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Estimates of the distribution of different fractions of PM_{10} in the ambient atmosphere in the Netherlands

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ABSTRACT

This report presents some methods to synthesize the present knowledge into a comprehensive overview. Several assumptions sometimes of a more or less speculative nature had to be introduced to arrive at a complete picture. These assumptions have been mentioned explicitely as “seven rules of thumb”. Using these rules, two statements i.e. 1) the ratio between Dutch and foreign contributions to PM_{10} levels is 20% and 80% and 2) a local contribution of 15% of primary PM_{10} by Dutch sources, are shown to contradict. As a way of hypothesis generation a contribution of 40% to 45% by local Dutch sources to PM_{10} levels is presented. This difference can be accounted for by assuming a larger share of local emissions of coarse particles > 2.5 μm to PM_{10} concentrations than was estimated previously. PM_{10} concentrations for five areas of the Netherlands are estimated using the seven rules, simple model calculations and assumptions on concentration gradients caused by differences in source characteristics and size distributions. It appears that the original “blanket” of homogeneous PM_{10} concentrations covering the Netherlands, indicating rather small concentration gradients, turns out to be more like a “patchwork quilt”. The estimated concentration ranges are comparable with the ranges of annual PM_{10} concentrations of 35 - 45 $\mu g/m^3$, measured by the National Air Quality Monitoring Network.

SAMENVATTING

Dit rapport geeft een schatting van de concentraties van PM_{10} in de Nederlandse buitenlucht en de verdeling van de verschillende fracties die bijdragen aan de totale massa van PM_{10}. Deze schatting is tot stand gekomen met hypothesen voortkomend uit de bestaande aerosol kennis.

PM_{10} kan worden onderverdeeld in vijf categoriën. Drie daarvan hebben betrekking op de deeltjesgrootteverdeling van het aerosol. Deeltjes gemonsterd met een 50% afscheidingskarakteristiek van 10, 2,5 en 0,1 µm worden respectievelijk PM_{10}, $PM_{2,5}$ en $PM_{0,1}$ genoemd. De andere twee categoriën betreft een chemisch onderscheid: het secundaire aerosol (som van sulfaat, nitraat en ammonium) en het koolstofhoudend aerosol (som van elementair en organisch koolstof uit verbrandingsbronnen).

Het kwantitatieve inzicht in het ontstaan en het verdwijnen van atmosferisch stof is nog beperkt. Vanwege de mogelijk ernstige gezondheidsrisico's die geassocieerd zijn met deeltjesvormige luchtverontreiniging kan de voorbereiding van beleid echter niet wachten tot alle aspecten volledig begrepen zijn. Dit rapport tracht daarom de bestaande soms fragmentarische kennis, op een methodische en transparante wijze samen te voegen, om zo een eerste discussie daarover te kunnen aangaan met beleidsmakers en om hypothesen te genereren die met later onderzoek kunnen worden getoetst. Daarbij was het nodig om een aantal vooronderstellingen van meer of minder speculatieve aard te maken om het beeld compleet te krijgen. Deze aannamen worden bediscussieerd en de gevoeligheid daarvan op de gepresenteerde uitkomsten wordt beschreven.

Op grond van zeven vuistregels, die in hoofdstuk 3 worden beschreven, blijkt dat eerdere aannamen: namelijk dat 20% van het PM_{10} uit Nederland en 80% uit het buitenland komt en tevens dat 15% van het primaire PM_{10} afkomstig is uit Nederlandse bronnen onderling strijdig zijn. Bij wijze van hypothese vorming is een nieuwe schatting geformuleerd die wel aan de zeven vuistregels voldoet. Dit houdt in dat 40 to 45% van het PM_{10} uit Nederland komt. Dit verschil zit vooral in de aanname van een grotere bijdrage van lokale bronnen van grof (> 2,5 µm) PM_{10}.

Met behulp van de vuistregels, eenvoudige modelberekeningen en aannamen met betrekking tot concentratiegradiënten veroorzaakt door verschillen in bronkarakteristieken en deeltjesgrootteverdelingen zijn voor vijf gebieden in Nederland de PM_{10} concentraties geschat. De door de kleine concentratieverschillen over Nederland bijna als een "homogeen ogende deken" van PM_{10} blijkt bij nadere beschouwing meer weg te hebben van een "lappendeken" bestaande uit aerosol van verschillende samenstelling. De geschatte bandbreedte van

concentraties komt echter aardig overeen met de bandbreedte van de jaargemiddelde PM_{10} concentraties over Nederland van 35-45 µg/m^3, zoals deze gemeten zijn in het Landelijk Meetnet Luchtverontreiniging van het RIVM.

Een beter inzicht in de deeltjesgrootteverdeling en de chemische samenstelling van het totale deeltjesvormige aerosol in Nederland is nodig om de geformuleerde hypothesen te kunnen toetsen. Een dergelijk inzicht kan verworven worden met behulp van bronherkennings- en andere meettechnieken. Dergelijke gegevens zullen leiden tot een beter inzicht en vooral een betere kwantificering van de processen en de bronnen van deeltjesvormige luchtverontreiniging. Slechts met een betere onderbouwing kan nationaal en internationaal een adequaat en kosten-effectief bestrijdingsbeleid gevoerd worden.

SUMMARY

This report gives estimates of the distribution of different fractions of PM_{10} in the Netherlands, using various hypotheses, based on existing knowledge of PM.

PM_{10} may be devided into five categories. Three of them refer to the size distribution of the aerosol: 50% cut-off diameters of 10, 2.5 and 0.1 μm resulting in PM_{10}, $PM_{2.5}$ and $PM_{0.1}$ respectively, the two others, to the chemical speciation of the aerosol: secondary aerosol (the sum of concentrations of sulphate, nitrate and ammonia), and carbonaceous (the sum of elemental and organic carbon from combustion sources).

The insight in the sources and sinks of ambient PM is still very limited. Because of the possible serious health risks associated with PM, decisions on how to deal with this problem cannot be postponed untill all the aspects are fully understood. This exercise attempts to synthesize fragments of existing knowledge into a comprehensive overview, in order to provide a first basis for discussions with policy makers and to guide future research efforts by providing testable hypotheses. It was unavoidable to introduce several assumptions of a more or less speculative nature to arrive at a complete picture. These assumptions have been mentioned explicitely as seven rules of thumb. These will be used later on for calculations and their sensitivity is discussed in this report.

Using seven rules, which are described in chapter 3, the statements of 1) a 20% Dutch and 80% foreign contribution to PM_{10} levels and 2) a local contribution of 15% of primary PM_{10} by Dutch sources are shown to contradict. By way of hypothesis generation a new postulate for the contribution by local Dutch sources of 40 to 45% is adopted. This difference is caused mainly by assuming larger local emissions of coarse particles > 2.5 μm than was estimated previously.

The PM_{10} concentrations for five areas of the Netherlands are estimated using the seven rules, simple model calculations and assumptions on concentration gradients caused by differences in source characteristics and size distributions. The picture of a "blanket" of homogeneous PM_{10} concentrations covering the Netherlands, based on rather small concentration gradients turns out to be more like a "patchwork quilt". The estimated concentration ranges are comparable with the ranges of annual PM_{10} concentrations of 35 - 45 $\mu g/m^3$, measured by the National Air Quality Monitoring Network.

A better insight into the size distribution and chemical speciation of PM is needed. This may be found by source apportionment and other techniques. Hopefully such data will lead to a better knowlegde and quantification of sources, sinks and emissions. In the future this data should decide the debate to which the hypothesis in this report contribute and provide a better basis for national and international PM abatement policies.

1. INTRODUCTION

In 1995 a research project was started by RIVM to advise the Dutch Ministry of Housing, Spatial Planning and Environment on future policy concerning fine Particulate Matter (PM). Part of this project consists of a phenomenological analysis of PM_{10} concentrations as measured in the Netherlands at several locations of the Dutch National Air Quality Monitoring Network aimed at:

- facilitating the comparison of measurements and model calculations and
- providing emission estimates of sources of which the emission is uncertain or cannot easily be incorporated in models, such as the contribution of sea-salt aerosol or resuspended crustal material.

An analysis of the daily variations of PM_{10} concentrations is recently described by Van der Wal and Janssen (1996).
Another part of the project is modelling primary and secondary PM in the Netherlands using air pollution dispersion models based on emission scenarios, presented elsewhere (RIVM,1997).

The insight in the sources and sinks of ambient PM is still very limited. Because of the serious health risks associated with PM, decisions on how to deal with this problem cannot be postponed untill all the aspects are fully understood. This exercise attempts to synthesize fragments of existing knowledge into a comprehensive overview, in order to provide a first basis for discussions with policy makers and to guide future research efforts by providing hypotheses which can be tested. Various assumptions, sometimes of a more or less speculative kind, were needed to make a complete picture. These assumptions have been called explicitely "rules of thumb". These rules of thumb are used later for calculations and the sensitivity of their results is discussed in this report.

This report provides a description of the regional composition of PM_{10} in the Netherlands as a function of different emission source categories. The composition of PM_{10} is described by the so-called "pentagon parameters", the five different components of PM_{10} distinguished in this project. These different PM categories will be defined in Chapter 2. Several sources and types of information are used to produce a complete picture of the PM_{10} composition i.e. some rules of thumb -hypotheses- were developed to estimate the national and foreign contributions to PM_{10} concentrations in the Netherlands. Particle size distributions were based on measurements and calculations with models. A set of assumptions of the PM_{10} pentagon distribution in the ambient atmosphere in the Netherlands can also be considered a means of hypothesis generation. Further research can be directed to validate these hypotheses, eventually leading to a better understanding of the atmospheric behaviour of the constituents of PM_{10}.

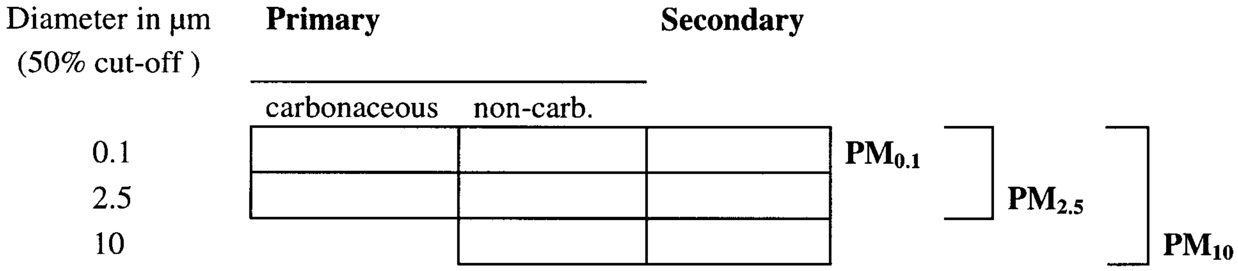
Chapter 3 presents the "seven rules of thumb" (assumptions) to determine the contribution of national and foreign emission sources to the five categories of PM_{10} which were distinguished in this project. The sensitivity of the assumptions on the results is discussed. The results of the calculations are compared with statements previously reported by Annema et al. (1996) and the differences are discussed. In Chapter 4 the contribution of major emission sources of PM_{10} to ambient concentrations of PM_{10} in the Netherlands is estimated, while Chapter 5 describes the characteristics of some typical background and source areas in the Netherlands. Chapter 6 intergrates the results of the analysis of the preceeding chapters into a description of the PM_{10} composition of some characteristic areas in the Netherlands. Chapter 7 concludes this report with a discussion on some of the assumptions, presenting conclusions and recommendations for further research.

