Mineral Dust as a component of particulate matter

This is a publication of the Netherlands Research Program on Particulate Matter

PBL Netherlands Environmental Assessment Agency

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TNO

BOP – WP2 – report

Mineral Dust as a component of Particulate Matter

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PBL Netherlands Environmental Assessment Agency

ECN
Energy research Centre of the Netherlands

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National Institute
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ECN Energy research Centre of the Netherlands
PBL Netherlands Environmental Assessment Agency
TNO Built Environment and Geosciences
RIVM National Institute for Public Health and the Environment

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Rapport in het kort

Atmosferisch fijn stof (PM10) bevat in Europa gemiddeld 5-20% bodemstof. Deze bodemstofdeeltjes kleiner dan 10 micron komen uit verschillende bronnen zoals wind erosie, landbewerking en resuspensie door wegverkeer. Hoewel de bijdrage van bodemstof aan totaal PM10 substantieel is, is de kennis van herkomst en het relatieve belang van de verschillende emissieoorzaken zeer beperkt. In dit rapport wordt met behulp van chemische analyses van fijn stof monsters uit Nederland, waarnemingen uit ons omringende landen en de rest van Europa, alsmede eenvoudige emissiemodules en een regionaal atmosferisch transport model de rol van bodemstof als fijn stof component onderzocht en gekwantificeerd. Het onderzoek levert inzicht in de bijdrage van verschillende processen en locatie afhankelijke bijdrages van bodemstof. De verhoging van het bodemstofgehalte in fijn stof in de stedelijke omgeving is nog niet goed begrepen, evenals de verschillen tussen Nederland en omringende landen. Een belangrijke conclusie van het BOP onderzoek is dat de rol van bodemstof in Nederland beduidend geringer lijkt dan tot nu toe aangenomen.

Contents

Summary

Mineral dust (MD), or soil particles, typically make up 5 to 20% of the mass of ambient PM_{10}. In certain regions and/or under specific meteorological conditions, contribution may even be higher. Mineral dust may originate from distinctly different sources, such as wind erosion of bare soils, agricultural land management, driving on unpaved roads, resuspension of road dust, road wear, handling of materials, and building and construction activities. Despite the importance of mineral dust in total PM_{10} mass, the sources are still poorly understood and not well represented in emission inventories. This is due to the fact that some sources can be defined as natural sources (e.g., wind erosion) while others, such as resuspension, are not recognised as primary emission sources, but are reemissions of already present particles. Separating the source contributions is difficult, because the unique tracers for mineral dust in PM_{10} samples (Al, Si, Ti) do not allow a distinction between the various sources of this material. Because of these complications, mineral dust is a big 'unknown' in the understanding of ambient PM_{10}. Often, the gap between observed PM concentrations and model-predicted PM concentrations is attributed to 'mineral dust and other semi-natural sources' that are not represented in the emission inventories.

To quantify and understand the role of mineral dust as a PM source, a combination is needed of flux measurements, emission estimates, chemical analyses of sampled ambient PM_{10} and atmospheric transport modelling. This report works towards a methodology to elucidate the role of mineral dust;

1. During 2007 and 2008, samples of ambient PM_{10} and $PM_{2.5}$, collected at various types of locations in the Netherlands, were analysed for chemical composition. Based on the aluminium and silica content in these samples, an approximation was made of the amount of mineral dust in ambient PM_{10}.
2. The thus estimated mineral dust contribution was compared with observations from neighbouring countries (Belgium, Germany) and with results from a similar campaign in the Netherlands, which was held 10 years earlier.
3. A European-wide literature survey and data collection activity was initiated to construct a database on mineral dust in PM_{10} across Europe.
4. Simple and therefore transparent emission functions were combined with activity data or land-use maps to make emission grids. The gridded emissions were then used as input for the LOTOS-EUROS model to calculate the resulting concentrations.
5. The predicted concentrations were compared with observations in various parts of Europe, derived from the literature survey.
6. A first correction of the emission functions was made to improve the match between modelled and observed concentrations.

The various steps outlined above, together, form a coherent approach, but are described individually in this report. Our understanding and representation of mineral dust in the air quality model is not yet complete. Further iteration, by comparing observed and modelled values, would be needed. However, from the work done so far, some important conclusions could be drawn.

Based on the chemical speciation data that were collected at six locations in the Netherlands, as part of the BOP programme, the contribution of mineral dust to PM_{10} in the Netherlands, in the 2007-2008 period, is estimated at no more than ~ 1.5 µg/m³. This is significantly lower than previously observed for 1998 and 1999 by Visser *et al.* (2001), and, more recently, for 2006, in Flanders, Belgium (VMM, 2009). In the neighbouring German province of North Rhine-Westphalia, data were collected by Quass *et al.* (2009) in a partly overlapping period with the BOP campaign. Again, the mineral dust estimates were higher than observed in the BOP campaign, but the gap between them was smaller. This discrepancy between estimates for neighbouring locations, as well as different years, showed that substantial variations in mineral dust contributions may exist. Unfortunately, chemical speciation of PM, including mineral dust tracers, is only obtained in specific campaigns and no good record for temporal variation is available. It is concluded that this variation in mineral dust is not yet fully understood and calls for a more in-depth comparison and collaboration between the research groups or agencies in the different countries.

The amount of mineral dust observed in PM_{10} during the BOP campaign in the 2007-2008 period showed an increase in MD in the urban environment and at traffic locations. This is consistent with the increase in MD across the European urban environment, as deduced from the compilation of European data. However, in the BOP data, the increment in MD from rural background to traffic locations was ~40 to 50%, whereas in a large number of European studies, MD in PM_{10} in urban or traffic locations was found to be roughly doubled. After verification it was concluded that the BOP data had correctly captured the increment at urban/traffic locations, with the surprising observation that this increase was not proportionally reflected in the mineral dust concentrations. This observation is quite remarkable, as it is contrary to the average observational data available on Europe, and contrary to what was expected for the Netherlands, based on the previous source apportionment study by Visser *et al*. (2001).

