses, moulds, spores, bacteria or products of bacteria such as endotoxins) entity. Quite a large amount of specific toxicological evidence seems to correspond to the results of epidemiological studies. An understanding of what in this complex mixture determines its toxicity would be of assistance in PM monitoring and control.
Intratracheal instillation studies, for instance, show dose-dependent health effects related to the chemical composition, albeit at fairly high exposure levels and - unlike epidemiological concentration-response relationships- with a threshold. Also, urban dust shows more health effects than rurally collected dusts, with some evidence for a traffic contribution. Several studies have shown that coarse and ultra fine mode PM can induce inflammatory responses, sometimes even stronger than the fine mode fraction. Other studies conclude that not the mass but the (reactive) surface area of PM is a better metric to link health effects with PM. The real problem, however, lies in the extrapolation of these toxicological results to the general population and their ambient exposures. All manipulation of collected PM in instillation studies tampers with the material, influencing the size distribution and the fraction that goes into the solution and the manner of exposure. It is therefore questionable whether the material used reflects ambient PM and how the results found should be interpreted. The possibility of studying the real-world effects of coarse, fine and ultra fine PM with concentrators promises to be very useful, but results have only recently started to emerge. No consistent health effects in either studies with dogs and rodents or human volunteers have yet been reported in the scientific literature. Slight changes in health parameters are, however, sometimes observed, but at present none of the groups applying the techniques is at a stage able to provide clear answers to the question of what is causing the health effects. Preliminary evaluations of studies using the only European ambient fine particle concentrator in the Netherlands indicate small but significant health effects in compromised animals. This field of real-world particle inhalation toxicology and human clinical studies is a completely new field of toxicology and probably still needs to mature for a couple of years.

To overcome possible artefacts introduced by fractionation and sampling, future toxicological studies can use concentrators to ensure exposure to the ambient PM mix.

PM fractions are able to induce inflammation and immunotoxicity in airways and lungs via oxidative stress or via a neurological mechanism by impairing respiratory and cardiac/neurological functions. Rather than the association between PM and mortality being assigned to the toxicity of PM itself, a more plausible explanation is that this association is the result of an organism's reduced capacity to withstand (oxidative) stress and maintain a stable, relatively constant internal environment. It is therefore likely that

susceptible individuals with failing health, attributable to ageing or illness, largely comprise the population at risk.

The current data have not yet produced sufficient evidence to demonstrate convincingly one mode of toxicological action that explains PM health effects at ambient levels, although it is likely that primarily susceptible people are at greater risk.

In the Netherlands the average life span is increasing. This gradual process of ageing has consequences for PM risk management, because the sub-groups presumably susceptible to the PM-associated health effects will become larger in relative and in absolute numbers in the future. As has been presented above, there are still a number of fundamental uncertainties concerning PM and its health impact. These will not be cleared up soon and so necessitate a long-term research effort. While it may be possible to succeed in reducing the PM problem in terms of air quality, it should be remembered that the size of the susceptible sub-population will increase and that exact quantification of the future health impact will become more complicated.

Concentrations of PM in ambient air will decrease in the future. Despite the improved air quality, it could be conjectured that the health impact associated with PM will nevertheless become more pronounced. In the Netherlands the gradual ageing of the population and other demographic developments might lead to a more than proportionate rise in the susceptible sub-groups. However speculative the previous remark, continuing vigilance seems required for this only partially understood problem of PM.

4 CURRENT AND FUTURE LEVELS OF AMBIENT PM AND PM EMISSIONS IN THE NETHERLANDS

4.1 Current PM concentrations and source contributions

In 2001, a yearly average PM_{10} concentration of 31 µg/m^3 was measured at regional sites in the Netherlands. Further details concerning current and future levels of PM and emissions in the Netherlands can be found in Chapters 2 and 6 of the full report.
In the Netherlands, which is virtually one air shed, the concentrations measured by the National Air Quality Monitoring Network (NAQMN) are representative for a larger space than just the vicinity of the monitoring site. Correlation of daily PM_{10} data is in the order of 0.6 to 0.8 between the various monitoring stations in the country. A high correlation indicates that the variation in daily levels of PM is evenly distributed over the Netherlands. Therefore, monitoring at central sites in the Netherlands adequately covers the day-to-day variation in exposures.

It has been shown in studies in the Netherlands and elsewhere that the correlation in time of personal and outdoor mass concentrations of $PM_{10,}$ and especially of $PM_{2.5}$, is reasonably high. This result is confirmed in other current time-series studies. For light absorption (as a marker for EC) the correlation coefficients were even higher than the 0.8 found for $PM_{2.5}$. In the Netherlands, in the absence of air conditioning and indoor sources, PM concentrations indoors are generally a fair reflection of the ambient PM levels outdoors. In absolute terms the outdoor part of PM that ends up indoors is a considerable fraction (0.6 to 0.8) of the ambient PM in the Netherlands. Research using "personal" monitors has indicated that outdoor levels are representative for personal exposure to ambient PM indoors in residential homes.

The levels of PM_{10} measured by a stationary monitor seem to be representative for the personal exposure of the general public to ambient PM_{10} in the residential environment.

The field of PM emissions and modelling has been developing in the past few years in the Netherlands. The emission databases for PM_{10} from anthropogenic sources and precursor gases of Secondary Inorganic Aerosols (SIA) in the Netherlands and European Union have been updated by RIVM and TNO and used in long-term dispersion models to estimate the yearly average PM_{10} levels. These anthropogenic emissions account for approximately half of the currently measured PM levels in the Netherlands. The other half is composed of PM that is not contained in the emission databases. Most of it is sea salt (4 to 7 µg/m^3) and (anthropogenic) crustal or natural material (3 - 4 µg/m^3). Approximately 1 µg/m^3 is a non-modelled contribution from the northern hemisphere. Due to a lack of adequate measurements it is not yet possible to present a reliable non-modelled value for $PM_{2.5}$.

Table 3. Emissions of PM_{10} in ktonnes per year by different economic sectors in the Netherlands

	1980	1995	2005	2010GC
Transport	32.6	21.2	15.9	11.7
Industry	52.8	21.3	13.0	12.9
Consumers	4.5	3.9	3.5	3.5
Agriculture	7.9	9.7	8.8	9.3
Storage and handling	2.0	2.8	2.6	2.7
Waste incineration	4.3	0.1	0.1	0.1
Energy sector	11.0	0.7	0.6	0.7
Other	0.8	1.1	1.3	1.5
TOTAL	**115.9**	**60.9**	**45.8**	**42.0**

Taking into account that a number of fractions and chemical compounds have not been measured, a non-modelled average value of 18 µg/m3 is currently added to the modelled anthropogenic PM_{10} fraction.

The PM_{10} emissions from 1980 to 2010 are presented in Table 3. The 2010 emissions are those of the Current Legislation of Emissions (CLE) scenario under Global Competition (GC), implying measures that have already been agreed upon.

Modelling of yearly average PM concentrations with long-term meteorology is done on 5×5 km^2 grids in the Netherlands. Only a few urban and industrial grids of the more than a thousand (5×5 km^2) Dutch grids are currently assessed to show yearly average PM_{10} concentrations above 40 µg/m^3. The modelled geographical distribution for 2005 is presented in Figure 7.