2. THE “PENTAGON”: FIVE PM_{10} CATEGORIES

The usual indicator of PM in the ambient air is PM_{10}. PM_{10} is particulate air pollution measured with a size-dependent aerosol analyser, which has its 50% aerodynamic cut-off diameter at 10 μm. There are several instruments available to measure PM_{10} (Van der Meulen, 1985 and EPA, 1996)

This means that mainly particles with an aerodynamic diameter of less than 10 μm, regardless of their composition, contribute to ambient concentrations PM_{10}. Because of the 50% cut-off a small fraction of particles with aerodynamic diameters greater than 10 μm contribute to PM_{10}. PM_{10} is thus a collection of particulates (a container) with very different physical sizes, chemical composition and possibly biological significance. For this reason five different categories of PM were defined at the beginning of the project in September 1995. PM_{10}, $PM_{2.5}$, $PM_{0.1}$, carbonaceous and secondary aerosol. It was believed that most probably one or more of the culprits causing the PM-related health effects would be contained in one of these categories. The later criteria document of the EPA (1996) showed that these choices still are valid, since there are indications supporting the patho-physiological hypotheses on any of these fractions.

The hypothesis of acid aerosol (as free H^+) was not deemed important for the PM circumstances in the Netherlands because the large amount of livestock breeding of pigs, cows and poultry emitting NH_3, which quickly neutralizes any acidic aerosols in the atmosphere. As a consequence almost no free H^+ aerosol is assumed to exist here.
In addition to the five categories mentioned above primary aerosol is also distinguished from secondary aerosol, so that we can make the following subdivision, which presents a partial overlap with different definitions. N.B. the contribution of the different size fractions is not proportional with the surface of the rectangles.



The **pentagon** contains the next five categories:

PM_{10} is the mass of the ambient aerosol with a 50 % cut-off diameter of 10 µm. Time series of several years of concentration measurements are available since this aerosol has been collected since 1993 by the National Air Quality Monitoring Network in the Netherlands by stationary sampling at locations in rural areas, city background or street stations on an hourly basis. After sampling through a special designed "PM_{10} head" the aerosol is heated to 50 degrees C and the resulting mass is monitored continuously and automatically by a FAG.

$PM_{2.5}$ is the ambient aerosol with a 50% cut-off diameter of 2.5 µm. There are only some occasional 24 h or 8 h filter measurements in the Netherlands of this fraction, though on an experimental scale automatic measuring of $PM_{2.5}$ has recently started at a few stations.

$PM_{0.1}$ is the ambient aerosol with a 50% cut-off diameter of 0.1 µm. There are not even any occasional measurements of this part of the pentagon. Compared to the other parts of the PM pentagon, this remains a "terra incognita". Due to a lack of data $PM_{0.1}$ is not treated in this report. $PM_{0.1}$ might be important for the policy makers because some of the patho-physiological mechanistic hypotheses concern the very small part of the PM spectrum. Instead of $PM_{0.1}$ one could also take the total number of particles, as this number is almost completely determined by this very small fraction.

Carbonaceous aerosol, defined as the sum of elemental and organic carbon (EC + OC) from combustion sources, was also a part of the PM_{10} which attracted special attention because of its possible role in the health effects of PM (i.e. "diesel soot"). Apart from some occasional measurements, general data for the average concentrations of the carbonaceous aerosol are lacking in the Netherlands.

Secondary aerosol comprises that part of PM_{10} that has been formed by 'gas to particle' conversions in the ambient atmosphere. In the Netherlands these are mainly the conversion products of SO_2, NO_x, and NH_3. The total amount of secondary aerosol is thus defined as the sum of the aerosol concentrations in µg/m^3 of SO_4^{2-}, NO_3^- and NH_4^+. This part of the PM was collected daily (24 h averages) for quite a number of years by the National Air Pollution Monitoring Network with low-volume samplers (LVS) on filters at a number of rural stations at different locations in the Netherlands.

An additional categorie used in this report is "**coarse aerosol**"which is defined as the difference between PM_{10} and $PM_{2.5}$, indicating an aerosol fraction between the two 50% cut-off diameters of 2.5 and 10 µm. This is a difficult fraction to measure experimentally, as the most reliable way to measure its concentrations is to use a dichotomous sampler (EPA, 1996). Calculating the coarse fraction by substracting independent but simultaneous measurements of PM_{10} and $PM_{2.5}$ generally leads to gros errors. (See appendix B)

3. ESTIMATES OF THE CONTRIBUTION TO THE PENTAGON PARAMETERS OF PM_{10}

3.1. Seven rules of thumb or assumptions

On the basis of seven rules of thumb, which are derived from measurements, reasoning, modelling or assumptions and which will be dicussed later (section 3.4.), an estimate will be presented of the way the aerosol mass is believed to be distributed over the different parts of the PM_{10} pentagon in the Netherlands. The reasoning, underlying data and assumptions are presented in the form of seven rules of thumb, used to indicate the composition of PM_{10} in the Netherlands, as shown in section 3.2. The division between Dutch and foreign contributions to the levels of PM_{10} in the Netherlands is presented in section 3.3. As such, each of the rules of thumb is supposed to be the most likely central estimate for the PM situation in the Netherlands. For the underlying evidence reference is made to the cited literature. When there are no literature references available, the reasoning for the choice of a certain value is made explicit. In section 3.4 a range of possibly plausible values for the rules of thumb is evaluted. Section 3.5 presents estimates of the possible ratios of Dutch versus foreign contribution. The fact that the rules of thumb or assumptions are supposed to be central estimates is an important element in the logical reasoning done later on in this chapter. The fact that these rules are supposed to be central estimates indicates, that for instance a Monte Carlo analysis will probably lead to rather similar results. Different values (meaning other central estimates) for the set of rules of thumb will lead to different outcomes, though. Therefore a clear distinction has to be made between the values that are attributed to the various rules of thumb and the logical frame work of the system of explicit and implicit rules of thumb and assumptions that has been developped in this report. If the central estimates that have been used in this excercise are incorrect, they have to be changed. But this does not impede the use of such a logical framework as a tool to generate hyopotheses for the PM situation in the Netherlands.

It has to be stressed that the following rules of thumb only hold for the yearly averge values for the Netherlands. For shorter time periods than a year and on smaller geographical scales, deviations of calculated concentrations, using the rules of thumb, of sometimes more than an order of magnitude may occur.

1. The first rule of thumb

Sixty percent of the PM_{10} mass is composed of particles with a 50% cut-off diameter of 2.5 μm and 40% of the mass is composed of particles between 2.5 and approximately 10 μm.
This will be indicated later on as: < 2.5 and > 2.5.

This first rule of thumb is based on results of particle measurements in the Netherlands collected with size specific sample heads for aerosol classification with cut-off diameters of 2.5 and 10 µm. Janssen et al.(1997) report that 57% of the PM_{10} mass is composed of particles with a 50% cut-off diameter of 2.5 µm for 56 (8 h average) filter measurements carried out in Arnhem. Van der Zee et al.(1997) report that 65% of the PM_{10} mass is composed of particles with a 50% cut-off diameter of 2.5 µm for ~ 80 (24 h average) filter measurements done in Amsterdam. As a best estimate for this rule of thumb the following value has been chosen: 60% of the PM_{10} mass in the Netherlands, in the nineties, is composed of particles with a 50% cut-off diameter of 2.5 µm. A sensititivity analysis will be carried out to test this rule of thumb in section 3.4.

2. The second rule of thumb

Fortytwo percent of the PM_{10} aerosol mass in the Netherlands is secondary aerosol.
This rule of thumb is based on the mean of the yearly averaged results of daily measurements at five rural locations from the National Air Quality Monitoring network (RIVM, 1994), which indicate that 33% of the weight of PM_{10} is secondary aerosol, as determined by wet chemical analysis. In such a chemical analysis the amount of water associated with the secondary aerosol can not be determined, though this water probabely has been included in the mass of the aerosol dertermined by filter weighing or Beta-gauge. A central estimate of 10% water in the PM_{10} measured in the Netherlands seems reasonable. As this water will be mainly associated with the primary and secondary salts and sea salt also is a constituent of the Dutch aerosol, a value of 42% seems a reasonable central estimate for the fraction of secondary PM_{10} in the Netherlands.

3. The third rule of thumb

Eightyfive percent of the mass of the secondary aerosol is < 2.5 µm and 15% is > 2.5µm.
This rule of thumb is based on the annual mean of daily measurements at a location in the centre of the Netherlands (Speulder forest) (Erisman et al., 1996).

4. The fourth rule of thumb

There are no carbonaceous particles > 2.5 µm.
By the definition in section 2 the carbonaceous aerosol by combustion sources is totally in the accumulation mode with a diameter up to 2.5 µm, as it caused by combustion. The predominantly submicron size distribution of the major contributor to the carbonaceous aerosol, traffic, is presented by Van de Brink (1996).

5. The fifth rule of thumb

The contribution of foreign emissions to primary particle levels < 2.5 μm in the Netherlands will be approximately 70%.

For the fifth rule of thumb is no simple reference in the literature. It is based on the outcomes of a tracer which is used as analogy, when the foreign contribution of a suitable tracer is known, this value may be used to estimate the foreign contribution of primary $PM_{2.5}$.