European data were compiled to estimate MD in PM_{10} for 19 European countries, often from more than one location per country. The compilation showed that MD in PM_{10} in urban centres ranged between 3 and 13 µg/m³ (with a few outliers), confirming the importance of MD in PM_{10}. Mineral dust in PM_{10} in the Netherlands appeared to be relatively low, compared to southern and northern Europe. These regions experience higher MD concentration levels due to a dryer climate (southern Europe) or the use of studded tires and road gritting in wintertime (northern Europe). In the context of this study, the Netherlands would rank with countries such as Belgium, Germany, and the United Kingdom.

To be able to model mineral dust in PM_{10}, specific modules have been developed for estimating MD emissions from wind-blown dust, agricultural land management and resuspension. These modules, or emission grids, have been implemented into the LOTOS-EUROS model. Explicit modelling of MD as a PM component is a novelty in Europe. The compiled European observational data on MD allowed a comparison with model-predicted MD and observed concentrations. This comparison was rather crude, since the observational data did not necessarily come from the same year or period that was covered by the model runs. However, the comparison did support some first modifications, which resulted in a better match between modelled and observed MD in PM_{10}. Average annual concentrations for a limited number of European rural locations were predicted with remarkable accuracy. However, a comparison between modelled concentrations and a larger set of observations indicated that current model approaches still underestimate total mineral dust contributions. The most remarkable feature of MD in PM_{10} remains its increase within the urban environment. Therefore, the importance of urban, inner-city processes for MD emission other than resuspension due to road traffic should be quantified in future studies.

The data from the BOP campaign showed that MD in the Netherlands is less important than previously thought. Since some contradiction with comparable, neighbouring countries appears present, the need for further monitoring of mineral dust in PM_{10} remains – at least until we understand why MD in the Netherlands is less important than elsewhere. Within the BOP project, we have been able to create a database of observational data and to make modules for modelling MD in PM_{10} with the LOTOS-EUROS model. A tool has been created to investigate the different source contributions, and is expected to enable further understanding by introducing the specific time of occurrence and spatial distribution for each source category. Based on the research done, recommendations for future research are presented.

Netherlands Research Program on Particulate Matter (BOP)

This study is conducted partly under the auspices of the Netherlands Research Program on Particulate Matter (BOP), a national programme on PM_{10} *and* $PM_{2.5}$*, funded by the Netherlands Ministry of Housing, Spatial planning and the Environment (VROM). The programme is a framework of cooperation, involving four Dutch institutes: the Energy Research Centre of the Netherlands (ECN), the Netherlands Environmental Assessment Agency (PBL), the Environment and Safety Division of the National Institute for Public Health and the Environment (RIVM) and TNO Built Environment and Geosciences.*

The goal of BOP is to reduce uncertainties about particulate matter (PM) and the number of policy dilemmas, which complicate development and implementation of adequate policy measures. Uncertainties concerning health aspects of PM are not explicitly addressed.

The approach for dealing with these objectives is through the integration of mass and composition measurements of PM_{10} *and* $PM_{2.5}$*, emission studies and model development. In addition, dedicated measurement campaigns have been conducted to research specific PM topics.*

The results of the BOP research programme are published in a special series of reports. The subjects in this series, in general terms, are: sea salt, mineral dust (this report), secondary inorganic aerosol, elemental and organic carbon (EC/OC), and mass closure and source apportionment. Some BOP reports concern specific PM topics: shipping emissions, PM trend, urban background, EC/OC emissions from traffic, and attainability of $PM_{2.5}$ *standards. Technical details of the research programme are condensed in two background documents; one on measurements and one on model developments. In addition, all results are combined in a special summary for policy makers.*

1

Introduction: Particulate matter and mineral dust

Particulate matter (PM) or aerosol is the generic term used for a type of air pollution that consists of complex and varying mixtures of particles suspended in the atmosphere. PM has been found to present a serious danger to human health. PM loadings in the Netherlands are relatively high, compared to other regions of Europe. Hence, there is a clear need to investigate the origin and source contributions for ambient PM in the Netherlands. An atmospheric transport model can be used to couple emissions of PM to observed PM concentrations. In the case of particulate matter, the modelled PM_{10} concentration (all particles < 10 µm) is the sum of the concentration of individual chemical species for which a source is defined in the model. Since most models do not include all PM sources and species, simulated concentrations are underestimated (by ~40-50%) compared to observed values (Schaap *et al.*, 2008). The non-modelled fraction of PM is often comprised of sea salt, mineral dust, secondary organic material and water. Within the BOP project, we aim to have a better understanding of the sources of aerosols in the Netherlands and greater Europe; the chemical composition of the aerosols, their concentrations, spatial distribution, temporal trends and ranking of importance. The current report focuses on suspended mineral dust (also called crustal material or soil dust) as it may contribute significantly to the total aerosol loading. Furthermore, mineral dust is thought to contribute significantly to the 'non-modelled' fraction of PM in the Netherlands (MNP, 2005). The majority of this so-called non-modelled fraction is grouped under 'soil dust and other', which is thought to be responsible for ~ 40% of the total ambient PM_{10} concentration in the Netherlands (MNP, 2005). This is more than two times the contribution from Dutch anthropogenic sources which contribute ~ 15% to ambient PM in the Netherlands (MNP, 2005). From these crude figures it becomes evident that uncertainty in the non-modelled fraction obscures the impact that policy measures can have on ambient PM concentrations, especially since it is unknown to what extent the category 'soil dust and other' varies between years or is influenced by measures. Therefore, one of the main ambitions of the BOP programme is to reduce the importance of the non-modelled fraction by explicitly adding (semi-)natural sources of PM, such as sea salt and soil dust to the predictive model. This can be done through source functions in the model that are driven, for instance, by meteorology or by providing gridded emissions as model input.

To achieve this ambition, a predefined strategy was followed (Figure 1-1). Because of encountered difficulties, the way towards a validated representation of mineral dust in the modelling of PM was adjusted several times, however, the outline given in Figure 1-1 remains valid.