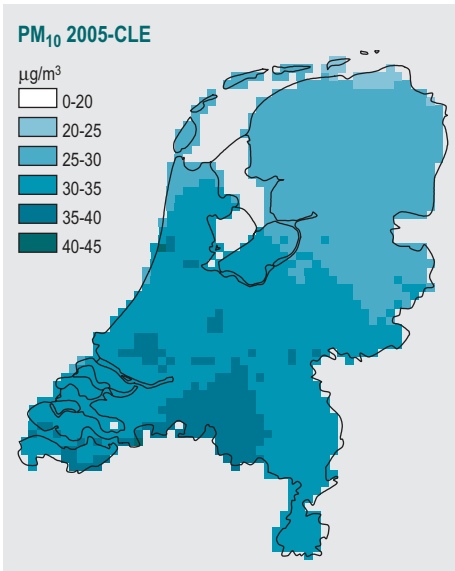

Figure 7. Modelled anthropogenic yearly average PM_{10} concentration in µg/m^3 with an additional non-modelled background of 18 µg/m^3 in 2005 in the Netherlands 5×5 km^2 grids

Table 4. Annually averaged primary and secondary inorganic concentrations of PM_{10} averaged over the Netherlands by anthropogenic source. Calculated for the year 1995, based on emissions for the Netherlands and the CEPMEIP inventory for European countries.

Dutch sources	Primary PM_{10} ($\mu g/m^3$)	NH_x ($\mu g/m^3$)	NO_y ($\mu g/m^3$)	SO_x ($\mu g/m^3$)	Summed concentration ($\mu g/m^3$)
Industry	0.4	0.0	0.1	0.1	0.6
Energy	0.0	0.0	0.1	0.1	0.2
Transport [1)]	1.5	0.0	1.0	0.1	2.6
Agriculture	0.5	0.9	0.0	0.0	1.4
Others	0.5	0.1	0.1	0.0	0.7
Sum	**2.9**	**1.0**	**1.4**	**0.2**	**5.5**
Other countries					
Industry	0.9	0.0	0.1	0.1	1.0
Energy	0.4	0.0	0.7	1.9	3.0
Transport 1)	0.9	0.0	2.1	0.3	3.3
Agriculture	0.1	1.2	0.0	0.0	1.4
Others	0.7	0.0	0.5	1.1	2.3
Sum	**3.0**	**1.2**	**3.3**	**3.4**	**10.9**
All sources					
Sum	**6.0**	**2.2**	**4.6**	**3.6**	**16.5**

[1)] Including international shipping

The long-term PM situation appears to be developing favourably, as can be seen in the measured PM concentrations presented in Figure 8. Similarly, the 24-hour averages are also decreasing, but not enough to meet the EU standards. This will be discussed in Section 5.1.

The dispersion model (OPS) calculates a yearly average PM_{10} level of 16.5 $\mu g/m^3$ (Table 4) based on the 1995 emissions (Table 3) using long-term meteorology. This calculated value needs, of course, to be augmented by the non-modelled part of the PM (18 $\mu g/m^3$): sea salt, crustal and biogenous material and the northern hemisphere background, as previously indicated. This results in an average concentration of 35 $\mu g/m^3$, which is slightly lower than the concentration actually measured in 1995 (38 $\mu g/m^3$). When the uncertainty in the emission database, the dispersion modelling and the PM measurements is taken into account, this agreement is good.

The modelled contribution for emissions from the Netherlands was 5.5 $\mu g/m^3$ and that of foreign countries 10.9 $\mu g/m^3$. Transport (including shipping) contributes the major part (5.9 $\mu g/m^3$), of which 2.6 $\mu g/m^3$ is the result of emissions in the Netherlands. It is estimated that 6 $\mu g/m^3$ of the anthropogenic PM is of primary origin, whereas 10.5 $\mu g/m^3$ is Secondary Inorganic Aerosol (SIA). By means of measurements taken at Cabauw (in the centre of the Netherlands) at heights of 20 and 200 m, ECN has established that the

yearly average foreign contribution of PM_{10} to Dutch aerosol levels is in the range of 7-17 µg/m^3. Such measurements corroborate a modelled long-term foreign contribution, which is of a similar order of magnitude.

In the Netherlands yearly average measurements of PM_{10} are in good agreement with modelled concentrations, taking into account the indicative contribution to the concentrations of the 'non-modelled' fraction largely of natural origin.

In annex A a similar calculation is presented for the annual average $PM_{2.5}$ concentrations in the Netherlands. It has to be stressed that due to a lack of reliable annual average measurements of $PM_{2.5}$ it is not yet possible to quantify the non-modelled fraction reliably for this size category of PM.

A meteorological analysis (presented in Figure 8) showed that the decrease of approximately 10 µg/m^3 in average PM_{10} concentrations in the last decade does not have to be attributed to meteorological influences. This decrease is the result of emission reductions in the Netherlands and elsewhere in Europe. The higher PM concentrations in the year 1996, and to a lesser extent in 1997 were due to two extremely dry and cold winters.

A difficult problem that currently complicates assessment of compliance is the correction factor actually used. Also, correction factors for automatic PM measurements are in use elsewhere in the EU. The readings of the PM measurements in the National Air Quality Monitoring Network (NAQMN) are corrected as a rule by multiplication by a factor of 1.3 to compensate for losses of semi-volatile material. It has been established in the present study that this correction is time- (season/temperature) and place-dependent.

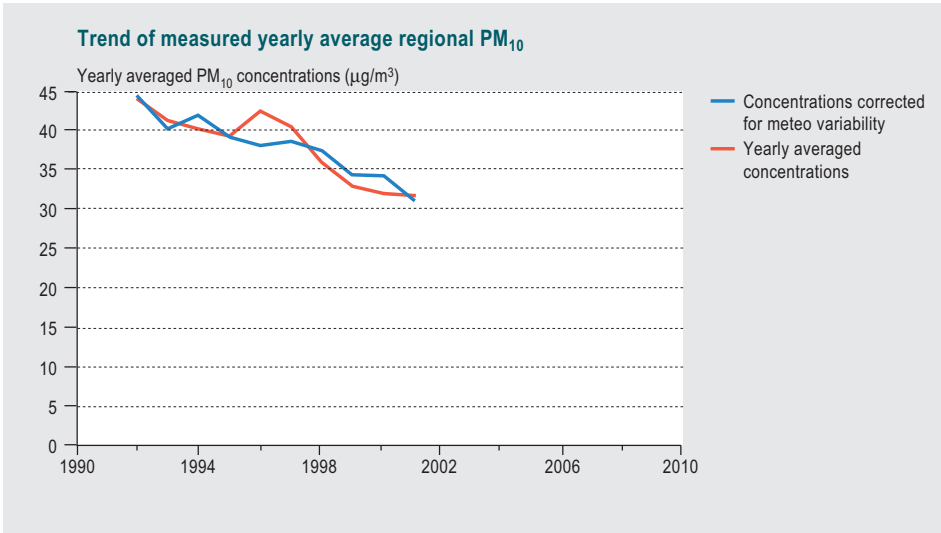

Figure 8. Trend of the measured yearly average regional PM_{10} concentrations 1992-2001 in the Netherlands and the meteorology corrected series.