Primary particles in this size class in the Netherlands are emitted mainly by stationary and non-stationary combustion sources. For such sources the concentrations of ambient atmospheric lead may be used as a particulate tracer. Nowadays lead can still be considered a tracer for the traffic contribution, even though the levels of lead in petrol have dropped considerably during the last decade and more cars are using lead free petrol, LPG or diesel. More than half of the Dutch lead emission is by cars and the remaining part is by industry. Approximately 85% of the industrial lead emissions in the Netherlands have a MMD < 1 μm. Therefore > 90% of the particulate lead emissions in the Netherlands is submicron and it seems valid to use the average national lead concentrations in the Netherlands and the foreign contribution of lead as a tracer for the foreign contribution of primary $PM_{2.5}$ This use of lead as a tracer can be confirmed with data of Erisman et al.,1996. They indicate that the dry deposition velocity of lead is the same as for sulphate. Therefore lead my be considered, just as sulphate, as a suitable tracer for the long range transport of $PM_{2.5}$. The RIVM (1995) estimates that the foreign contribution to lead levels in the Netherlands is approximately 70%. Analogous to this, the foreign contribution of primary $PM_{2.5}$ is taken to be approximately 70%.

6. The sixth rule of thumb

Eight percent of PM_{10} in the Netherlands consists of carbonaceous aerosol.

As there are no yearly average measurements of the carbonaceous aerosol in the Netherlands its value has to be estimated. If we assume that the carbonaceous emission in the Netherlands is slightly more than 20 ktonnes per year (RIVM, 1997) and if we use the box model described in Appendix A, a contribution to the average (rural) background concentrations in the Netherlands can be calculated of approximately 1µg/m^3. Using the fifth rule of thumb[1] with an average foreign contribution of ~ 70 % to the concentrations of carbonaceous aerosol in the Netherlands, a yearly average concentration of ~ 3.3 µg/m^3 of carbonaceous $PM_{2.5}$ results. As the yearly average level of PM_{10} in the Netherlands is ~ 40 µg/m^3, this means that approximately 8 % of the PM_{10} in the Netherlands consists of carbonaceous aerosol.

7. The seventh rule of thumb

Approximately 75% of the secondary aerosol in the Netherlands is of foreign origin.

Calculations by the OPS model with the European emissions of the precursor gases for secondary PM (SO_2, NO_x and NH_3) have indicated a perfect match between predicted and measured secondary PM values in the rural Netherlands (RIVM, 1997). From these calculations it follows that 76% of the precusors of the secondary aerosol are emitted by foreign sources.

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[1] Implicitly more than the mentioned seven rules of thumb have to be considered in order to solve the twelve unknowns presented in the tables of 3.3. These implicit rules of thumb follow logically from the seven explicitely formulated rules. For instance when rule of thumb #4 states that there are no carbonaceous particles by combustion sources that are > 2.5 µm, this statement is deemed to be true for Dutch and foreign sources. When rule of thumb #5 states that 70% of the primary $PM_{2.5}$ is from foreign origin, this of course also relates to foreign sources of carbonaceous particles, as the are < 2.5 µm. Another of such implicit rules is that the size distribution of secondary aerosol for Dutch and foreign sources is considered to be similar.

3.2. Estimation of the distribution of the pentagon of PM_{10} in the Netherlands

A picture of the distribution of the different parts of the pentagon can be given on the basis of five of the rules of thumb (#1, #2, #3, #4 and #6). The results of the application of these rules are shown in Table 1.

Table 1 *Estimated distribution (%) of yearly average concentrations of* PM_{10} *in the Netherlands for the different parts of the pentagon*

	primary			
	carb.	non-carb	sec.	
< 2.5	8	*16*	36	60 %
> 2.5	0	*34*	6	40 %
	8 %	50%	42 %	100 %

The numbers in Roman script appear in the table on the basis of five of the rules of thumb, discussed previously. The numbers in italics can be easily deduced from the resulting two equations with two unknowns. In these and in the following calculations the distribution over the various boxes in the tables is rounded off to the nearest integer, while assuring that the numbers of all the boxes amount to a total of 100(%).

3.3. Estimation of the distribution for the PM_{10} pentagon, broken down into Dutch and foreign contributions

In combination with the other two rules of thumb (5 and 7) and the implictit rules it is possible to break down the PM_{10} distribution into national (Dutch) and foreign contributions. It is interesting to see how the contribution of different parameters for the PM_{10} pentagon changes when a distinction is made between aerosol emitted in the Netherlands and aerosol originating from foreign countries. In the Integrated Criteria Document Fine Particulate Matter (Annema et al., 1996), 20% of our PM_{10} is concluded to originate in the Netherlands and the remaining 80% is of foreign origin. This statement, the 20-80 statement, will be taken as a basis for the next exercise.

If the national contribution to levels of primary particles < 2.5 um in the Netherlands is 30 % (rule of thumb #5) and if the unknown contribution of this part of the aerosol is called x, it means that the ratios of Dutch and foreign aerosol in this size range are $x : (2.3)x$. When the other unknowns are called y and z, the following table results under the assumption of 20 % local and 80 % foreign contribution:

	Netherlands 20 %			Foreign origin 80 %			
	carb.	non-carb	sec.	carb.	non-carb	sec.	
< 2.5	2	x	9	6	$2.3x$	27	60
> 2.5	0	y	1	0	z	5	40
	2	8	10	6	42	32	100

We arrive at a system of three equations and three unknowns, which can be easily solved:

$3.3x = 16$so $x = 5$

$x + y = 8$so $y = 3$

$2x + z = 42$so $z = 31$

This leads to the following distribution, illustrated in Table 2.

Table 2 *Estimated distribution (%) of yearly average concentrations of PM_{10} for the different parts of the pentagon, with a contribution of 20% from the Netherlands and 80% from foreign sources*

	Netherlands 20%			Foreign origin 80%			
	carb.	non-carb	sec.	carb.	non-carb	sec.	
< 2.5	2	5	9	6	11	27	60
> 2.5	0	3	1	0	31	5	40
	2	8	10	6	42	32	100 %

In the opinion of the authors the figures from Table 2 should not be considered to represent a realistic picture of the PM situation in the Netherlands. The reasons for this rejection of the results are fourfold, the first two arguments are more of an absolute nature, whilst the second two are more relative.

I. The first reason is that the resulting 3 % of the Dutch PM_{10} concentrations (~ 1 $\mu g/m^3$) in the category of non-carbonaceous > 2.5 μm is less than the average yearly concentrations which are regularly measured from Dutch sources.

II. The second reason is that the resulting 31% of foreign contribution (~ 12 $\mu g/m^3$) for this non-carbonaceous particles > 2.5 μm is much more than can be accounted for in modelling excercises and can be explained with the mean residence times and travelling distances of aerosols in this size category (Table 11).

III. The third reason is that the fraction of foreign $PM_{2.5}$ is much less than can be derived from modelling exercises.

IV. The fourth reason is that the fraction of foreign PM > 2.5 μm (coarse PM) in relation to Dutch coarse PM is much higher in Table 2 than can be derived from modelling excercises.

The four arguments will now be elaborated.

Ad I.

The first reason to reject the results of Table 2 is that the Dutch fraction of the non-carbonaceous PM > 2.5 µm of approximately 1 µg/m^3 of the PM_{10} seems to be too low. The picture resulting from this table does not correspond with the local contribution of for instance sea salt, resuspended crustal material or other Dutch sources of “coarse” PM_{10}. Sea salt is an example of non-carbonaceous aerosol with an aerodynamic diameter predominantly above 2.5 µm. The calculation of the contribution of sea salt aerosol to PM_{10} was based on the measured Cl^- concentrations in particulate material, which have been multiplied by 1.75 to convert them to NaCl (see Chapter 4). The yearly average concentration of sea salt can be estimated at 5-7 µg/m^3 in the coastal part of the country, decreasing to 2-3 µg/m^3 in the east of the country (Van der Wal and Janssen, 1996 and Chapter 4). Size selective measurements by Erisman et al., 1996 in the centre of the Netherlands show that the yearly average sea-salt aerosol >2.5 µm is approximately 1.6 µg/m^3. Additionally to sea salt the yearly average crustal contribution to this size category is estimated to be higher than 2.2 µg/m^3 for the Netherlands (RIVM,1997). The combination of these two sources already leads to an average Dutch contribution of 3.8 µg/m^3 or approximately 10% of the non-carbonaceous and non-antropogeneous primary PM in the size range > 2.5 µm. The antropogeneous local Dutch contribution of PM in this size range has not been taken into account yet in the previous figure of approximately 10%. TAUW (1996) has estimated a non-neglible Dutch emission in this coarse size range. The known measurements of natural aerosols and the antropogenic emissions obviously contradict with the calculated 1 µg/m^3 of Table 2. This would imply by consequence that one (or more) of the assumptions or rules of thumb used to derive Table 2, is incorrect.

Ad II.

The second argument is that a foreign contribution of over 30% seems to be unrealistically high for this coarser part of PM_{10} with a limited residence time due to dry deposition. According to Table 10 particles with a diameter of > 2.5 to 10 µm have a transport range of up to approximately 200 km. Calculations with the OPS model based on PM emissions for the whole of Europe (RIVM, 1997), indicate that the foreign contribution to the Dutch yearly average concentration in this size category > 2.5µm is 0.9 µg/m^3 and much less than the ~12 µg/m^3 implied by Table 2. Even if the rather extreme position would be chosen that European emissions have been underestimated by a factor of 4 to 5, the resulting yearly average European PM_{10} contribution in this size category >2.5 µm would be approximately 4 µg/m^3, which is still far off from the value of approximately 12 µg/m^3 in Table 2. (Another possible reason for this mismatch, a fault in the modelling, has been refuted by Van Jaarsveld, 1995. The OPS model has proven itself a suitable tool for dispersion calculations of yearly average concentrations on a European scale). The second argument to reject the results of Table 2, based on the foreign contribution > 2.5 µm, also indicates that one (or more) of the assumptions or rules of thumb used to derive Table 2 is incorrect.

If in Table 2 the 12 μg/m^3 of foreign contribution and 1 μg/m^3 of Dutch contribution would have been correct, it implies that > 90% of the primary PM > 2.5 μm in the Netherlands is of foreign origin. Also this figure is unrealisticly high as one of rules of thumb (#5) indicates that of the primary $PM_{2.5}$ of smaller particles the foreign contribution is approximately 70%. On the basis of these arguments the foreign contribution of larger particles, with shorter residence times and higher deposition rates, therefore cannot be higher than the 70% of rule of thumb#5.

Of course one has to explore first if the contribution of our neighbouring countries for the various PM size categories is not very dissimilar to that of the Netherlands.