1.1 Mineral dust

Particulate pollution comes from such diverse sources as power plants, engine exhausts, wood burning, mining, and agriculture. Because of the variation in sources, airborne PM includes many different chemical constituents. PM_{10} samples collected on a filter can be analysed for chemical composition. Sources contributing to PM_{10} concentrations may emit particles with a unique chemical composition that are enriched in certain elements.,. For example, vanadium and nickel have typically been used as tracers for emissions from heavy fuel-oil combustion. Similar tracers exist for mineral dust. Mineral Dust (MD), also called crustal material or soil dust, refers to atmospheric aerosols originating from minerals in soil or the earth's crust. It is mainly composed of oxides (such as SiO_2, Al_2O_3, FeO, Fe_2O_3, CaO[1]) and carbonates (such as $CaCO_3$, $MgCO_3$) (Figure 1-2). Of all the different other elements oxygen is present as oxide and is never directly measured in PM samples. Therefore, the dominant elements that can be used to quantify mineral dust contributions are silicon and aluminium (Figure 1-2). However, also calcium (Ca) and iron (Fe) are frequently used to estimate the mineral dust contribution to PM. The use of tracers for estimating mineral dust contributions is further discussed in Chapter 2.

1.2 Sources of mineral dust contributing to PM_{10}

Mineral dust may originate from distinctly varying sources. Dust aerosol particles produced by wind erosion in arid and semi-arid regions significantly affect climate and air quality, but the magnitude of these effects is largely unquantified. Major dust source regions include the Sahara, and the Arabian and Asian deserts. Global annual dust emissions are currently

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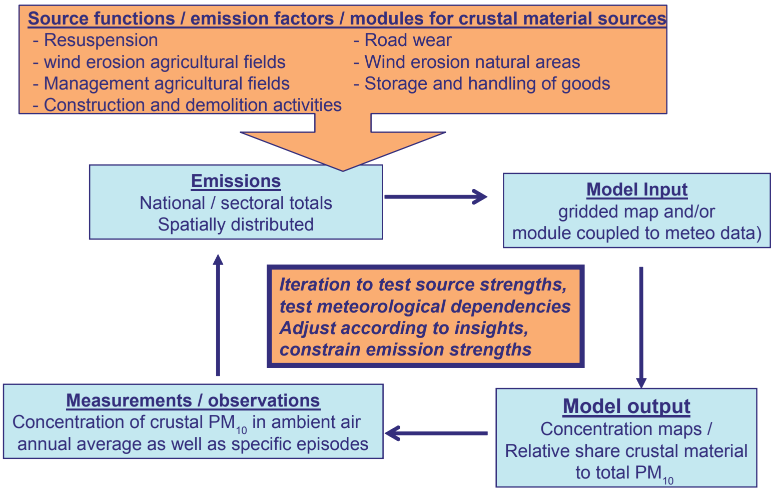
[1] SiO_2=silicon dioxide or quartz or sand; Al_2O_3=aluminium oxide, FeO=ferrous oxide; Fe_2O_3=ferric oxide, CaO=calcium oxide or lime; $CaCO_3$=calcium carbonate; $MgCO_3$=magnesium carbonate.

Strategy to understand and define the contribution and distribution of mineral dust

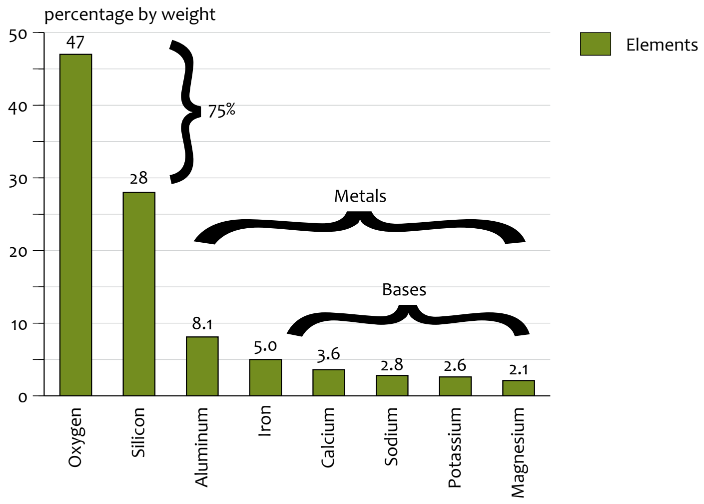
Figure 1.1



Strategy to understand and define the contribution and distribution of mineral dust (MD) in PM_{10} and $PM_{2.5}$ across Europe and the Netherlands.

The average composition of the Earth's crust

Figure 1.2



The average composition of the earth's crust in weight percentages of the eight most abundant elements (Source: Strahler and Strahler, 1992).

estimated to range between 1000 and 3000 Mt/yr (Tegen and Schepanski, 2009). Next to wind erosion, which is partly anthropogenic because of human-induced desertification, various other human activities may cause mineral dust to enter the atmosphere. Despite the importance of mineral dust in total PM_{10} mass, the sources are still poorly understood..

On the global scale, mineral dust emissions are dominated by desert dust. However, in Europe, other sources dominate mineral dust contributions. The sources are (see also Figure 1) :

- Wind-blown dust
- Agricultural activities
- Construction and demolition
- Handling of mineral goods
- Road wear
- Road gritting and use of studded tyres
- Driving over unpaved roads
- Resuspension due to road traffic

Several of these sources, such as road gritting and the use of studded tyres, have a distinct regional pattern. The sources are discussed in detail in Appendix 1 and partly in Chapter 4, where source functions are introduced that are incorporated in the LOTOS-EUROS model to facilitate the modelling of mineral dust < 10 µm.

Average annual levels (µgm^{-3}) of PM10, $PM_{2.5}$, mineral elements, and the equivalent contributions to bulk mass concentrations (% wt), recorded at regional background (RB), urban background (UB) and traffic locations (TS) in central Europe (examples from Austria, Germany, Switzerland, the Netherlands, the United Kingdom), northern Europe (13 locations in Sweden) and southern Europe (10 locations in Spain). **Table 1.1**

	Central Europe			**Northern Europe**			**Southern Europe**		
	RB	UB	TS	RB	UB	TS	RB	UB	TS
PM_{10} (µgm^{-3})	14–24	24–38	30–53	8–16	17–23	26–51	14–21	31–42	45–55
Mineral matter (µgm^{-3})	1–2	3–5	4–8	2–4	7–9	17–36	4–8	8–12	10–18
% Mineral matter PM_{10}	5–10	10–15	12–15	20–30	35–45	65–70	12–40	25–30	25–37
$PM_{2.5}$ (µgm^{-3})	12–20	16–30	22–39	7–13	8–15	13–19	12–16	19–25	28–35
Mineral matter (µgm^{-3})	0.5–2	0.4–2	1–2	1–3	2–4	4–6	1–3	2–5	4–6
% Mineral matter $PM_{2.5}$	2–8	2–8	5	15–25	25–30	30–40	8–20	10–20	10–15