This produces quite some uncertainty concerning the 'true' concentrations of PM and their spatial distribution.

Accurate measurement of PM is complicated and the automatic PM_{10} monitoring network in the Netherlands uses a factor of 1.3 to correct for losses of semi-volatile material.

This situation with a substantial correction factor on the one hand makes compliance with standards difficult to assess and on the other also results in crude PM levels that cannot easily be used very reliably for epidemiological research.
More extensive PM measurements relating to PM_{10}, $PM_{2.5}$, particle number and composition (e.g. elemental and organic carbon content), possibly with a high time-resolution as well, are needed at this stage to provide a sounder basis for ongoing and future epidemiological and toxicological research. Studying the role of atmospheric processes resulting in chemical conversions or radical formation of PM might also be relevant. A complete chemical speciation of the organic material and the semi-volatiles will be essential. Due to the high correlation of PM metrics a few typical sites in the Netherlands could suffice to provide a reasonable picture of the temporal and spatial distribution of this better characterised PM. In particular, these typical sites should take into account the need for more experimental data on the impact of traffic on PM in urban areas. A second issue that could be addressed at these sites is the need for better insight into the mass of semi-volatiles currently lost in the measurements taken by the available automatic instruments. The presently used correction factor of 1.3 is substantial and produces cruder, and on a daily basis incomplete, PM data which do not allow a better understanding of the still unknown parts of the PM problem. Better knowledge of the actual correction factors (spatially and seasonally) is essential to demonstrate compliance in the future with the EU standards. Furthermore, a future standard for a smaller PM fraction (whether $PM_{2.5}$ or PM_1) needs to be measured reliably. At present, correction factors are needed because of the loss of semi-volatile components. These components are to be found in a higher proportion in the fine fractions than in the coarse fraction of PM_{10}. So based on the current measurement principles, a correction factor for this smaller PM fraction would probably be even larger than that for PM_{10}. An undesirable situation like this should be prevented before a new and different particle metric is promulgated as a standard.

Measurements of PM should be made more accurate. More information on the specific chemical composition and size distribution of PM representative for typical situations in the Netherlands should be generated in a way that it facilitates the testing of relevant hypotheses concerning health effects, source contributions and possibly atmospheric influences.

Research on health effects has indicated that the transport sector is an important source of PM emissions. Therefore, a more accurate estimate of its contribution to the various fractions of PM is desirable in order to quantify its health impact. Primary traffic-related PM emissions are Elemental Carbon (EC), Organic Carbon (OC) and ultra fines (UF).

Table 4 presents the traffic contribution to the SIA by the secondary route through precursor gases. Accurate carbonaceous measurements are quite complicated. The reported EC and OC values for carbonaceous PM therefore have large confidence intervals. RIVM measurements have indicated that in the regional parts of the Netherlands central estimates of yearly average levels of Elemental Carbon (EC) are 0.7 µg/m^3 and of Organic Carbon (OC) 3.9 µg/m^3.

Traffic-oriented measurements in the Rotterdam area made by TNO indicated that the average local elemental carbon contribution of a highway with heavy traffic was 1.7 µg/m^3. This figure is quite similar to the values modelled by TNO (Figure 9). The measured average urban background of EC in Rotterdam was 1.4 µg/m^3. Reasonably similar urban background measurements made by RIVM in a prior year came to 1.6 µg/m^3, indicating good agreement. Tunnel measurements in Amsterdam have shown that the contribution of Organic Carbon (OC) by traffic is of a similar magnitude to that of EC. At an urban background site the average EC + OC contribution made by traffic would therefore be approximately 3 µg/m^3. This value compares well with the long-term modelled primary traffic concentration of 3.5 µg/m^3 in the corresponding urban background grid. For highways, tunnel measurements in Rotterdam made by TNO established a high contribution to the particle numbers. A current emission factor of 10^{14} UF particles per vehicle km is reported as a fleet average. In the late 1970s, similar UF emission factors were still approximately 10^{16} particles per vehicle km. Long-term health effects of living near major roads are being studied in an ongoing epidemiological cohort study in the Netherlands.

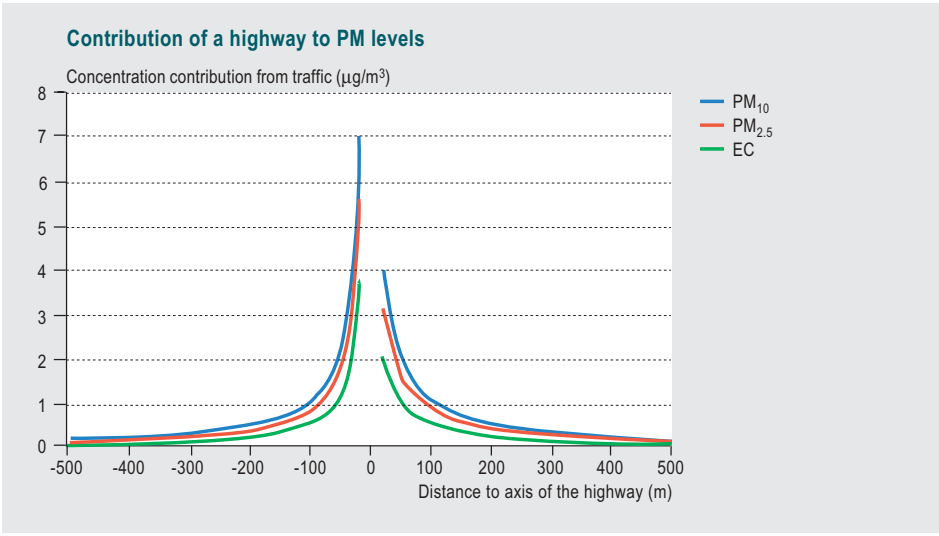

Figure 9. The modelled contribution by TNO of local highway traffic to annual concentrations of PM_{10}, $PM_{2,5}$ and EC (µg/m^3) in Rotterdam (the Netherlands) as a function of the distance to the axis of the highway.

Traffic is an important source of carbonaceous PM and ultra fines, which are emitted at breathing height, close to a large part of the population in the Netherlands.

4.2 Future trends in PM concentrations at current control policies

A comparison of the calculated PM levels for 2010 with 1995 shows that, on average, the yearly concentrations of modelled anthropogenic PM_{10} will decrease by 5.4 $\mu g/m^3$ from 16.5 to 11.1 $\mu g/m^3$ for the Netherlands. Of this decrease, 1.2 $\mu g/m^3$ results from reductions of primary PM_{10} mainly in the transport sector; the greater part (0.9 $\mu g/m^3$) is due to abatement in the Netherlands.