Table 3. *PM emissions in tonnes/km^2 by the Netherlands (1993) and by neighbouring countries (1990). (RIVM, 1997)*

	PM_{10} tonnes/km^2	$PM_{2.5}$ tonnes/km^2	coarse PM_{10} > 2.5 μm tonnes/km^2
Belgium	2.8	1.6	1.2
France	0.7	0.4	0.3
former GDR	3.5	2.2	1.3
Germany	1.7	1.0	0.7
Netherlands*	1.6	0.9	0.7
United Kingdom	1.1	0.6	0.5

* Apart from the year, also the basis for the emission data was slightly different for the Netherlands compared to other European countries

Table 3 indicates that the emissions for the Netherlands and its neighbours for the coarse PM fraction are not very dissimilar, even is the possibility of a relative underestimation for the Dutch data, or conversely an overestimate of the European emssions, is considered. For the former GDR it has to be born in mind though, that the emissions are for the year 1990, less than a year after the 'Wende', when a lot of the polluting industries were still operating which were closed down by 1993, the base year for the calculations of this report.

Ad III.
The third argument to reject the results of Table 2 can de derived from modelling. Modelling the yearly average concentrations with the OPS model and using the European PM emissions leads to a $PM_{2.5}$ to PM_{10} ratio of 76% for the European contribution of primary PM_{10} (RIVM, 1997). This figure is much higher than the 35% that can be derived from Table 2 as the $PM_{2.5}$ to PM_{10} ratio of the European contribution of primary PM. Even though the absolute values of the European (and Dutch) emissions may not be very certain, such a ratio is influenced mainly by the geographical scale and the proces of dispersion , conversion and deposition and to lesser extent by the absolute values of the various emissions. Therefore such a ratio of foreign primary $PM_{2.5}$ to PM_{10} on the basis of a modelling exercise could be used a guide to evaluate the probability of various alternatives that are presented in section 3.5. As the third argument, like the previous two, also leads to a contradiction between expected results and Table 2, this indicates that one (or more) of the assumptions or rules of thumb used above is incorrect.

Ad IV.
The ratio in Table 2 between the Dutch and foreign contribution to primary coarse PM, which is about 0.1, is considered to be unrealisticly low taking into account the mean transport distances (Table 10) and national and foreign emission profiles (Table 3).

3.4. Discussion on the assumptions 1 to 7

After the conclusion in the previous section that one (or more) of the assumptions is incorrect, it has to be determined which one(s) is (or are) the most likely to be incorrect. It has to be born in mind though, that this excercise is a thought experiment to be used as a way of generation of hypothesis and ideas for further experimental research, so no conclusive answer should be expected.

First of all the central estimate of the seven rules of thumb will be analysed and after that it will be explored if with some bending of the rules the calculated results based on the stated 20% Dutch and 80% foreign contribution to PM_{10} (the 20-80 statement) can be accomodated with the existing knowledge of PM_{10}.

Rule of thumb #1.

This rule of thumb, indicating that 60% of the PM_{10} mass < 2.5 μm, seems to be in the low end of the range as data from the UK indicates 62% for an urban site and 64% for a rural site (QUARG, 1996). Data from the USA indicate 63% for the North-Eastern region of the US (EPA, 1996) and a high value of 70% for Washington DC. (For the arid regions in the South-Western part of the Sates this may drop to 37-49%, but for the Netherlands such low figures are of no concern as we have no deserts and a different climate.) An extreme value for this rule of thumb therefore seems to be 70%. So as a matter of sensitivity analysis an alternative rule of thumb is formulated (#1'): *Seventy percent of the PM_{10} mass is composed of particles with a 50% cut-off diameter of 2.5 μm and 30% of the mass is composed of particles between 2.5 and approximately 10 μm.*

Calculations with rule of thumb #1', similarly as done previously for Table 2, lead to the result that the local Dutch contribution to primary PM_{10} >2.5 μm becomes zero, and that the foreign contribution in this class of PM is 24% of the PM_{10} (~10 μg/m^3), indicating that 100% of the coarse particles in the Netherlands are of foreign origin. The foreign $PM_{2.5}$ contribution is 50% of the foreign contribution to PM_{10}. The reasons for rejecting the original results in Table 2 have only become stronger with this alternative #1' for the decisive criterium stated in 3.4.

The discussion of rule of thumb #1 may be concluded by considering that the range of values 60-70% all lead to a violation of the decission criteria.

Rule of thumb #2.

This rule, which assumes that 42% of the PM_{10} is secondary aerosol in the Netherlands, is based on filter measurements by LVS and subsequent wet chemical analysis of the filters, in this figure it is assumed that approximately one quarter of this figure is water associated with the secondary salts. The sampling efficiency of the LVS is partly wind speed dependent, as 85% of the secondary aerosol is < 2.5 μm this will probably not influence the measured secondary aerosol concentrations to a great extent. In the Netherlands measured values of secondary aerosol are therefore not customarily corrected for the sampling efficiency. Water is not analysed chemically and water associated with the secondary aerosol might distort the picture on which rule 2 is based. The PM_{10} concentration is determined physically (absorption of beta-radiation) by a FAG as total mass, so it incorporates any PM associated water. The influence of humidity on the aerosol mass is minimised in the National Air Quality Monitoring Network by heating the sampling tube. Combining a chemical and physical analysis in one figure (rule of thumb #2) might thus lead to a problem of validity with this rule.

For the sake of argument two alternatives are formulated, the sensitivity of which will be analysed both. Let us assume that 0% and 100% of the mass of secondary aerosol is associated with water.

This would lead to two alternative rules of thumb;
(#2'): *Thirtythree percent of the PM_{10} aerosol mass in the Netherlands is secondary aerosol.*
or rule of thumb
(#2''): *Sixtyseven percent of the PM_{10} aerosol mass in the Netherlands is secondary aerosol.*
The effect of both alternative rules of thumb will be evaluated on the results of Table 2 and the subsequent arguments to accept or reject the results.
Rule of thumb #2' would lead to a local Dutch contribution of coarse PM_{10} of 2% (~1 μg/m^3), a foreign contribution of coarse PM_{10} of 33% (approximately 13 μg/m^3) and a foreign $PM_{2.5}$ contribution of 42% of the foreign primary PM_{10}, which on the basis of the prior reasoning would also lead to the same conclusion that one (or more) of the assumptions is incorrect.
Rule of thumb #2'' even leaves less room for a local Dutch contribution of primary PM as 17% of the 20% Dutch contribution then consists of secondary aerosol. Therefore the conclusion is the same as for #2'.
The discussion of rule of thumb #2 may be concluded by considering that the range of values of the mass of the secondary aerosol with 0-100% of associated water all lead to a violation of the decission criteria.

Rule of thumb #3.
The third rule of thumb, that 85% of the secondary aeosol < 2.5 μm, has been derived directely from measurements in the Netherlands. However, this does not necessarily indicate it to be correct. In the UK (QUARG, 1996) a slightly different particle size distribution for the secondary aerosol has been reported: for an urban situation 82% of the secondary aerosol <2.5 μm and for a rural station this was 80%. Let us therefore explore an extreme alternative to test this rule to its limits.
Rule of thumb
#3': *Seventyfive percent of the mass of the secondary aerosol is < 2.5 μm and 25% is > 2.5μm.*
Calculations with rule of thumb #3' lead to 2% (~1 μg/m3) of Dutch contribution to coarse PM_{10}, 28% (~11 μg/m^3) of foreign contribution and a foreign primary $PM_{2.5}$ contribution of 42% of the foreign PM_{10}, which also violates the decission criteria.

Rule of thumb #4.
The fourth rule of thumb, carbonaceous aerosol < 2.5 μm, is based on the definition of carbonaceous aerosol as being a combustion aerosol and the fact that combsution aerosols are predominatly submicron in size range (Van den Brink, 1996). Of course it could be possible to formulate alternatives of this rule of thumb by assuming that part of the carbonaceous PM would be in the size range > 2.5 μm. This though, would leave less mass for the Dutch non-carbonaceous > 2.5 μm and less mass for the foreign $PM_{2.5}$ and therefore would lead to the same conclusion as is drawn from rule of thumb #4.

Rule of thumb #5.

The fifth rule of thumb, assuming that 70% of primary $PM_{2.5}$ is of foreign origin, seems to be rather sensitive for the basic assumptions, as it can not be substantiated by direct measurements and needs additional arguments. It may be argued for instance, because 75% of the secondary aerosol seems to be of foreign origin (rule of thumb #7), as 15% of the secondary aerosol seems to be > 2.5 µm (rule of thumb #3) and knowing that particles > 2.5 µm generally do not travel much further than 200 km (see table 10), that it might be reasonable to estimate that more than 75% of the primary $PM_{2.5}$ would be of foreign origin. Maybe 80% would be a reasonable estimate as an alternative rule of thumb for the foreign contribution to primary $PM_{2.5}$. (Annema et al.,(1996) even states a foreign contibution of 85% of all primary PM, including coarse PM_{10}, this will be treated later on as alternative #5''.).

Rule of thumb #5': *The contribution of foreign emissions to primary particle levels < 2.5 µm in the Netherlands will be approximately 80%.*

Before any calculations can be made, it has to be remembered that rule 5 (or any alternative) is not independent from rule 6. Based on the contribution of the foreign primary $PM_{2.5}$ the carbonaceous content of the Dutch aerosol has been calculated by augmenting the modelled Dutch carbonaceous contribution of 1 $\mu g/m^3$ with the fraction of the foreign contribution to primary $PM_{2.5}$. If a foreign contribution of 80% is assumed as in rule of thumb #5', this larger foreign contribution automatically leads to a higher yearly average carbonaceous content, so keeping the carbonaceous aerosol fraction in the aerosol fraction <2.5 constant.. The original 8% carbonaceous aerosol now becomes 12%.

Rule #5' leads to 6% (~2 $\mu g/m^3$) of primary Dutch coarse PM_{10}, 28% (11 $\mu g/m^3$) of foreign coarse PM_{10} and a foreign $PM_{2.5}$ contribution of 42% of the foreign PM_{10}, which on the basis of the prior reasoning would also lead to the same conclusion that one (or more) of the assumptions is incorrect.

A foreign contribution of 75% to secondary aerosol levels in the Netherlands could be used as a plea for a higher foreign contribution than 70% for primary $PM_{2.5}$, as has been done in the motivation of rule of thumb #5'. However, secondary aerosol is formed from primary gaseous emissions after chemical conversions, which takes time. From a gas it becomes an aerosol. Depending on the reaction velocities for the different gasphase and heterogeneous reactions this may be a matter of hours to a day (or days). Because the residence time in the Netherlands may be estimated to be < 10 hours (based on the modelling of the Netherlands as a 'square surface' with sides of 200 km and an assumed mean transport velocity of 6 m/s) it could be assumed that the Dutch contribution to the precursors of the secondary aerosol < 2.5 μm could very well be the 30% implied by rule of thumb #5, while at the same time rule of thumb #7 is correct. This at a first sight contradictory statement can be easily explained. For the sake of argument let us assume that the Dutch contribution of secondary aerosol <2.5 μm is emitted as PM, than rule #5 indicates that the Dutch contribution to the secondary aerosol would be 30%. As it is not emitted as PM but as a precursor gas which needs time to become PM, in this time the gas is transported out of the country and therefore the Dutch contribution to secondary PM becomes lower than 30%. The value of 24% from model calculations therefore may accomodate with rule #5. As such the 75% foreign contribution of rule #7 is no argument against the correctness of rule of thumb #5.