Source: Querol *et al.* (2004)

1.3 Mineral dust as a fraction of total Particulate Matter

Based on the concentrations of tracers, such as Si, Al and Ca, the total amount of mineral dust in ambient PM_{10} can be estimated. In Europe, mineral dust or soil particles typically contribute 5 to 20% to ambient PM_{10} mass (Table 1-1). In certain regions and under specific meteorological conditions contributions may be higher. Examples of such region-specific conditions are the use of studded tires in Nordic countries and the intrusion of Sahara dust into the atmosphere of southern Europe, causing elevated crustal contributions to PM_{10} (Table 1-1). Despite the importance of mineral dust in total PM_{10} mass, the sources are still poorly understood and not (well) represented in emission inventories or air quality models. However, a better understanding of MD contribution to PM is essential for understanding contributions from various sources to the total PM burden, and because different components of PM may have different health impacts.

An estimate of the contribution of mineral dust to PM_{10} in the Netherlands based on the BOP measurement campaign is described in the next chapter. These results were compared with previous Dutch data (Chapter 2) and with data from neighbouring countries Belgium and Germany (Chapter 3).

The modelling of mineral dust in PM_{10} for Europe starts by making first-order estimates of the various source strengths of mineral dust sources (Figure 1-1). Simple and therefore transparent emission functions are proposed for the processes that are assumed to contribute the most in north-western Europe. These functions are implemented in the LOTOS-EUROS model (Chapter 4). A review of observational European PM_{10} and derived mineral dust contributions is presented in Chapter 5. The thus obtained European data set is needed to validate the LOTOS-EUROS model . Through the collected data we estimated the share of mineral dust in the total aerosol burden for the Netherlands and Europe. The results from modelling PM concentrations with these new source functions are described in Chapter 6. The final conclusive chapter puts the results in perspective and provides suggestions for further improvements (Chapter 7).

Mineral dust in PM_{10} in the Netherlands

2

The Netherlands Research Program on Particulate Matter (BOP) is aimed to reduce gaps in our knowledge on composition, distribution and origin of particulate matter in the Netherlands. In the framework of the BOP programme an intensive, one-year measurement campaign from August 2007 to August 2008 was carried out. During this year, PM_{10} and $PM_{2.5}$ was sampled, simultaneously, every other day at five locations. For about half of the sampling days the filters were analysed for PM composition. The results from this campaign for mass closure and source apportionment have been reported by Schaap *et al.* (2010). Here, we have used the results from this measurement campaign to estimate the contribution of mineral dust to PM_{10}. Furthermore, a comparison was made between previous estimates on the contribution of mineral dust to PM_{10} in the Netherlands, taken from a measurement campaign in 1998-1999 by Visser *et al.* (2001).

2.1 Estimation of mineral dust

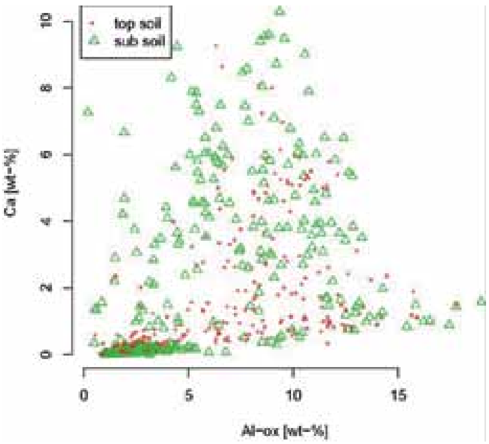
A number of elements are known to be present in mineral dust, the most important being Si, Al, Ca, K, Fe and Ti (Figure 1-2). Strong correlations between these elements in PM samples suggest they originate from the same source. Clay minerals in soil and rock have complex alumino-silicate structures (feldspars) and although measuring the aluminium and silicon content is relatively straightforward, the associated oxygen has to be estimated indirectly by assuming a certain oxidation state. So, elemental weight alone is not enough, but requires a correction for the oxidised form (e.g., Si is mostly present as SiO_2) to estimate total contribution of mineral dust to PM_{10}. Apart from Si and Al minerals, all kinds of other minerals may be present but contribute much less to total mineral dust, with the exception of areas dominated by calcareous soils where carbonates, such as $CaCO_3$, may be important. Since chemical analysis often provides all relevant elements, it is potentially possible to account for all elements present in mineral dust. After correction for the oxidation status this would then result in total MD mass. However, there are two major points of caution. First, a particular element may also (partly) originate from other sources, for example, Fe from wear of metal machinery, brakes and the iron and steel industry. For K, wood combustion and fertiliser manufacturing and application may contribute significantly. Assigning all mass to mineral dust could thus result in overestimation. Therefore, such elements are less suited to be used as tracers and their respective mass contribution needs to be estimated from the ratio to, for example, Si and Al in soil material. Second, some elements are really trace elements in soil material hence the low concentrations are may be close to detection limits introducing considerable uncertainty in the final estimate. Both points can be practically solved by focusing on the two most abundant elements (Si and Al) which are hardly present in PM from other sources. Nevertheless, a check on correlation between Si and Al is still crucial to confirm that both elements are most likely to originate from the same source. A further confirmation can be obtained for relatively high MD samples to also check for correlation with the trace element Ti, because in high MD samples the trace element is also accurately determined. A formula [3.1] was derived by Denier van der Gon *et al.*, 2007; 2008), based on the chemical composition of the fine fraction of Dutch top soils to calculate the contribution of mineral dust (MD) by using concentrations of Al and Si. The formula includes the average mass from other non-Al and non-Si minerals, and is used to estimate MD in PM_{10} from the chemical analysis of PM_{10} samples in the BOP programme.

$$\text{Mass MD} = 0.49*[\text{Si}] + (2.36*[\text{Si}]+2.70*[\text{Al}]) \qquad [2.1]$$