With respect to 1995, international acidification policy has reduced SIA concentrations by 4.1 $\mu g/m^3$. So, 75% of the downward trend between 1995 and 2010 is the outcome of emission reductions of the precursor gases SO_2, NO_x and NH_3. In total, nitrate concentrations drop by almost 2 $\mu g/m^3$, followed by 1.6 $\mu g/m^3$ for sulphate. In particular, the abatement of acidifying species in foreign countries has an impact on the SIA levels in the Netherlands. Almost 75% of the modelled reduction in SIA ensues from acidification policy in these countries. This emphasises the importance of international collaboration in abating particulate matter. The reduction in SIA causes the aerosol to become 'blacker' and more linked to emissions from combustion processes.

In spite of the major reductions in SIA following cuts in foreign countries, a relatively constant 30-35% from modelled PM_{10} stems from Dutch emissions in both 1995 and 2010-CLE. Primary emissions of PM_{10} in the Netherlands are expected to be controlled with greater efficiency than abroad. Figure 10 illustrates the changes in the modelled composition of PM_{10} as an average over the Netherlands. The 18 $\mu g/m^3$ 'not modelled' bar represents the average of the difference between models and measurements explained in more detail in the full report. The decrease in SIA formed from emissions in foreign countries is clearly visible in this diagram.

The modelled yearly average PM_{10} contributions in $\mu g/m^3$ from the transport sector at an urban background point in Rotterdam are presented for 1995 and for 2010 in Figure 11. It has to be remembered that a considerable part of the transport contribution in Rotterdam, which is the world's largest port, consists of shipping.

PM_{10} emissions in the Netherlands in 1998 are estimated at 54 ktonnes per year, of which 40% is combustion-related. Emissions of $PM_{2.5}$ are estimated to amount to about 32 ktonnes, i.e. 60% of PM_{10} emissions. From 1998 to 2010, total emissions of PM_{10} will decrease by approximately 20% to a level of 42 ktonnes. The fraction of PM_{10} that is combustion-related and that seems to be more health-relevant will show an even larger decline of 40%. For carbonaceous combustion emissions made by the transport

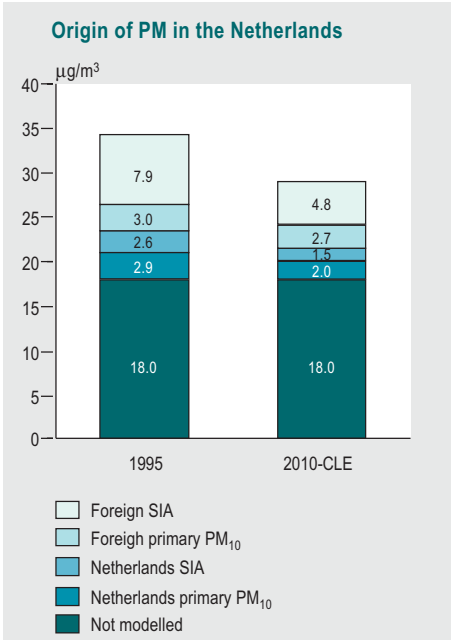

Figure 10. Changes in the composition of yearly average modelled PM_{10} between 1995 and 2010-CLE

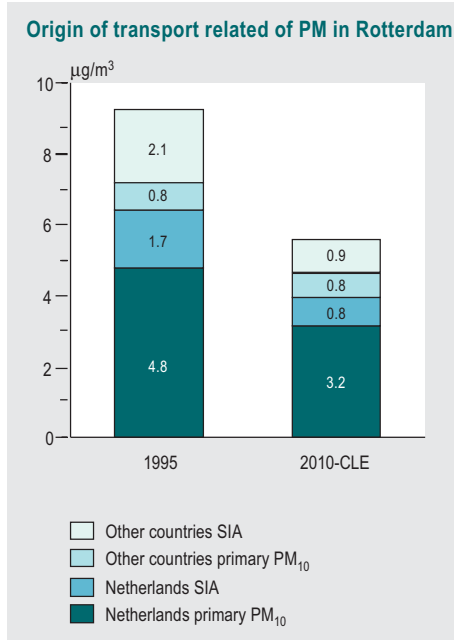

Figure 11. Contribution from the transport sector in Rotterdam (road transport and other mobile sources, mainly shipping) modelled as yearly average PM_{10} in an urban background in µg/m³

sector, which are mainly diesel-related, a downward trend is projected of about 45%. Total national emissions of $PM_{2.5}$ are anticipated to drop by about 30% from 1998 to 2010.

Under current policies, emissions of PM_{10} will decrease by about 20% from 1998 to 2010. The fraction of PM_{10} that is combustion-related and that seems to be health-relevant will show an even larger decline of 40%. National emissions of $PM_{2.5}$ will decrease by about 30%.

In 2010, dominant classical sources of PM in the energy sector, industry, waste treatment sector and road transport will be strictly controlled in the Netherlands. At present, Dutch directives NeR and BEES for industrial sources and combustion plants have been almost fully implemented. Dutch industries already comply with almost all EU reference documents on Best Available Techniques. This successful abatement of classical sources renders other less controlled sources more important. Less controlled sources are still found in a wide range of sources, i.e. in industry (smaller point sources with low emission flows, ventilation air of industrial buildings, storage and handling), transport (inland ships, sea-going ships, mobile machines, wear of tyres, brakes and road surface), agriculture (animal housing systems), consumers (wood stoves and fireplaces), construction (construction sites) and the commercial and institutional sector (storage

and handling of materials). It should be realised that the chemical and size characteristics of PM emissions of these less controlled sources are rather different, and some may be less health-relevant than others. Information on the health relevance of PM emissions from different sources is scarce, with the exception of combustion-related diesel emissions from traffic for which the general opinion is that these emissions are probably health-relevant. In this respect, the anticipated future trend in emissions from shipping (inland shipping and international maritime shipping in Dutch ports) is a cause of concern. International initiatives to limit these emissions have not been very effective up to now, so the contribution of shipping emissions to combustion-related transport emissions has increased over the last twenty years from about 10% in 1980 to 20% in 1998, and will increase further to about 40% in 2010. Emission trends for mobile machines are decreasing because the EU has agreed on the first effective steps (phase 1 and phase 2) to control these emissions. However, for these sources too it should be realised that currently agreed EU emission limits are much less stringent than agreed EURO4/5 emission limits for heavy-duty vehicles.

PM emissions from diesel engines used in maritime shipping, inland shipping and off-road mobile machines will be less controlled than similar PM emissions from comparable engines used in heavy-duty vehicles for road transport.

5 STANDARD SETTING AND POLICY OPTIONS

In this last part of the executive summary the conclusions for standard setting for PM and a number of the available policy options and their consequences will be presented. More details can be found in Chapters 5 and 7 of the full report.

5.1 Considerations on the current daily standard for PM

The first conclusion for standard setting refers to the EU standards for PM_{10} for the year 2005. The yearly average value of 40 µg/m^3 and the daily average of 50 µg/m^3 with 35 permitted exceedances per year are not equivalent in the Netherlands. The original EU position paper was based on the equivalence of both standards. Figure 12 shows that a yearly average value of 40 µg/m^3 for PM_{10} is equivalent to a daily value of 50 µg/m^3 with 80 allowed exceedances per year in average Dutch atmospheric conditions.

The EU yearly average and daily standards are not equivalent in the Netherlands.