Based on the statement of Annema et al.,(1996) that 85% of primary PM is of foreign origin a second alternative to rule of thumb #5 may be formulated.
Rule of thumb #5'': *The contribution of foreign emissions to primary particle levels < 2.5 μm in the Netherlands will be approximately 85%.*
If a foreign contribution of 85% is assumed a yearly average carbonaceous content of 16% is estimated. Rule #5'' leads to 7% (~3 μg/m^3) of Dutch coarse PM_{10}, 27% (~11 μg/m^3) of foreign coarse PM_{10} and a foreign $PM_{2.5}$ contribution of 42% of the foreign PM_{10}. Though the value of 7% of the Dutch contribution to coarse aerosol is nearing the ~10% of the natural primary contribution to the coarse aerosol in the Netherlands it still does not take any antropogenous coarse aerosol into account. Based on the prior reasoning this leads to the same conclusion that not all decission criteria are fullfilled.
With rule of thumb #5'' the original postulate of Annema et al., (1996) is not realised yet, as only 80% of the primary PM is of foreign origin and not 85%. Therefore a third alternative is explored that postulates that 90% of primary PM is of foreign origin.
Rule of thumb #5''': *The contribution of foreign emissions to primary particle levels < 2.5 μm in the Netherlands will be approximately 90%.*
If a foreign contribution of 90% is assumed a yearly average carbonaceous content of 24% is estimated in the Netherlands. Rule #5''' results in 8% (~2 μg/m^3) Dutch contribution to primary coarse PM_{10}, 26% (~10 μg/m^3) of foreign coarse PM_{10} and a foreign $PM_{2.5}$ contribution of 46% of the foreign PM_{10}. This eventually leads to the same conclusions as those of rule #5'' and does not fullfill the decission criteria.

Rule of thumb #6.

Rule of thumb #6, that 8% of PM_{10} consists of carbonaceous aerosol, has been expored a little bit by the influence of the alternatives of rule #5, but also can be easily analysed by itself by doubling or halving the carbonaceous content as we do not have a sound basis for its value.

Rule of thumb #6':*Sixteen percent of PM_{10} in the Netherlands consists of carbonaceous aerosol.*

Rule #6' leads to 3% (~1 µg/m^3) of Dutch contribution to primary coarse PM_{10}, 31% (12 µg/m^3) of foreign coarse PM_{10} and a foreign primary $PM_{2.5}$ contribution of 35% of the foreign primary PM_{10}. Based on the prior reasoning this leads to the same conclusion that one (or more) of the assumptions is incorrect.

Rule of thumb #6'':*Four percent of PM_{10} in the Netherlands consists of carbonaceous aerosol.*

Rule #6'' leads to 3% (~1 µg/m^3) of Dutch contribution to primary coarse PM_{10}, 31% (~12 µg/m^3) of foreign coarse PM_{10} and a foreign primary $PM_{2.5}$ contribution of 35% of the foreign PM_{10}. Based on the prior reasoning this leads to the same conclusion that one (or more) of the assumptions is incorrect.

A further conclusion could be that the calculated results do not seem very sensitive to a doubling or halving rule 6. The discussion of rule of thumb #6 may be concluded by considering that the range of values of the carbonaceous content 4-16% all lead to the rejection of one or more of the rules and postulates.

Rule of thumb #7.

Rule #7, that 75% of the secondary aerosol is of foreign origin, is based on model calculations. Though the models describing the secondary aerosol concentrations seem rather trustworthy as the modelled and measured yearly average values of secondary aerosol match with an error of 1% (RIVM,1997). Nevertheless two alternatives are explored ± 5% of the calculated ratios.

Rule #7':*Approximately 70% of the secondary aerosol in the Netherlands is of foreign origin.*

Rule #7' leads to 0% (0 µg/m^3) of a Dutch contribution to primary coarse PM_{10}, 34% (~13 µg/m^3) of foreign coarse PM_{10} and a foreign $PM_{2.5}$ contribution of 33% of the foreign PM_{10}. Based on the prior reasoning this leads to the same conclusion that the decission criteria are not met.

Rule #7'':*Approximately 80% of the secondary aerosol in the Netherlands is of foreign origin.*

Rule #7'' leads to 5% (2 µg/m^3) of a Dutch contribution to primary coarse PM_{10}, 29% (~12 µg/m^3) of foreign coarse PM_{10} and a foreign $PM_{2.5}$ contribution of 34% of the foreign primary PM_{10}. Based on the prior reasoning this leads to the same conclusion that the decission criteria are not met.

Conclusions

After this analysis of these seven rules of thumb the first conclusion could be that, assuming the correctness of the statement that 85% of the primary PM is of foreign origin and at the same time assuming the correctness of their other statement, that 80% of PM_{10} in the Netherlands is of foreign origin (Annema et al., (1996)), leads to an inconsistency and therefore at least one of the two postulates has to be incorrect.

A second conclusion could be that reasonable ranges of ± 5% or ± 10% of the rules of thumb (#1, #3 and #7) which are based on measured and modelled parameters, and wider ranges of -100% and + 100% when the rules are not based on measurements but on analogies (#2, #5 and #6) all lead to conclusion that the decission criteria are not met when the rules are analysed one by one.

An exercise has been done to explore if, with some bending of the rules, the calculated results on the basis of the postulate of 20% local Dutch and 80% foreign contribution to PM_{10} and the existing knowledge of PM_{10} can be accomodated in any way. In no way this is the case, it may be concluded that bending the rules and streching them beyond their limits leads to inconsistencies.

The sensititvity of plausible alternatives of the seven rules of thumb has been examined one by one and none of these alternatives has led to a situation in which no inconsistencies arose. Therefore it is concluded that the inconsistencies arrise from the 20-80 statement as such and that this statement is the more likely to be incorrect than any of the rules of thumb. Of course in this stage it can not be ruled out that varying a number of rules simulataneously might also lead to the inconsistencies, allthough this possibility has been minimised by using the central estimates. In Table 4 a simultaneous variation of rules and assumptions is analysed.

3.5. Estimates of the likely ratios of Dutch versus foreign contributions

By estimating various ratios of local Dutch versus foreign contribution of PM_{10} and indicating the most likely ones, hypotheses are generated that are to be tested. The excercise with the 20-80 statement has presented some possible outcomes. Table 4 presents the percentage of foreign primary PM > 2.5 μm of all primary coarse PM for various ratios (from 15-85 to 50-50, Dutch versus foreign contribution of PM_{10}). In Table 4 a value of 100 indicates that all primary coarse PM is of foreign origin, a value of 0 means that all PM >2.5μm is of local Dutch origin.

Both extremes are unlikely. More boudaries of plausible values may be given by realising that the foreign contribution to PM > 2.5 μm has to be lower than the foreign contribution to $PM_{2.5}$ as particles > 2.5 μm have a higher depostion velocity (see table 10).

Another consideration can be that the average travelling distance of particles > 2.5 μm is approximately 200 km (van Jaarsveld, 1996). This implies that when the Netherlands is considerd to be a square of 200 x 200 km (appendix A), the foreign contribution in this size class would be approximately 50%, if all foreign countries have similar emission densities per km^2 as the Netherlands (Table 3). Though this last condition is reasonably met, the local geographical situation and the meteorology in the Netherlands indicate that 50% of foreign contribution probably is too high, as approximately 3/8 of the Dutch 'square' of 200 x 200 km is surrounded by squares of sea with little emissions. Taking this geographical situation and the prevailing westerly winds into account and the fact that Belgium has a somewhat higher emission density than its neighbours (Table 3), possibly a value of 35% for the foreign contribution would be appropriate. In Table 4 the results that fullfill this requirement of 35 ± 5% will be put in a dotted rectangle (a range of 30-40% foreign contribution of coarse primary PM is deemed plausible).

A second criterium to evaluate the various alternatives has been presented before. This is the value of 75% of the ratio of foreign primary $PM_{2.5}$ versus primary PM_{10} resulting from the modelling exercise with the emissions from all over Europe (RIVM,1997). When the calculated ratio of foreign $PM_{2.5}$ versus PM_{10} is 75 ± 10% (a range of 65-85% is deemed plausible for the ratio of foreign primary $PM_{2.5}$ to PM_{10}) this range is indicated with a colsed rectangle in Table 4. For the different alternative of rules of thumb #5 the calculations have been executed, as rule #5 is the assumption that is the least based on existing measurements or modelling results. In this way it is possible to analyse the sensitivity of the different alternatives of rule of thumb #5 in combination with one of the other alternatives.

Table 4 *Percentage of foreign versus Dutch contribution to primary PM >2.5 μm for the different alternatives of rule of thumb #5 for various postulates of Dutch versus foreign* PM_{10}

Dutch versus foreign contribution to PM_{10}	Rule of thumb #5	Rule of thumb #5'	Rule of thumb #5''
15-85	100	97	94
20-80	93	82	79
25-75	79	68	65
30-70	64	53	50
35-65	49	38	35
40-60	34	24	21
45-55	19	9	6
50-50	4	0	6

Table 4 shows that the possible alternatives for the local Dutch versus foreign ratio of PM_{10} lie between 35-65 and ~45-55. If the rules of thumb #5 is taken to be correct, it narrows down to between 40-60 and 45-55 as a possible range for the Dutch versus foreign contribution to PM_{10}. In Table 5 the postulate of 40% local Dutch contribution to PM_{10} and 60% foreign contribution is explored. The rules of thumb that are used are the same seven rules as before.

Table 5 *Estimated distribution (%) of yearly average concentrations of* PM_{10} *for the different parts of the pentagon, with a contribution of 42% from the Netherlands and 58% from foreign sources*

	Netherlands 42%			Foreign origin 58%			
	carb.	non-carb	sec.	carb.	non-carb	sec.	
< 2.5	2	5	9	6	11	27	60
> 2.5	0	25	1	0	9	5	40
	2	30	10	6	20	32	100 %

Table 5 complies with both of the boundary conditions that 30-40% of the primary PM >2.5 μm has to be foreign (36%) and the other that: foreign primary $PM_{2.5}$ is 65-85% of foreign PM_{10} (65%). As both boundary conditions are not based on actual data, a range of 40-60 to 45-55 for the Dutch versus foreign contribution seems plausible. If a central estimate was preferred though, based on the seven rules of thumb the values of Table 5 emanate.