2.1.1 Calcium as a tracer for mineral dust in PM_{10}

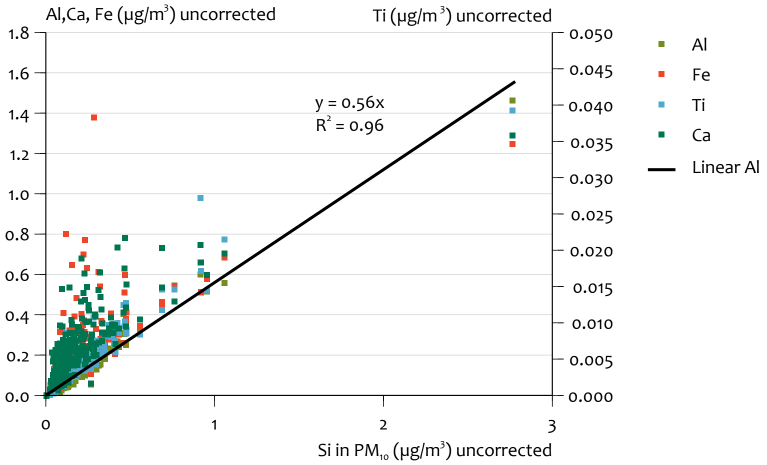
As an alternative for silicon and aluminium, calcium (Ca) is often used as a tracer for mineral dust. The main reason being that Ca is more often analysed in samples, is abundant in soils, and in PM samples often correlates well with Si and Al. Indeed, calcium is commonly present in soil, but in varying concentrations. For the Netherlands, the average Ca content in soil is 2 to 3% (Spijker *et al.*, 2008). For top soil, which is the most likely source of mineral dust entrainment into the atmosphere, there is no relation between aluminium (or silicon) content and calcium (Figure 2-1). The suitability of Ca as a tracer for mineral dust is further discussed in Annex 2, which mainly concludes that Ca often correlates with mineral dust content, but does not have a fixed correlation with Al and Si in soils. Hence, estimating soil dust in PM_{10} can be done by using empirically derived relationships between total soil dust and Ca concentration in PM_{10}, but not by using the average Ca content of soils. The last would imply

The relationship between Calcium and Aluminium in Dutch top soils and subsoils **Figure 2.1**



The relationship between calcium and aluminium in Dutch top soils and subsoils (Van der Veer; 2006; Spijker et al., 2008).

Al, Ca, Fe and Ti as a function of Si in PM_{10} for three rural stations in the Netherlands **Figure 2.2**



Al, Ca, Fe and Ti as a function of Si in PM_{10} for three rural locations in the Netherlands in 2007-2008.

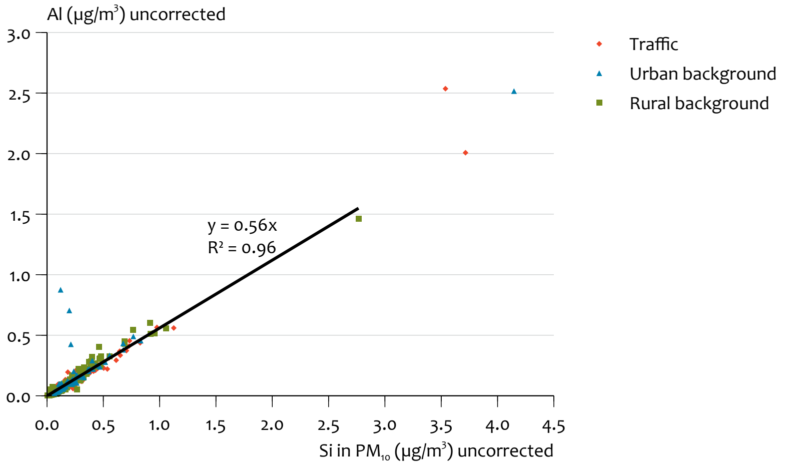
multiplication of the Ca concentration in PM_{10} by a factor of 20 to 50 (based on a 2-3% Ca content in soils), resulting in unrealistic or even impossible shares of soil dust to total PM. The empirical factors are more in the order of a factor 7, implying that Ca is significantly enriched by other sources than by soil material, for example, calcareous materials and concrete. Therefore, tracers of choice for mineral dust in the present study were Si and Al, together with further consistency checks using, for example, Ti and Ca.

2.2 Mineral dust in PM_{10} in the Netherlands in 2007-2008

In the BOP programme, a chemical analysis of PM_{10} samples collected over one year (2007-2008) was carried out to determine their chemical composition. For a detailed description of the methodology we refer to van Arkel *et al.* (2009) and Schaap *et al.* (2010). Based on the collected chemical data, we gathered information on the MD contribution to ambient PM_{10}. First, the correlation between the known tracer elements for MD was investigated (Figure 2-2). Even for the rural background locations the correlation for Fe and Ca with Si shows considerable scatter, suggesting that the elements did not necessarily originate from the same source. The correlation between Al, Ti and Si was very good ($r^2 = 0.96$). This supports the approach discussed earlier in this chapter, which considers that Al and Si are most reliable for estimating MD. Furthermore, the correlation between Al and Si for three different types of locations showed a consistent pattern (Figure 2-3). For comparison, the trend line from Figure 2-2 has also been presented in Figure 2-3. Only three outliers could be identified for the urban background location, the other ~500 data points showed a high correlation between Al and Si, consistent with the elements present in rather fixed ratios in soil material.

Al as a function of Si in PM_{10} for rural background, urban background and street locations

Figure 2.3



Al in PM_{10} as a function of Si content for rural background, urban background and traffic locations included in the BOP programme in 2007 to 2008.