The horizontal and vertical red lines in Figure 12 show the EU daily standards of 2005 (35 days allowed in which 50 µg/m^3 is exceeded) and yearly limit value (40 µg/m^3). The red dotted lines: indicative EU standards for 2010.

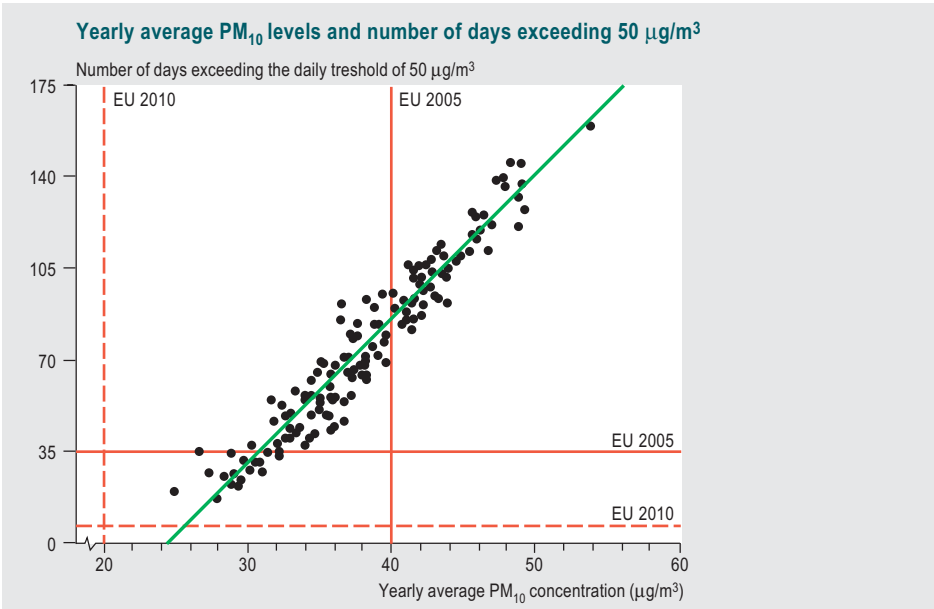

Figure 12. The yearly average PM_{10} concentration level and the number of days the EU standard of 50 µg/m^3 is exceeded for all different PM_{10} monitoring stations between 1992 and 2001.

One of the reasons for this mismatch in the two EU standards was that the daily average standard had been derived from UK measurements made using an instrument (TEOM) that had not been corrected for losses of semi-volatile material. Consequently, the measured values derived from this study were too low, resulting in the low value of the standard. If this loss had been taken into account, the daily average value measured by TEOM would have been considerably higher. Its value would have been at least a factor of 1.3, and possibly a factor of 1.9, larger if the average Dutch situation had been taken into account. The resulting daily average values would then have been 65 or 95 $\mu g/m^3$ instead of the presently used figure of 50 $\mu g/m^3$.

When reasons of risk communication to the public are considered, a standard of 50 $\mu g/m^3$ with either 35 or 80 permitted exceedances will be difficult to communicate. Up to now, a level of 50 $\mu g/m^3$ is exceeded somewhere in the Netherlands during six months of the year. So the attention value of a specific PM warning issued every other day will soon decrease. A value greater than 50 $\mu g/m^3$, but with a proportionally decreasing number of accepted exceedances (at the same time being equivalent to the yearly average value of 40 $\mu g/m^3$), could be better suited for purposes of alerting the public.

With 80 allowed exceedances per year a daily average value of 50 $\mu g/m^3$ would be equivalent to a yearly average PM_{10} standard of 40 $\mu g/m^3$.

As has been indicated above, risk communication to the general public with either 35 or 80 allowed excursions per year is quite complicated. If this communication were limited to a maximum of 7 times a year, it would become a 98-percentile and would be more feasible.
The numerical value of a 98-percentile for the EU daily standard (equivalent to a yearly average value of 40 $\mu g/m^3$) would be 100 $\mu g/m^3$ for the Dutch situation (with a GSD of 1.7). This has been elaborated in Chapter 5 of the full document. By the definition of a 98-percentile, a daily limit of this value may not be exceeded on more than 7 days a year.

A daily average of 100 $\mu g/m^3$ PM_{10} with 7 exceedances per year is also equivalent to a yearly average level of 40 $\mu g/m^3$ in the Netherlands. For reasons of practicability, a value of 100 $\mu g/m^3$ with 7 exceedances is preferred to a value of 50 $\mu g/m^3$ with 80 exceedances.

In the Netherlands a large fraction (approximately 85%) of the *temporal, daily variations* in PM_{10} concentrations is caused by weather variations, whereas only 15% of the *spatial variation* in annual average concentrations is influenced by meteorology. This indicates that local, regional or national authorities have limited possibilities for influencing the daily average PM values. On the other hand, reducing annual average concentrations also results in a decrease in the daily concentrations, although the pattern of daily variations remains unaltered. Furthermore, it must be remembered that the current measurement uncertainty in the automatic PM measurements becomes more manifest in daily concentrations than in average annual concentrations. The shorter the averaging

time, the higher the chances of some of the measurements indicating exceedances. As a consequence of these measuring uncertainties, in a 'real world' situation with PM concentrations below, but close to, the standard, a number of daily PM measurements will indicate otherwise.
A third consideration concerning a yearly average and a daily average standard is modelling. At present, deterministic dispersion models that can reliably model PM concentrations on a daily basis are not operational and have not been validated in the Netherlands. Model tools that have demonstrated they can do an adequate job with a reasonable level of precision for the modelling of yearly average concentrations are currently operational. In the past, modelling tools have been very helpful and, as can be seen in the full report, they can be called pivotal in estimating, predicting and evaluating the effectiveness of various abatement strategies.

For local (or regional) authorities the availability of a daily standard seems to be a helpful instrument for risk communication and advice to the public. As mentioned, episode levels as such are not amenable to control by taking short-term emission reduction measures.

On the other hand, a 24-hour-average standard could well be used for risk communication to the public. A more or less similar position regarding risk communication and "alert" levels has previously been taken for other components in EU daughter directives. For instance, an information system is currently in use in the Netherlands for ozone, PM_{10}, SO_2 and NO_2 and for pollen.
However, the 24-hour standard of 50 $\mu g/m^3$ for PM_{10} is so low that this daily average is exceeded somewhere in the Netherlands during six months of the year. A complicating factor with PM is that it is less clear how individual members of the public can actually protect themselves from the PM-associated health risks. Contrary to pollutants like ozone, SO_2 and pollen, indoor levels of PM_{10} and $PM_{2.5}$ are only slightly lower than ambient levels. Nevertheless, a reduction of physical activity can generally be recommended to reduce the amount of air inhaled and hence, the amount of pollution inhaled on high pollution days. Also, avoidance of participation in motorised traffic, and avoidance of being on major roads will help to reduce exposure to PM.