The proof of the pudding is the eating. Are there any measurements or modelling excercises that can confirm the results of Table 5?
A honest answer would be: not really. Though this report is a hypothesis generating excercise, an attempt will be made to answer the previous question.

Reconciliation of this 25% of a Dutch contribution to primary coarse PM_{10} with measured and modelled values indicates that one has to be able to explain 10 $\mu g/m^3$ for a yearly average PM_{10} concentration of 40 $\mu g/m^3$. As has been presented earlier ~3.8 $\mu g/m^3$ in this size class can be explained by natural sources in the Netherlands. A modelling excercise with the Dutch emission inventory results in a local contribution of 0.4 $\mu g/m^3$ in this size range (RIVM, 1997). This indicates that still more than 50% of the local Dutch mass in this coarse size range is missing.

The 5% (2 µg/m^3) of the Dutch contribution to primary $PM_{2.5}$ can be explained better. Modelling leads to 0.6 µg/m^3 (RIVM, 1997) which still has to be augmented with the average sea spray contribution to this size class of 0.6 µg/m^3 and the crustal part of $PM_{2.5}$. The local Dutch primary $PM_{2.5}$ contribution seems to be estimated within a factor of 2 in Table 5, when the calculated value of 2 µg.m3 is compared with the modelled value > 1.2 µg/m^3.
A hypothesis generated by this excercise is that the missing mass of primary PM is suspected to be predominantly in the coarse mode.

The average foreign contribution of 9% (~4 µg/m^3) of coarse PM > 2.5 µm has to be reconciled with the modelled 0.8 µg/m^3. This still is a difference of a factor 4-5, but it is less big than the difference of a factor 14 resulting from the 20-80 postulate.

The average foreign contribution of primary $PM_{2.5}$ of 17% (~7 µg/m^3) has to be reconciled with the modelled 2.0 µg/m^3, which differs a factor of 3-4. The difference between modelled and calculated European values for $PM_{2.5}$ is less than for the coarse fraction of PM_{10}.

Table 5 is not based on measurements. Therefore this table should not be regarded as the definitive solution, but is to be seen more as a plausible hypothesis, as it does not violate any of the rules of thumb. Further research trying to identify missing sources by way of source apportionment and a better size classification of the whole size range of airborn PM should help to fill in the existing gaps in our knowledge.

As a final conclusion to this section, it can be stated that the model based on the seven rules of thumb and the implicit assumptions seems rather robust.
A hypothesis generated by this excercise is a higher local Dutch contribution to PM_{10} than originally envisaged in the 20-80 postulate. This assumption seems to remain unaffected under various assumptions.

In the next three sections an effort is made to complete the PM picture from what we know about sources of PM. There were we don't have the knowledge some likely values for the yearly average contributions of various source categories will be chosen. For various areas in the Netherlands the contributions of the source categories will be added and compared with the measured values by our National Air Quality Monitoring Network. It will be explored if the resulting guesstimates and measurements coincide.

4. THE CONTRIBUTION OF DIFFERENT SOURCE CATEGORIES AND FOREIGN COUNTRIES TO PM_{10} LEVELS IN THE NETHERLANDS

An estimate of the composition of PM_{10} in the Netherlands and the contributions of foreign countries to concentration levels of PM_{10} in the Netherlands were discussed in the previous chapter. This chapter describes the contributions of the different source categories in the Netherlands to PM_{10} concentration levels under the following categories:

- Natural sources of PM_{10} in the Netherlands
    - Resuspended aerosol (wind-blown crustal material)
    - Sea-salt aerosol
- Antropogenic sources of primary PM_{10} in the Netherlands
    - Traffic
    - Agricultural emissions
    - Large-scale combustion
    - Industry
    - Households
- Emittors of precursors of secondary PM_{10} in the Netherlands
- Foreign sources influencing PM_{10} in the Netherlands
    - Traffic
    - “Other” foreign sources

4.1. Natural sources of PM_{10} in the Netherlands

4.1.1. Resuspended aerosol (wind-blown crustal material)

There is very little information available on the contribution of soil particles to PM_{10} levels in the Netherlands. Bloemen et al.,(1996) reported very high contributions of wind-blown dust (>90%) to PM_{10} during dry periods at exceptionally high wind speeds (>10 m/s). During these episodes concentrations of PM_{10} > 150 μg/m^3 were measured. Simultaneous measurements of PM_{10} and ^{210}Pb (a radioactive element which is associated with soil) in Utrecht show a good correlation between concentrations of PM_{10} and ^{210}Pb (Rijnen, 1996; Van der Wal and Janssen, 1996). This indicates a noticeable contribution of crustal material to PM_{10} levels. Janssen et al., (1997) present some data of 14 measurements of Si concentrations in PM_{10} and $PM_{2.5}$ sampled at an urban background station. The median concentration of total Si in PM_{10} was 286 ng/m^3 and in $PM_{2.5}$ was 41 ng/m^3. If the $PM_{2.5}$ is assumed to originate from long range transport of antropogenic emissions (fly ash), the difference of PM_{10} and $PM_{2.5}$ indicates the contribution of the soil to PM_{10}. Using a mean Si content of 27.7% of the earth crust, a crustal contribution of 0.9 μg/m^3 to PM_{10} during the 14 days of sampling can be calculated. Erisman et al .,(1996) published results of measurements of concentrations of secondary aerosol and basecations in aerosols; see Tables 6 and 7.

Table 6 *Yearly average concentrations (μg/m^3) and size distribution in the Speulder forest (1995) and average rural concentrations (1993) of secondary PM_{10}*

	Rural 1993 (μg/m^3)	Speulder f.1995 (μg/m^3)	Percentage > 2.5 μm
sulphate	5.1	5.2	10
nitrate	5.0	5.9	19
ammonia	3.1	3.3	6

Table 7 *Concentrations of crustal elements (µg/m³) in the Speulder forest (1995)*

	Speulder f. 1995 (µg/m³)	percentage > 2.5 µm	percentage in earths crust [1]	contribution crustal material [2] (µg/m³)
Cl	1.1	64	<<1	[3]
Na	0.791	67	2.83	[3]
Ca	0.202	80	3.63	5.6
K	0.132	34	2.59	5.1
Mg	0.111	70	2.09	5.3 [3]

[1] The average percentage of different elements in the crustal material of the earth (Handbook of Chemistry and Physics, 56th ed. p. F-195.)

[2] The aerosol which contains these elements is found mainly in the coarser part of PM_{10} with an aerodynamic diameter > 2.5 µm. This often is an indicator of natural sources. A contribution of approximately 5 µg/m³ of crustal material to the PM_{10} concentrations can be derived from the data sampled in the Speulder forest, if the average occurrence of these elements in the Dutch topsoil are identical to those in the earths crust in general.

[3] Na and Mg are not only elements of the earths crust but also constituants of sea-salt aerosol. As sea spray is thought to be an important contributor to PM_{10} concentrations in the Netherlands, the total Na and Mg concentrations in PM_{10} cannot be attributed to soil dust alone.

If the ratio of 56.7% of Cl^- in sea water (Handbook of Chemistry and Physics, 56th ed. p. F-199) is used, and all Cl^- is assumed to be originating from sea spray, for the situation in the Speulder forest a yearly average contribution of 1.94 µg/m³ of sea-spray aerosol can be calculated. For Mg^{2+} which constitues 3.8% of the solid mass of sea water, it can be calculated that the contribution of sea spray to this component is 0.074µg/m³. In table 5 it is implicitely assumed that all the Mg^{2+} measured (0.111µg/m³) was originally crustal material. If Mg^{2+} contibution of sea spray is sub stracted from the measured Mg^{2+} concentrations than the Mg^{2+} contribution of soil dust to PM_{10} levels would be 0.037 µg/m³ ; with an Mg^{2+} content of 2.09 % this leads to a yearly average crustal contribution of approximately 1.8 µg/m³.

Therefore the calculated 5 µg/m³ seems to be a bit too high, a contribution of resuspended soil dust in the range 1-3 µg/m³ is preferred.

4.1.2. Sea-salt aerosol

Based on LVS measurements Van der Wal et al.,(1996) estimate a yearly averaged concentration gradient of calculated sea-salt aerosol of 5-7 µg/m³ near the coast in the western part of the Netherlands and 2-3 µg/m³ inland in the eastern part of the Netherlands at a few hundred kilometer from the coast.

These figures correspond with results of measurements reported by Erisman et al.(1996) above a forest in the centre of the Netherlands presented in Table 7. The results reported by Erisman et al.(1996) are collected by the same PM_{10} head as is used by the National Air Quality Monitoring network for the collection of size-dependant aerosols and therefore there is no need to correct them for the lower sampling efficiency of the LVS. The contrinbution of the directly measured sea-salt aerosol has been calcuated as approximately 2 µg/m^3. This similarity of the calculated and measured concentrations of sea-spray aerosol indicate that a correction for the lower sampling efficiency of the LVS is not really necessary.

4.2. Antropogenic sources of primary PM_{10} in the Netherlands

4.2.1. Traffic-related urban PM_{10}

Fischer et al.(unpublished data) did some measurements in Amsterdam (SAVIAH). In busy streets the mean concentrations were 43 µg/m^3 PM_{10} (n=18) and in streets with a low traffic density 36 µg/m^3, resulting in an average traffic contribution of 7 µg/m^3.

Van der Wal and Janssen (1996) analysed time-series of PM_{10} measurements in cities and background stations. They calculated a relative increase of about 6 µg/m^3 of PM_{10} concentrations in street stations on Sundays compared with working days. Assuming this relative increase to be due to halving all the PM emissions on Sundays, an upper limit of about 12 µg/m^3 due to both traffic and other urban and industrial emissions may be estimated. This leads to the following estimate of the primary PM_{10} contribution by traffic in an urban street

Table 8 *The contribution of primary PM_{10} by traffic in an urban street*

carb. tailpipe (< 2.5 µm)	non carb. resusp. dust (> 2.5 µm)	non carb. tyre wear (> 2.5 µm)	total PM_{10}
4.2 µg/m^3	3.7 µg/m^3	0.4 µg/m^3	8.3 µg/m^3

Besides this direct emission of PM_{10}, SO_2 and NO_x emissions from traffic are also supposed to contribute to PM_{10} concentrations in the Netherlands in the form of their conversion products: sulphates and nitrates. Because of the size range of these secondary aerosols (mainly < 2.5 μm) and the time which is needed to convert SO_2 to sulphates, and NO first to NO_2 and later on NO_2 to nitrates, this contribution is not supposed to affect the local geographical scale. In the Netherlands the contribution that the source category traffic makes to SO_2 and NO_x emissions is 18 % and 57 % respectively (Annual Report Air Quality, RIVM 1994). If this contribution were similar for a larger area in Europe, this would mean that on average the traffic contribution of Dutch and foreign sources to the secondary aerosol in the rural and urban background would be approximately 4.6 $\mu g/m^3$.