Estimated mineral dust in PM_{10} in 2007-2008 for 5 Dutch locations

Table 2.1

Location	type	MD_uncorrected	MD_corrected[a)]
		μg/m³	μg/m³
Cabauw	rural	0.80	1.45
Hellendoorn	rural	0.65	1.18
Vredepeel	rural	0.87	1.57
Schiedam	urban background	1.26	2.25
Breda	traffic	1.06	1.92
Rotterdam	traffic	1.11	2.01

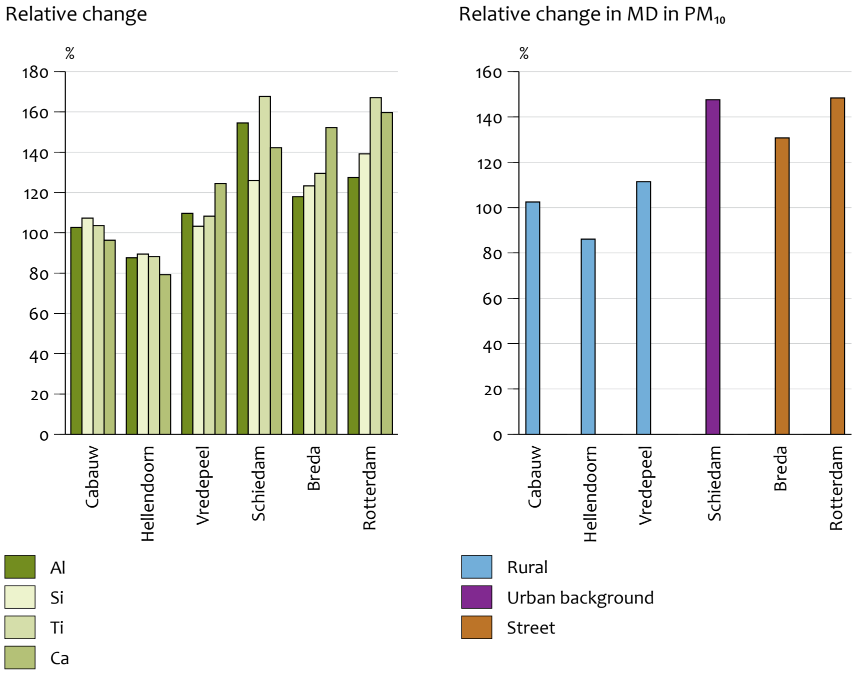
[a)] Correction implies that the Si and Al concentrations have been adjusted for a 50 and 70% recovery in the chemical analysis (see Van Arkel *et al.*, 2009; Schaap *et al.*, 2010).

Unfortunately, a complication in quantification of the MD component in the PM_{10} samples was the recovery of elements with the chosen destruction and analysis techniques. Van Arkel *et al.* (2009) have reported recoveries for silicon and aluminium of 50 and 70%, respectively. As shown in Figure 2-2 and Figure 2-3, the ratio between both elements is well-defined and remains intact from high to low concentrations and across different locations. Therefore, it is our conclusion that a simple across-the-board correction for the recovery is allowed. This correction was also applied to the same samples by Schaap *et al.* (2010) to achieve mass closure. The MD contribution to $PM_{10,}$ estimated by inserting the observed Al and Si concentrations (with and without recovery correction) in Equation 2.1 is presented in *Table 2-1*

The data show an increase in MD in PM_{10} in the urban background and traffic locations, compared to those in the rural background. The change in the most important tracer elements for MD, compared to the Dutch rural background locations (an average of results from Cabouw, Hellendoorn and Vredepeel) is presented in Figure 2-4. Although variation existed for the different elements, also between the rural locations, all elements showed an increase in traffic and urban background locations. Overall can be seen that, compared to the average rural background in the Netherlands, MD is ~ 45% elevated in urban background and traffic locations (Figure 2-4, right). This suggests that urban sources made an important contribution to the observed MD in PM_{10}, in line with observations elsewhere (see Table 1-1 and e.g., Querol *et al.*, 2004; Putaud *et al.*, 2004) . The most likely explanations for this increase are resuspension caused by road traffic and construction and demolition activities within the city.

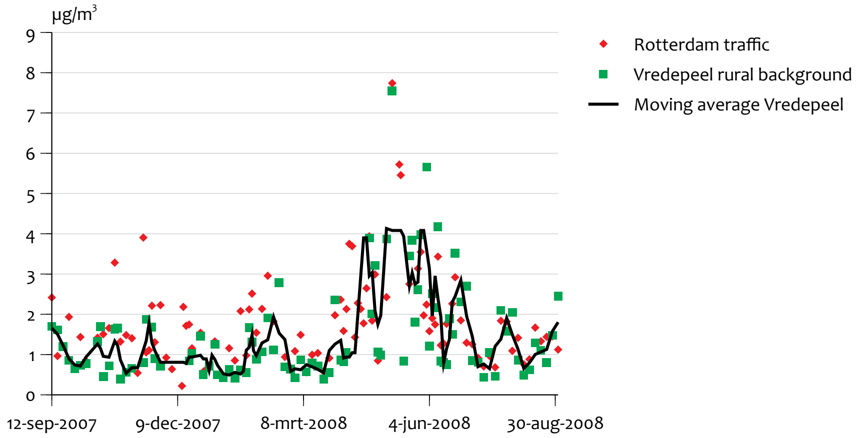
In line with observations elsewhere, the concentration of mineral dust in PM_{10} varied over time (Figure 2-5). Although average observed concentrations were between 1 and 2.5 μg/m³, during certain episodes the concentrations could go up to 5 to 10 μg/m³

Relative change of tracer elements and the estimated mineral dust in PM_{10} **Figure 2.4**



Relative presence of the tracer elements Al, Si, Ti and Ca (left) and estimated mineral dust in PM_{10}, (average of rural background taken from locations at Cabouw, Hellendoorn, Vredepeel = 100%).

Concentration of mineral dust in PM_{10} for a kerbside station and rural background station **Figure 2.5**



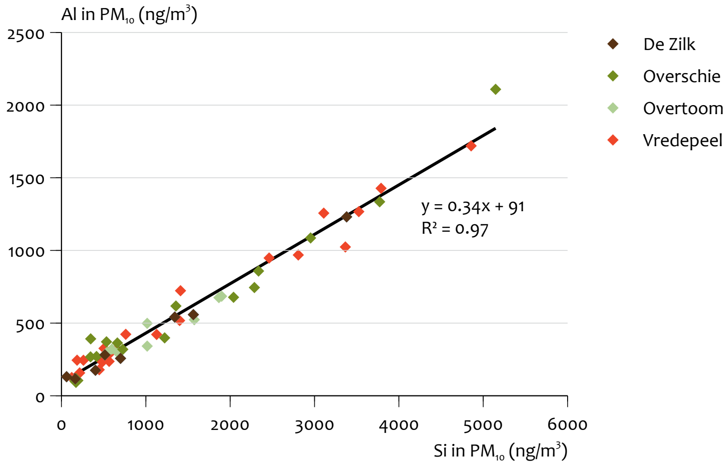
Concentration of mineral dust in PM_{10} at a traffic and a rural background location in 2007-2008, over time.