A value of 100 $\mu g/m^3$ with 7 exceedances per year is equivalent to a yearly average value of 40 $\mu g/m^3$ in the Netherlands. If this value were chosen as the EU daily standard, 'alerting' the public would become more feasible. A complication with a risk communication scheme of this kind is that at present the prediction of exceedances above a certain level of PM_{10} on a daily basis is not well established. In the early 1990s, there were two types of risk communication or 'smog alert' in the Netherlands: 'summer smog' and 'winter smog'. The first one was triggered by a one-hour maximum ozone level of more than 240 $\mu g/m^3$ and the second by the sum of the daily PM_{10} and SO_2 concentrations exceeding a value of 450 $\mu g/m^3$. These alert levels for the general public were chosen because they were deemed to correspond to the threshold levels for health effects. This means that below these levels no health effects were presumed to occur and risk communication below these levels was therefore not considered necessary.

However, recent epidemiological research has demonstrated that there no longer seems to be a threshold for PM-related health effects, as health effects appear to exist at any level of PM. So, risk communication for non-threshold toxicants, indicating that any level of exposure carries a certain risk, would probably be more appropriate than one which specifically comes into operation when a certain PM_{10} threshold of 50 µg/m^3 or 100 µg/m^3 is exceeded.

Although the EU has proposed two standards for PM, there are several arguments that only one standard would suffice – annual mean concentrations being the best choice. However, for reasons of communication to the public and preventing exceedances above a certain threshold, daily standards may be appropriate.

5.2 Compliance with EU standards

Compliance with the yearly average EU standard of 40 µg/m^3 seems feasible in 2005 if the currently proposed and envisaged policy measures for PM abatement are achieved. The full report still mentions a number of 'hot spots' in regard to this yearly average. One is that for one (largely industrial) grid of 5 x 5 km^2, a yearly average value in excess of 40 µg/m^3 is assessed for 2005 based on differential mapping and concentration modelling. However, this modelled value is not supported by current PM measurements taken by the competent regional authorities at that location, as current yearly average PM levels are below 40 µg/m^3. The high values in this grid generated by this modelling assessment may be due in part to some technical modelling problems with the spatial attribution of a number of sources of coarse PM_{10} by the source category: storage and transhipment of material. For this category the modelling is currently under scrutiny. The second type of 'hot spot' is urban street canyons. Though the average value for the urban background was 33 µg/m^3 in 2001, values above 40 µg/m^3 can not be excluded, based on the measurements by TNO. This indicates that for a number of urban situations the combination of traffic and the local dispersion parameters, which are probably heavily influenced by the current urban building pattern and spatial layout, may result in unfavourable situations. This is a point of concern that demands extra attention in the near future.

For the 50 µg/m^3 daily average for PM_{10} an assessment has been made for 2010 which takes into account the influence of future abatement policies up to that year. Note that this assessment has been made with a given value (1.3) of the correction factor used to compensate for losses of semi-volatiles. If this value changes in the future (either for specific locations or seasons) the results of the assessment presented here will also change. Based on this assessment it can be concluded that even in a favourable situation with fewer emissions, the standard of 50 µg/m^3 would still be exceeded in the Netherlands on average approximately 36 to 40 times a year in 2010. It also indicates that now and in the near future it will probably be impossible to comply with the standard of 35

permitted exceedances of the daily average in 2005 or with the 7 exceedances in 2010.

Compliance with the yearly average value of 40 µg/m^3 seems feasible for PM$_{10}$ in the Netherlands, though at present local exceedances cannot be ruled out. However, compliance with the daily average value of 50 µg/m^3 with 35 permitted exceedances does not seem feasible throughout the country in 2005.

The indicative EU values for 2010 are 20 µg/m^3 as a yearly average value and a daily average of 50 µg/m^3 with 7 permitted excursions per year. Modelling the expected yearly average values of PM$_{10}$ in 2010, based on all the current and envisaged abatement measures, results in a modelled concentration of 11 µg/m^3. This is 5 µg/m^3 lower than the modelled concentration for 1995. In 2010 the transport and shipping sector will contribute 3.6 µg/m^3 of which 1.3 µg/m^3 is a result of Dutch emissions. Compared with 1995, the largest reduction modelled is due to abatement in the transport sector (2.3 µg/m^3) and the energy sector in other countries. As observed earlier, the larger decrease in total PM$_{10}$ between 1995 and 2010 is the result of an abatement of acidifying species (3.5 µg/m^3), whereas 1.2 µg/m^3 is of primary origin.

If the non-modelled sources are kept constant in the 2010 estimate, similar to 1995 as they are for a large part more of natural origin, it would indicate that the expected yearly average concentration in 2010, conservatively estimated, will be some 5 µg/m^3 lower than in 1995. This indicates that at a regional level a yearly average value of 30 µg/m^3 can be expected for 2010. This is considerably higher than the indicative standard of 20 µg/m^3. The urban situation will be even more unfavourable as PM$_{10}$ concentrations are generally somewhat higher here (approximately 2-3 µg/m^3).

OPS models the highest yearly average PM$_{10}$ concentration of 36 µg/m^3 for 2010 under the CLE scenario in an urban background grid in Rotterdam (the aforementioned industrial grid remains the highest even in 2010, of course. In more precise terms this background grid in Rotterdam is the penultimate.) This value of 36 µg/m^3 presents the maximum reduction that can be achieved under current legislation, a condition being that this yearly average has to be complied with on every grid in the Netherlands. It should be remembered, of course, that higher PM$_{10}$ concentrations may well be possible in street canyons in this urban background.

The ultimate technical reduction potential (20 ktonnes emission reduction in addition to current legislation in 2010CLE) would lead to a further concentration reduction of 1.1 µg/m^3 averaged across the Netherlands. Locally, maximum additional reductions of 2.5 µg/m^3 are modelled along the Rhine shipping route and the Rotterdam harbour area. A local reduction of 7 µg/m^3 is modelled near an industrial facility in the west of the country. The package is not expected to produce different conclusions with respect to the ability to meet the 20 µg/m^3 2010 indicative annual standard.

A separate analysis by linear extrapolation of the trend of the meteorologically corrected regional PM$_{10}$ levels in the Netherlands from Figure 8 indicates that if all the abatement

measures continue to be taken in the future with the same vigour as they have been taken during the last ten years, a regional average value of approximately 21 μg/m^3 could be expected to be reached in 2010. This value can only be reached if the current slope of the downward trend continues in the future. So this linear extrapolation is probably an overoptimistic projection, as all the relatively easier and promising emission reductions have already been implemented. Even in this scenario, though, future regional PM concentrations based on dispersion models will still be considerably higher than the indicative standard of 20 μg/m^3. Yearly average urban background and street concentrations will, of course, be higher still than those at the regional level.

Compliance with the indicative yearly average value of 20 μg/m^3 in 2010 is not feasible for PM$_{10}$ in the Netherlands with current and foreseen abatement technologies and policies; compliance with the daily average value of 50 μg/m^3 with 7 permitted excursions is not feasible either in 2010.