4.2.2. Agricultural emissions

An estimate for the aerosol contribution from Dutch pig farms may be determind from several figures presented in Preller (1995). In Table 2.1 of Preller (1995) the following average concentrations in stables are noted, which can be transformed into the regular stable emissions from Dutch pig farms under assumed ventilation rates:

respirable PM 0.15 mg/m^3 leads to an emission of 0.8 ktonnes/y
TSP 4.0 mg/m^3leads to an emission of 21.2 ktonnes/y
ammonia 4.4 mg/m^3 leads to an emission of 23.3 ktonnes/y

The ammonia emissions from pig breeding in the Netherlands as estimated above, are underestimated when compared with the 33 ktonnes/y determined by LLO (1995) to be the yearly emission from this source. This indicates that also the PM emissions estimated above will be more likely to be underestimated than overestimated. This leads to an extra contribution of rather coarse aerosol in the neighbourhood of pig farms and a skewed size distribution of the total rural aerosol in pig farming areas in the Netherlands.

This example only relates to pig breeding; there are other rural activities, for instance, poultry breeding, harrowing and ploughing, etc., and natural processes such as wind blown crustal material, which could also lead to a local and skewed aerosol distribution and influence the local PM_{10} levels. Van der Wal and Janssen (1996) estimated a local increase of PM_{10} concentrations due to agricultural emissions of at least 6 $\mu g/m^3$. A range of 5-8 $\mu g/m^3$ from local contributions to PM_{10} concentrations due to agricultural activities does not seem unreasonable at this stage.

4.2.3. Large-scale combustion

Because of the burning of natural gas in Dutch utilities and the use of efficient emission reduction devices, notably the desulpherisation in coal-fired power stations, no noticeable contribution of large-scale combustion to PM_{10} levels in the Netherlands is assumed.

4.2.4. Industry

Fine-particle emission of Dutch industry is assumed to be about 10 ktonnes/y. An average contribution of industry to PM_{10} levels in the Netherlands of 0-1 $\mu g/m^3$ is calculated using the simple box model (see appendix A). However, industrial emissions of the coarser part of PM_{10}, i.e. > 2.5 μm, may contribute to elevated PM_{10} levels on a smaller scale than the national one. Locally, on a scale of about 35 km, an extra contribution of 6 - 8 $\mu g/m^3$ may be assumed, based on measurements of PM_{10} in a industrialized area near a steel industry in the Netherlands (van der Wal and Janssen, 1996) and model calculations (Van den Hout, personal communication)

4.2.5. Households

Households are assumed to contribute to PM_{10} levels due to burning of wood in fire places. The carbonaceous particles are assumed to be in the size range of 2.5 < μm. The total emission is estimated at 7.5 ktonnes/y, so a contribution by consumers of 0 - 1 $\mu g/m^3$ does not seem unreasonable (appendix A).

4.3. Emittors of secondary PM_{10} precursors in the Netherlands

4.3.1. Contribution of different source categories to secondary aerosols

If, as assumed in Table 5, about 10 % of the PM_{10} is secondary aerosol originating from national sources and an average PM_{10} concentration of about 40 $\mu g/m^3$ is measured, an average contribution of about 4 $\mu g/m^3$ due to secondary aerosol from sources in the Netherlands is estimated.

In order to break this contribution down into the various source categories, the ratio of the contribution to the precursor gases of the different source categories is taken to be representative of the gases contribution to Dutch secondary aerosol levels. For industry this will probably lead to an overestimation because emission heights for SO_2 are so high; as there are no better estimates this will be used. For 1993 the SO_2 contribution from agriculture is: 0%, industry 57%, energy 16%, traffic 18% and consumers 0%. For NO_x the contribution from agriculture is: 2%, industry 16%, energy 12%, traffic 57% and consumers 4%. For NH_3 the contribution from agriculture is: 94%, industry 1%, energy 0%, traffic 0% and consumers 5%. Taking into account the relative of occurrence of sulphates, nitrates and ammonia in the Netherlands, the following contributions can be estimated.

Table 9 *Dutch contribution to secondary aerosol*

Dutch contribution to secondary aerosol	
agriculture	0.8 µg/m^3
industry	1.1 µg/m^3
energy	0.4 µg/m^3
traffic	1.2 µg/m^3
consumers	0.1 µg/m^3

4.3.2. Various other sources

No contribution of "other" unidentified sources is assumed in this analysis.

4.4. Foreign sources influencing PM$_{10}$ in the Netherlands

4.4.1. Traffic

If it is assumed that the contribution of traffic to secundary aerosol is about 30 % and that approximately 25 % of secundary PM$_{10}$ is supposed to originate from foreign sources (Table 6) a contribution of about 3 µg/m^3 of foreign traffic to the secundary aerosol in the Netherlands may be estimated. Small carbonaceous particles (<2.5 µm) come mainly from traffic, and because of the analogy with Pb as a traffic tracer, the contribution of carbonaceous to the yearly average PM$_{10}$ levels of 40 µg/m^3 in the Netherlands is estimated to be about 3 µg/m^3 as previously explained.

4.4.2. Other foreign sources

If a foreign contribution to PM$_{10}$ levels of 58% is assumed (Table 5) then a total contribution of ~24 µg/m^3 may be calculated using an average PM$_{10}$ concentration of 40 µg/m^3 . If the foreign traffic contribution is ~5.7 µg/m^3 then the contribution of "other foreign" sources to PM$_{10}$ levels in the Netherlands is about 18 µg/m^3, which may be broken down into 8 µg/m^3 of secondary aerosol and ~5 µg/m^3 of non-carbonaceous < 2.5 µm and a similar amount > 2.5 µm.

5. REGIONAL DISTRIBUTION OF SOURCES IN THE NETHERLANDS

The Netherlands is divided into different areas (described under 5.2) to compare the calculated contribution of the different emission categories as defined in Chapter 4 with the measured PM_{10} levels in the Netherlands. This spatial classification has been based on an estimation of the influence of different sources to the measured PM_{10} concentrations in the Netherlands, notably:

- a contribution of the large-scale background
- a contribution of local sources, mainly consisting of the coarser fraction of PM_{10} (> 2.5 µm).

5.1. Concentration gradients

Table 10 presents the residence time based on the dry deposition velocity as a function of particle size (aerodynamic diameter in µm, Van Jaarsveld, 1995). If it is assumed that particles are homogeneously distributed up to a height of 1000 m and the mean wind speed is assumed to be 6 m/s, which is a typical value that is used in the modelling to calculate the yearly average concentrations of air pollution in the Netherlands, then the approximate transport distance can be calculated if the mean residence times have been calculated when both dry and wet deposition have been accomodated.

Table 10 *Mean residence time (h) and approximate transport distance (km) for different aerodynamic particle sizes of aerosols for Western-European conditions*

Particle diam. (µm)	residence time (h)	transport distance (km)
<1	60	1200
1-4	10	200
4-10	8	160
10-20	6	120
>20	2	40

From the numbers in Table 10 one may conclude that sources which emit PM_{10} in the coarser size fraction (> 2.5 µm) can influence local PM_{10} levels at ranges up to ~ 200 km.

5.2. Characterics of the different areas influencing the PM_{10} levels

1. Background levels in the western part of the Netherlands

- long-range transport of emissions from foreign countries
- a limited contribution from national sources
- a larger influence of sea spray
- a contribution from resuspended crustal material.

2. Background levels in the eastern part of the Netherlands

- a stronger influence by foreign sources notably the coarser part of PM_{10}
- a limited contribution from national sources
- a limited contribution from sea spray
- a contribution from resuspended crustal material

3. An urban area in the centre of the Netherlands

- an average (between east and west of the Netherlands) contribution from foreign sources
- a substantial influence of local sources, notably traffic
- an average contribution of sea spray

4. An industrialised area in the western part of the Netherlands

- long-range transport of emissions from foreign countries
- sea spray
- a substantial contribution of local industrial sources, notably to the coarse fraction of PM_{10}

5. An agricultural area in the south eastern part of the Netherlands

- long-range transport of emissions from foreign countries
- a substantial influence of local agricultural sources
- a limited contribution of sea spray
- a contribution from resuspended crustal material

6. INTEGRATION

6.1. Integration of the contribution by source category and regional characteristics

The characteristics of the different source areas as described in Chapter 4, together with the calculated contributions of the different source categories of Chapter 3, are used to describe the regional PM_{10} composition of the Netherlands. The tables for the different areas present the yearly average contribution in µg/m^3 per source category.
In the following tables the yearly average contribution in µg/m^3 of the total secundary aerosol in the size ranges <2.5 µm and > 2.5 µm by the various source categories is presented under the heading: Sec. The column Carb. respresents the contribution to the carbonaceous aerosol by combustion sources, all in the size fraction < 2.5 µm. The symbols <2.5 µm and > 2.5 µm stand for the contribution of primary PM_{10} in the indicated two size ranges.