2.3 Mineral dust in ambient PM_{10} in the Netherlands in 1998-1999

The current BOP programme is not the first to investigate the chemical composition of airborne PM in the Netherlands. Samples of the fine fraction ($PM_{2.5}$) and the coarse fraction ($PM_{2.5\text{-}10}$) were collected from six locations in the Netherlands by Visser *et al.* (2001) during one year in 1998-1999. The samples were analysed for chemical composition; for a description of the measurement procedures and analytical specifications, we refer to Visser *et al.* (2001). In keeping with the data from the current BOP programme (Figure 2-2 and Figure 2-3), the data from Visser *et al.* (2001) show a strong correlation between Al and Si (Figure 2-6), indicating that Al and Si indeed are most likely to have the same origin. The number of days analysed in the study of Visser *et al.* (2001) with a complete elemental analysis of PM is rather limited. To bypass the problem of limited data availability, the strong correlation between Al and Si in the available data (Figure 2-6) was used to calculate soil material on the basis of Si alone, when no Al data were available (Denier van der Gon *et al.*, 2007) . The average concentration of MD in PM_{10} for the Netherlands in 1998-1999 was estimated at between ~2.5 µg/m³ at the coast to ~ 5.5 µg/m³ at inland locations (Table 2-2).

Al as a function of Si in PM_{10} at four locations in the Netherlands

Figure 2.6



Aluminium (Al) concentration as a function of silicon (Si) in PM_{10} at four locations in the Netherlands (based on data from Visser et al., 2001).

Average annual contribution of MD to ambient PM_{10} concentrations at four locations

Table 2.2

Location	**De Zilk**	**Vredepeel**	**Overschie**	**Overtoom**
method	**MD_alt**	**MD_alt**	**MD_alt**	**MD_alt**
No. of observations	31	45	42	49
Average (ug/m³)	2.4	5.4	5.0	3.3
Stdev	2.5	4.7	3.9	2.2

MD_alt = similar to MD eq[1], except for the fact that for the latter missing Al data are estimated based on the Al to Si correlation (Figure 2-6)

Mineral dust contribution to PM_{10} at De Zilk and Vredepeel at different wind directions

Table 2.3

	De Zilk			**Vredepeel**		
	All	**Continental**	**Sea**	**All**	**Continental**	**Sea**
Wind direction	0-360	(0-180)	(180-360)	0-360	(0-180, 315-360)	(180-315)
Observations	31	18	13	45	19	26
Average (ug/m³)	2.4	2.9	1.8	5.4	8.6	3.0
Stdev	2.5	3.1	1.4	4.7	4.8	2.9

Denier van der Gon *et al.* (2007) investigated the variation of the MD concentration with wind direction for the data from Visser *et al.* (2001. The samples were split into those measured while the wind direction brought air masses from the sea or from the continent (Table 2-3). Air masses with a more continental/land origin show an elevated mineral dust component, compared to air masses coming from the sea (2.9 vs 1.8 µg/m³ for De Zilk and 8.6 vs 3.0 µg/m³ for Vredepeel). This may help us to identify source regions, using trajectories in the models for particular days with elevated concentrations, and to investigate the possible dependency on meteorological conditions.

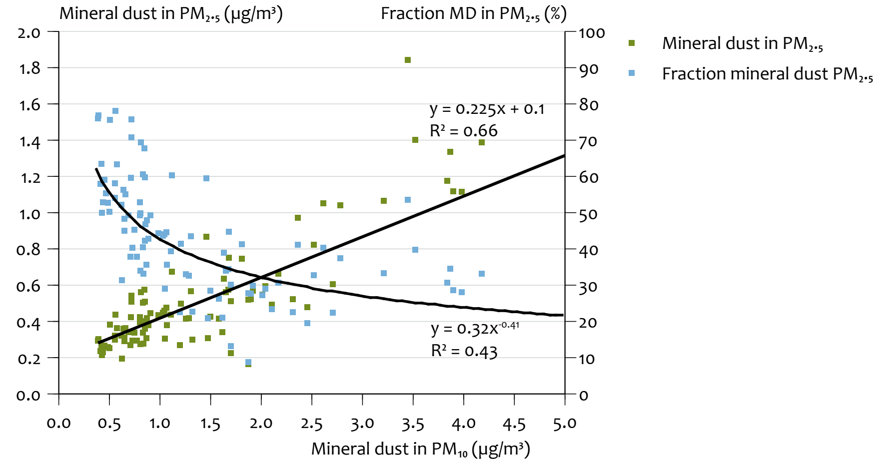
2.3.1 Size fractionation of mineral dust in PM_{10}

An important model feature, needed to predict the contribution of particulate emissions to ambient concentrations, is the size fractionation. The smaller size fractions are transported over longer distances and have a longer lifetime because of a slower deposition rate. The starting point, based on averaging observational data with chemical speciation in $PM_{2.5}$ and PM_{10}. was that 20% of MD is contained in the fine fraction ($PM_{2.5}$) of PM_{10}. This assumption can be checked and refined by taking a closer look at individual observational data for a particular location (instead of looking at the overall average).

As expected, the absolute amount of crustal $PM_{2.5}$ increases when the amount of crustal PM_{10} increases (Figure 2-7). However, the fraction of $PM_{2.5}$ in PM_{10} is not constant. When total MD in PM_{10} increases, the fraction of $PM_{2.5}$ decreases (Figure 2-7). A similar pattern was observed previously by Denier van der Gon *etal.* (2007) in the data available for 1998-1999 (Figure 2-8). When MD in PM_{10} is less than 0.5 µg/m³, the fraction $PM_{2.5}$ may be as high as 40 to 80%, but when amounts of MD in PM_{10} become substantial, for example, >2 µg/m³, the fraction of MD in $PM_{2.5}$ drops to ~20% (Figure 2-7). When higher contributions of MD to PM_{10} are observed (e.g., > 7 µg/m³) it drops further to ~ 10% (Figure 2-8). This pattern can be understood from the notion that if nearby sources dominate MD in PM_{10}, it is mostly in the coarse fraction, in contrast to MD being added from long-range transport – which is dominated by smaller particles. So, the more substantial the