5.3 Control strategies and options for additional reductions

The current level of health risks associated with PM in the Netherlands is considerable and surpasses the maximal tolerable risk levels as defined in the Dutch policy document 'Premises for Risk Management'. However vague and elusive the presented risk estimates sometimes seem to be, it should by any means be very clear that a maximum tolerable risk of one excess mortality per year per million inhabitants (which is the yardstick in the prevailing policy document 'Premises for Risk Management') is considerably exceeded by the PM-associated health effects. The Dutch policy document was, however, completed before the current PM enigma emerged, and its practical application to PM is rather complicated because of its far-reaching consequences. An option of risk reduction by halving the PM$_{10}$ standards in 2010 seems neither practically feasible, nor very sensible when current knowledge of PM$_{10}$ as a complex mixture with fractions that are to a greater or lesser extent health-relevant is taken into account.

In spite of the fact that falling emission trends indicate that the 2005 EU standard of 40 μg/m^3 seems feasible, large uncertainties still exist and the scientific debate continues. Because there appears to be no threshold for PM-associated heath effects, even a PM$_{10}$ level of 20 μg/m^3 will still have a large impact on health in the general population. The extent and the seriousness of the PM enigma warrant further and permanent action in the foreseeable future by the Government and other parties concerned.

On the basis of the precautionary principle, supplementary policy action could be directed at further control of sources that are possibly more health-relevant e.g. primary combustion-related emissions and more in particular transport- related diesel soot.

Technical possibilities for further emission reduction in the Netherlands have been explored (see Table 5). Two emission abatement packages have been developed.

1. The ultimate Maximum technically Feasible Reduction abatement package ("MFR_{ult}") shows emissions in 2010 assuming full implementation of the measures and not looking at cost. The "MFR_{ult}" abatement package shows that the available technology is no constraint for further reduction in emissions of PM_{10}. Technical options studied have the ultimate potential to reduce emissions by about 60% from 42 ktonnes to 18 ktonnes per year. The cost, however, is about 6000 million Euro per year in the Netherlands.
2. The second abatement package "$2010_{quart\ red}$" incorporates only those measures with marginal costs of up to 55 Euro per kg PM_{10} reduction. In this abatement package, emissions of PM_{10} are reduced by about a quarter from 42 ktonnes to 32 ktonnes. These reductions are made in industry (refineries, food industry, building materials and chemical industry), shipping and storage and handling companies. Measures for the basic metals industry also fall into this cost category, but have already been included under current policies. Measures in other industrial sectors, not within this group of five high emitting sectors, are relatively more expensive. Annual national costs are estimated to be 210 million Euro per year, with relatively low costs for the inland shipping sector (8 million Euro per year) and relatively high costs for industry (174 million Euro per year) and the commercial and institutional sector (25 million Euro per year).

Table 5. Averaged results of air quality calculations using the 2010 scenario variants. In order to facilitate the comparison with calculations from past years, data from 1980 and 1995 are listed as well. The non-modelled fraction of 18 mg/m3 is not shown in this table

Year	Modelled average anthropogenic PM_{10} concentration ($\mu g/m^3$)	Modelled average concentration of primary PM_{10} ($\mu g/m^3$)	Dutch primary PM_{10} emissions in ktonnes	Additional costs per year compared to 2010 CLE (M€)
1980	29.7	11.4	116	
1995	16.5	6.0	61	
2005-CLE	12.9	5.2	48	
2010-CLE	11.1	4.8	42	
$2010_{quart\ red}$	10.8	4.5	32	210[a]
$2010\text{-}MFR_{ult}$	10.0	3.6	18	6000[a]

CLE = Current Legislation decided upon before 1-1-2000 according to the Global Competition C scenario
$2010_{quart\ red}$ = CLE with an extra reduction of approx. a quarter, marginal costs of up to 55 euro per kg PM_{10} reduction.
MFR_{ult} = ultimate Maximum technical Feasible Reductions

[a] Excluding costs for technical measures on sea-going ships

The ultimate technical potential for PM_{10} reduction, on top of currently agreed measures, could be 60% of current PM emissions in the Netherlands. The cost of achieving this reduction is about 6000 million Euro per year. A reduction of 25% in Dutch PM_{10} emissions could be achieved at a cost of 210 million Euro per year.

There still remain options for further reductions when the marginal costs of various abatement options for primary PM emissions are considered more closely, although the cost of measures is generally high. Information on cost is presented in Chapter 6 of the full report.

Other measures for achieving further reductions in *primary* PM_{10} emissions in the Netherlands are:

- reduction in the S-content of residual oil used by sea-going ships;
- application of technical measures (optimised engines, particulate traps) that will reduce uncontrolled emissions from diesel engines on inland and sea-going ships;
- implementation of additional technical measures in the already controlled classical high-emitting industrial sectors, and in companies specialising in materials handling.

The first abatement option demands that Annex VI of the International Convention for the Prevention of Pollution from Ships (MARPOL) come into force (reduction of S–content from 3% to 1.5%). In a following step international agreements need to be made on a further reduction (from 1.5% to 0.5%). The second option requires stringent Stage-2 PM emission limits for inland ships (e.g. Euro4/5 limits for Heavy-Duty Vehicles), and control measures for sea-going ships should also be agreed. The cost of these measures is about 4 Euro per kg PM_{10}. With respect to the third option for industrial sources, three conclusions can be drawn. In the first place, the tightening of generic national PM emission standards will not be the most cost-effective option for achieving reductions in industry. This strategy will affect sectors/processes with high marginal costs while there are still opportunities for further reduction in other sectors/processes at less expense. Other policy instruments are needed to achieve this cost-effective reduction. Secondly, a cost-effective reduction policy for PM_{10} will result in a similar or probably even larger reduction in $PM_{2.5}$ emissions. Finally, marginal costs of measures in industry will be more expensive than the upper cost limit of 2.3 Euro per kg currently in force in the Netherlands through the instrument of the Dutch NeR directive.

As already mentioned, supplementary source-oriented actions could in the first place be directed at the further control of sources that may be more health-relevant. In this respect, the abatement of uncontrolled shipping emissions has been identified as the most cost-effective control option. The abatement of other combustion-related sources such as industrial combustion, wood burning in fireplaces and off-road machinery are also possible, but prove to be less cost-effective.
An emission reduction by a quarter (abatement package "$2010_{quart\ red}$") and marginal costs of 55 Euro/kg of PM_{10} reduction could be achieved at a cost of 210 million Euro per year, resulting on average in a 0.3 $\mu g/m^3$ lower PM_{10} concentration. The ultimate technical reduction potential ("MFR_{ult}": 20 ktonnes emission reduction in addition to

current legislation in 2010CLE) would result in a concentration reduction of 1.1 $\mu g/m^3$ averaged across the Netherlands compared with 2010CLE.
From these values it can be concluded that, averaged on a national level, these PM_{10} reductions seem rather small. Locally, however, higher reductions of 1 to 5.5 $\mu g/m^3$ in PM_{10} levels are modelled in "$2010_{quart\ red}$". The maximum reductions will be achieved in Rotterdam, which is densely populated. It is interesting to note that the measures directed at *transport* in the "$2010_{quart\ red}$" scenario are devoted to the shipping sector only. When the focus is placed on more health-relevant fractions of PM, e.g. traffic-related diesel soot, modelled reductions are relatively higher even. The presented abatement packages ("$2010_{quart\ red}$" and "MFR_{ult}") correspond to a drop of 20% and 50% respectively in average traffic-related diesel soot concentration levels of Dutch origin. These effects would be further intensified if similar reduction technologies were also applied in foreign countries.