Background west Netherlands	Sec.	Carb	< 2.5	>2.5		PM_{10}
resuspended crustal material				1-3		1-3
sea spray				5-7		5-7
agriculture	1					1
industry	1-2	0-1		1-2		2-5
energy	0-1					0-1
traffic	1-2	1-2		1-2		3-6
consumers						
FOREIGN traffic	3-4	2-3				5-7
FOREIGN other sources	6					6
total	**12-15**	**3-6**		**8-14**		**23-35**

Background east Netherlands	Sec.	Carb	< 2.5	>2.5		PM_{10}
resuspended crustal material				1-3		1-3
sea spray				2-3		2-3
agriculture	1					1
industry	1-2	0-1		1-2		2-5
energy	0-1					0-1
traffic	1-2	1-2		1-2		3-6
consumers						
FOREIGN traffic	3-4	2-3				5-7
FOREIGN other sources	10		1-3	1-3		12-16
total	**16-19**	**3-6**	**1-3**	**6-13**		**26-43**

Urban area central Netherlands	Sec.	Carb	< 2.5	>2.5		PM_{10}
resuspended crustal material				0-2		0-2
sea spray				4-5		4-5
agriculture	1					1
industry	1-2	0-1		1-3		2-5
energy	0-1					0-1
traffic	1-2	4-6		2-3		7-11
consumers		0-1				0-1
FOREIGN traffic	3-4	2-3				5-7
FOREIGN other sources	8		1-2	1-2		10-12
total	**14-18**	**6-11**	**1-2**	**8-15**		**29-46**

Industrial west Netherlands	Sec.	Carb	< 2.5	>2.5		PM_{10}
resuspended crustal material				1-3		1-3
sea spray				5-7		5-7
agriculture	1					1
industry	1-2	0-1		6-8		7-11
energy	0-1					0-1
traffic	1-2	1-2		1-2		3-6
consumers						
FOREIGN traffic	3-4	2-3				5-7
FOREIGN other sources	6					6
total	**12-15**	**3-6**		**13-20**		**28-41**

Agricultural SE Netherlands	Sec.	Carb	< 2.5	>2.5		PM_{10}
resuspended crustal material				1-3		1-3
sea spray				2-3		2-3
agriculture	2			5-8		7-10
industry	1-2	0-1				1-3
energy	0-1					0-1
traffic	1-2	1-2		1-2		3-6
consumers						
FOREIGN traffic	3-4	2-3				5-7
FOREIGN other sources	10		1-3	1-3		12-16
total	**17-21**	**3-6**	**1-3**	**10-19**		**31-51**

6.2. Comparison of the regional concentrations with measurements and discussion

The background level in the west of the Netherlands can be compared with a station such as the Zilk or Wieringerwerf. The Zilk has only been operational for a year and a half so the reference year of 1993 is not included in its measurements. Wieringerwerf had a yearly average concentration of 32 $\mu g/m^3$ in 1993, which compares fairly well with the 23-35 $\mu g/m^3$ estimated on the basis of the integration.

The background level in the east of the Netherlands can be compared with a station such as Eibergen or Wageningen. Wageningen had a yearly average concentration of 43 $\mu g/m^3$ in 1993 and Eibergen had a concentration of 37 $\mu g/m^3$. This compares fairly well with the 26-43 $\mu g/m^3$ estimated on the basis of the integration.

An urban station can best be compared with a station in Utrecht. There is a measuring station in Utrecht, but this is not an urban background one, but a street station, so concentrations tend to be higher at such a station. The yearly average concentration in Utrecht was 43 $\mu g/m^3$ in 1993. An urban background station can be found in Amsterdam-Noord, but this is nearer to the coast than the centre of the country. The yearly average concentration at this station was 41 $\mu g/m^3$ in 1993. The estimated concentrations for an urban area in the centre of the country would be 29-46 $\mu g/m^3$, which is in reasonable agreement with measured ones.

There are no measuring stations of the National Air Quality Monitoring network near an industrial area. So a real comparison with the estimations is not possible. Amsterdam-Noord is not too far away (~ 30 km) from a big Dutch steel mill, but it cannot be called a typical industrial station.

An agricultural area in the southeast of the country can be compared with a measuring station such as Vredepeel. The yearly average concentration of PM_{10} measured in 1993 at this station was 49 $\mu g/m^3$. This seems to be in the range of 31-51 $\mu g/m^3$ estimated in 6.1.

In 6.1 some simplifications have been made which could have been accounted for. For instance, the fraction of primary particulate matter (< 2.5 μm) seems to be missing. This of course is not the case, but as a matter of simplifcation resuspended crustal material and sea spray is all grouped into the category > 2.5 μm. Real measurements (see Erisman et al., 1996) of size-dependent chemical fractions of PM indicate that a portion of 20-30 % of the particulate matter of these categories will be in the smaller sized range. If the tables in 6.1 then had been filled with such size fractions the simplicity of the exercise would have been violated. Therefore the results of tables in 6.1 should only be used for comparison with total PM_{10} numbers. Neither has the secondary aerosol been divided into > 2.5 μm and < 2.5 μm.

7. CONCLUDING REMARKS AND RECOMMENDATIONS

It can be concluded from these calculations that the two postulates of 20% Dutch and 80% foreign contributions to PM_{10} levels and a local contribution of 15% of primary PM_{10} by Dutch sources (Annema et al.,1996) contradict with each other. Therefore at least one of the two has to be incorrect.
As a way of hypothesis generation for the distribution of the contributions by local Dutch and foreign sources a postulate of 40 to 45% of local Dutch contribution is generated. This postulate is not in contradiction with the seven rules of thumb that have been formulated to include the existing knowlegde concerning PM in the Netherlands. Further expermental research into the validity of these new postulates is recommended as such postulates might indicate a different emphasis on control options and policy advise than the 20-80 postulate.

The current knowledge on Dutch primary aerosol emissions (Luchtkwaliteit, Jaaroverzicht 1993) of 53 ktonnes/y, leads to an average yearly (rural) background concentration of 2-3 $\mu g/m^3$, as can be calculated with the box model. This means that not all Dutch primary emissions are as yet reliably known, (see also 4.2). As this box model does not contain any sinks (no dry or wet deposition), the estimated concentrations with the simple model are more likely to be overestimates than underestimates. If approximately 40% of the PM_{10} levels is supposed to be contributed by local Dutch sources, some emissions are implied to be still missing. Therefore research into this problem and the influence of water on the missing mass is recommended.

Considering the “homogeneous PM blanket” covering the Netherlands (as virtually all different stations report yearly average concentrations of PM_{10} of 35 - 45 $\mu g/m^3$) and the missing mass of emissions to explain these measurements, it probably would be better to consider this “blanket” as a “patchwork quilt”. The different patches of the “quilt” are filled up by different local sources, stationary or non-stationary, or regular or irregular, which mainly emit PM_{10} in the coarser range (> 2.5 μm) of the size spectrum with a short approximate transport distance, as is shown in Chapter 6. Because of the rather even geographical distribution of these sources over a so densely populated country as the Netherlands and their shear numbers, a rather homogeneous looking “patchwork quilt” of PM_{10} seems to cover the Netherlands.

A recommendation of this, to a certain extent speculative and theoretical, exercise is, that more and reliable data based on measurements are needed. In particular more has to be known of the yearly (and episodic) size distribution and chemical speciation of PM in rural, urban and street settings, as well as the carbonaceous content (EC + OC) of PM and its size distribution in different parts of the Netherlands and the other constituents of PM_{10}. More insight into the size distribution of different parts of the secondary aerosol (SO_4, NO_3, and NH_4 and possibly also the organic fraction of the secondary aerosol) would be helpful to establish the Dutch and foreign contributions to the different fractions of the secondary aerosol. For source apportionment and other techniques to fill in the picture for sources of an intermittant and non-stationary nature, PM should be characterised with the use of suitable tracers.

Concerning the measurement techniques and the interpretation of their results, the skewness of the aerosol distribution, together with a size dependent sample efficiency of the sampling head, influence the reported levels of PM_{10}, as not only 50% of the particles of 10 μm diameter are sampled, but also some bigger particles of 15 or 20 μm, even larger ones may be sampled depending on the skewness of the aerosol, although with a very much reduced, but not zero, efficiency. Especially a situation with a very skewed size distribution of large particles (diam. > 20 μm), e.g. those generated mechanically or by animals, may influence the local PM_{10} readings. In such a case it may happen that the aerosol concentrations with an aerodynamical diameter less then 10 μm may seem to be overestimated. Knowledge of the total aerosol size distribution is necessary to investigate and quantify this effect, which may influence the size of the health risk if this risk is estimated by using the PM_{10} levels as a proxy for exposure.

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APPENDIX A BOX MODEL

In order to make a first-order approximation a simple box model will be used to describe the dispersion of concentrations on a yearly and a national basis for the Netherlands . Such a model, though with a circular surface instead of a square, has been used previously in the Netherlands to calculate, as a first approximation, the influence of foreign emissions on rural concentrations of benzene (Slooff, 1988).

In this model a yearly average effective mixing height (H) of 500 m is assumed for Netherlands and surrounding countries. The Netherlands is densely populated and emissions can be assumed to be fairly evenly distributed. The effective mixing height is also assumed constant over the year and the time of day, and wind speeds and concentrations are assumed constant with height.
The Netherlands is represented as a square with sides (L) of 200 x 200 km. The effective mean windspeed (u) in the Netherlands used in the OPS model is 6 m/s (Van Jaarsveld, personal communication). The stationary situation can be described by:

$dC/dt=0$

and C the averaged concentration in the box

Equating the emission flux: S (kg/s) and
the outward flux of the mass through the boundaries of the box $C*L*H*u$ (kg/s)
gives:

$S = C*L*H*u$ and

$C = S / (L*H*u)$ (1)

The yearly average concentration in the box (the rural background in the Netherlands) can be easily calculated using equation (1).

With this simple model an emission of 20ktonnes/y contributes approximately 1 µg/m^3 to the average (rural) background concentration.

APPENDIX B RELATIVE VARIANCE OF COARSE AEROSOL

The concentration of coarse aerosol (PM between the two 50% cut-off diameters of 10 and 2.5 µm) is often "established" by substracting the measured $PM_{2.5}$ concentration from that of PM_{10}. In principle this calculation leads to the correct averages, when a large number of individual measurments are used.
In practice, though, due to the low signal to noise ratio in the individual PM measurements such a calculation leads to a large relative variance of the concentrations of coarse aerosol. When the health effects are established with the individual (24 h average) concentrations, as usually is the case in time series analysis, this may lead to spurious results for the coarse aerosol.

Statistically speaking such a large noise in the coarse PM will in its turn lead to lower associations between PM concentrations and health effects, epidemiologicaly speaking this leads to "lower" betas in univariate analysis[2]. Therefore it is difficult to disentangle this statistical effect from the possibly "lower" patho-physiological nature of coarse PM also resulting in "lower" betas.

An example may clarify the described problem.

If: $Z = A - B$

than the variance of Z is: $\Delta Z^2 = \Delta A^2 + \Delta B^2$

Assuming that a certain measurement is 10 µg/m^3 PM_{10} and 6 µg/m^3 $PM_{2.5}$, which is a PM_{10} to $PM_{2.5}$ ratio generally observed in the Netherlands, and also assuming that the measurement error is 25% for the individual 24h averaged PM measurements, the following picture arises:

	standard deviation as % of measurement
$PM_{10} = 10 \pm 2.5$ µg/m^3	25%
$PM_{2.5} = 6 \pm 1.5$ µg/m^3	25%
coarse = 4 ± 2.9 µg/m^3	73%

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[2] Multivariate analysis leads to different results

This example indicates that the relative standard deviation of the coarse aerosol gets much larger (a factor of ~ 3) than the original basic values of the example. As the ratios and measurement errors presented in this example are not unreasonable compared to generally observed values in the Netherlands, the problem with coarse PM is probably represented correctly.

When the coarse fraction is sampled directly with a dichotomous sampler the problem described above does not arise, because the nature of such measurements is different from calculations.