The most cost-efficient control options are the technical adaptation or replacement of diesel engines on inland and sea-going ships. Reductions can be achieved by retrofitting particulate traps on old ships and/or by setting stringent EU emission limits for new ship engines, which in the most ambitious case could be harmonised with Euro 4/5 emission limits for heavy-duty machines.

Apart from the cost aspect, what would be the consequences of choosing PM_{10} or $PM_{2.5}$ as a basis for reduction policies in industry?
PM_{10} includes all $PM_{2.5}$, while $PM_{2.5}$ excludes all larger particles within PM_{10} (2.5 μm < diam. < 10 μm). Reducing PM_{10} also results in a $PM_{2.5}$ reduction, depending on the $PM_{2.5}$ fraction. Reducing $PM_{2.5}$ will in most cases also lead to a reduction in larger particles in industry because the $PM_{2.5}$ fraction is in almost every case less than unity.
Coincidentally, in a cost-effective PM_{10} reduction strategy the most attractive PM_{10} sources for reductions also contain high fractions of $PM_{2.5}$. So, PM_{10} reductions result in relatively high reductions for $PM_{2.5}$ as well. The full report illustrates the fact that the first 50% of PM_{10} reduction results in a 60% $PM_{2.5}$ reduction. It is a matter of coincidence that sources with cost-effective PM_{10} reductions also contain relatively high fractions of $PM_{2.5}$.
Vice versa, $PM_{2.5}$ reduction always results in PM_{10} reduction as well. However, a cost-effective strategy for $PM_{2.5}$ focuses on the sources with high $PM_{2.5}$ fractions, resulting in relatively low additional PM_{10} reductions. In the full report it is shown that the first 50% of $PM_{2.5}$ reduction produces only a 30% reduction in PM_{10}.

The conclusion is that an industrial reduction policy taking PM_{10} as a general approach also effectively reduces $PM_{2.5}$, but is more expensive than a similar cost-effective strategy focusing solely on $PM_{2.5}$. A cost-effective reduction policy formulated in terms of $PM_{2.5}$ is cheaper, but results in lower reductions of PM_{10}.

Figure 10 also limits the influence of the ultimate Maximum technically Feasible Reduction abatement package ("MFR_{ult}") from Table 5 to its mass proportion. With "MFR_{ult}", the absolute yearly average Dutch contribution could be reduced from the

calculated 2010CLE contribution of 2.0 to 0.9 μg/m^3. Though this reduction seems small in absolute numbers, its impact could be much larger as it may be a more health-relevant fraction.
However, this still does not lead to a prospect of compliance with 20 μg/m^3 in 2010. Reducing the non-modelled part of 18 μg/m^3 becomes essential to achieve such low levels. This non-modelled part is currently known only to a certain extent. The numerical value of 18 μg/m^3 is also influenced by the 'true' value of the correction factor of 1.3 that is used, correcting for the losses of semi-volatiles. Of this 18 μg/m^3, approximately 4-7 μg/m^3 is sea salt, 2 μg/m^3 is the natural crustal contribution and 1-2 μg/m^3 are non-modelled contributions of road dust, while 1 μg/m^3 is the northern hemisphere background. There is an unknown (but probably in the Netherlands not very large) contribution of Secondary Organic Aerosols (SOA).

More insight into the chemical composition (specific tracers) and contribution of different sources to the currently 'non-modelled' part of PM_{10} is necessary to find out how much of the current PM levels may eventually be influenced by abatement measures.

Nearly all abatement options presented above are directed at anthropogenic sources of PM. This anthropogenic fraction of course is a substantial part of our PM levels, but there is also an important more natural contribution to PM levels.
In the Netherlands a considerable part of PM_{10} is of natural origin. On a yearly average basis we have quite a large contribution of sea salt: 4 - 7 μg/m^3. Part of PM_{10} is of crustal origin. Sometimes this is wind-blown dust that is re-suspended due to meteorological conditions, but part of the crustal contribution in the Netherlands is anthropogenic, as it is re-suspended by traffic-induced turbulence. No adequate information is available about the composition and contribution of biogenous material. Current information shows that the total natural contribution to present levels of PM_{10} in the Netherlands is considerable and will probably even rise proportionally.

It would be wrong to conclude that this natural contribution does not have any associated health effects. In a previous Dutch epidemiological time-series study it was shown that pollen is associated with mortality independent of the occurrence of ambient PM_{10}. It should be realised that due to continuing measures to abate PM emissions from anthropogenic sources, the relative contribution in mass terms of natural sources will become more important in the future. At the moment, however, available information about the health effects does not suggest that known biogenous sources and agents (e.g. viruses, endotoxins, pollen, etc.) are largely responsible for the statistically observed health effects in the population in epidemiological studies.

Part of the PM_{10} levels in the Netherlands cannot be influenced by policy measures, as natural sources are responsible for their ambient concentrations. Because future abatement measures will further reduce the anthropogenic fraction, the natural fraction will increase proportionally.

The current PM enigma and accompanying scientific debate indicate there are still such fundamental uncertainties that PM and its associated health problems will continue to be on the agenda of national and European policy makers in 2010. In spite of these uncertainties the extent and seriousness of the problem mean that an active PM policy will remain necessary. Further PM abatement measures, on top of what has been achieved already, will be very costly in the Netherlands. Nevertheless, further source-oriented measures can be based on the precautionary principle. These actions could focus on more cost-effective reductions of the total PM_{10} aerosol mass, or could be directed at those fractions that are expected to be more health-relevant, and this last option is preferred. So, a cost-effective way of achieving concrete risk reduction for specific source categories seems to be indicated.

Annex A.

Annually averaged primary and secondary inorganic concentrations of $PM_{2.5}$ averaged over the Netherlands by anthropogenic source. Calculated for the year 1995, based on emissions for the Netherlands and the CEPMEIP inventory for European countries.

Dutch sources	Primary PM_{10} ($\mu g/m^3$)	NH_x ($\mu g/m^3$)	NO_y ($\mu g/m^3$)	SO_x ($\mu g/m^3$)	Summed concentration ($\mu g/m^3$)
Industry	0.2	0.0	0.1	0.0	0.4
Energy	0.0	0.0	0.1	0.1	0.2
Transport [1)]	1.4	0.0	0.8	0.1	2.3
Agriculture	0.1	0.9	0.0	0.0	1.1
Others	0.4	0.1	0.1	0.0	0.6
Sum	**2.2**	**1.0**	**1.1**	**0.2**	**4.6**
Other countries					
Industry	0.6	0.0	0.1	0.1	0.7
Energy	0.2	0.0	0.5	1.7	2.5
Transport 1)	0.8	0.0	1.7	0.3	2.8
Agriculture	0.1	1.2	0.0	0.0	1.3
Others	0.6	0.0	0.4	1.0	1.9
Sum	**2.3**	**1.2**	**2.6**	**3.1**	**9.9**
All sources					
Sum	**4.5**	**2.2**	**3.7**	**3.3**	**13.8**

[1)] Including international shipping