Mineral dust in PM_{10}, in $PM_{2.5}$ and the fraction of $PM_{2.5}$ in mineral dust PM_{10} for a Dutch background station

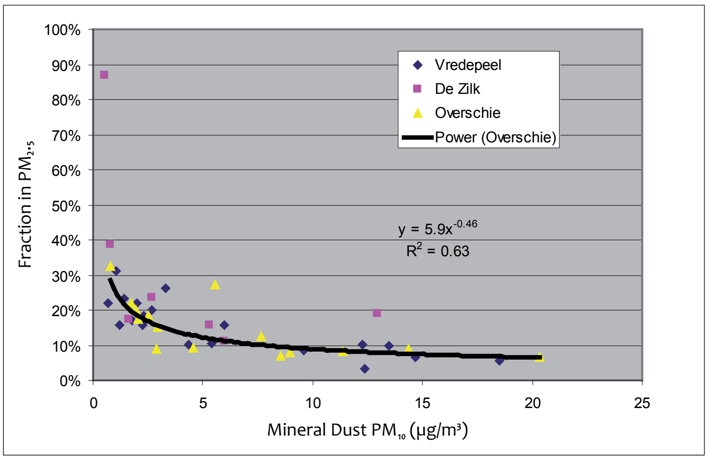
Figure 2.7



Mineral dust in PM_{10} and $PM_{2.5}$, and the fraction of $PM_{2.5}$ in mineral dust PM_{10}, at a Dutch background location, in 2007-2008.

The fraction of $PM_{2.5}$ in mineral dust PM_{10} for two Dutch background stations

Figure 2.8



The fraction of $PM_{2.5}$ in crustal PM_{10}, at two Dutch background locations, in 1998-1999.

MD contribution to PM_{10}, the less relevant the $PM_{2.5}$ fraction. The linear relationship between MD in $PM_{2.5}$ and PM_{10} shown in Figure 2-7 suggests that, overall, 22.5% of MD is in the fine fraction. From this we concluded that, in general, a generic fraction of 20% of MD in $PM_{2.5}$ would be a good estimate. Occurrences of strongly elevated MD contributions to PM_{10} will have been caused by relatively nearby sources. In such cases, the fine fraction does not increase proportionally and will be relatively unsubstantial. This observation may be relevant to a proper assessment of the health impact of such occurrences.

2.4 Comparison between mineral dust contribution to PM_{10} in 1998-1999 and 2007-2008

In addition to the BOP data set for 2007-2008, an ambient PM data set with chemical speciation is available for 1998-1999 (Visser *etal.*, 2001). The patterns in both data sets are similar; they show a good correlation between elements that can be used as a tracer for mineral dust (Al, Si, Ti), a decreasing fraction of MD_2.5 when the absolute contribution of MD to PM_{10} increases, and an elevated MD concentration in PM in urban and traffic locations compared to those in the rural background. However, there are also some remarkable differences. Most notable is the difference in absolute concentration levels of MD in PM_{10}. The amount of mineral dust observed in PM_{10} during the BOP campaign in 2007-2008 was significantly lower (by about a factor of 2 to 3) than estimated when using data from Visser *etal.* (2001) for the 1998-1999 period. Although the locations in both studies were not exactly the same, this difference was also observed for overlapping locations, such as Vredepeel, which was included in both data sets. If conditions at specific locations are not the explanation for this discrepancy, then it could be due to 1) the meteorology in both 1998 and 1999 causing higher emissions of MD, 2) differences in analytical techniques and sampling, 3) changes in MD source strengths, or 4) a combination of these three possibilities.

It is unlikely that we will learn the exact cause of this difference, for example, because an intercomparison of samples is no longer possible. Before trying to further understand the cause of these different MD levels in PM_{10}, we first made a comparison with available data for neighbouring countries. Especially in Belgium and in German provinces bordering on the Netherlands we would expect similar contributions of mineral dust to PM_{10}, as this would not originate from a local source, but from one with a more regional character and influenced by regional climate, meteorology and soil types.

3

Mineral dust in PM_{10} in surrounding countries

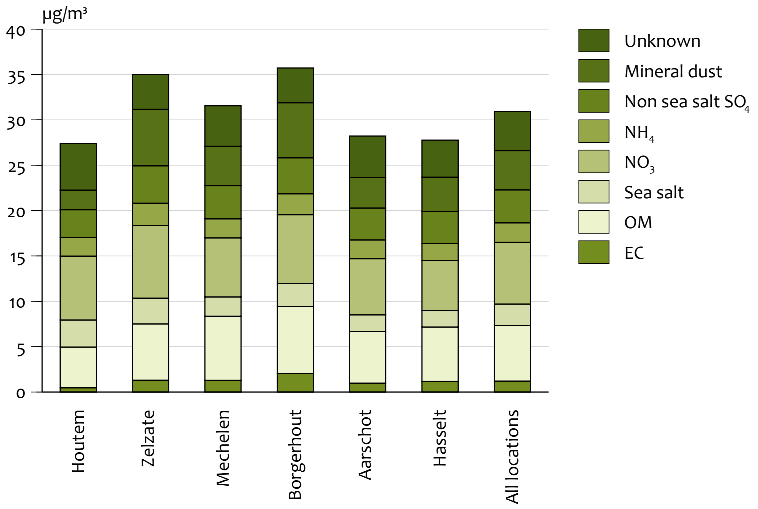
Data on ambient PM_{10} and/or $PM_{2.5}$ with full chemical speciation are scarce. Therefore, it is almost impossible to obtain a data set for a large geographical domain (such as north-western Europe) which covers the same period in time. Having such a data set is desirable because it would reduce the uncertainty introduced by different meteorological conditions and wind directions. Chemically speciated PM data are available for border provinces in Belgium (Flanders) and Germany (North Rhine-Westphalia). The Belgian data have no time overlap with the Dutch BOP data, but for the data from North Rhine-Westphalia a period of overlap exists. The data from both countries are briefly described in this chapter.

3.1 Belgium

From September 2006 to September 2007 chemical characterisation of PM_{10} was carried out by the *Chemkar PM_{10}* project at six measuring locations in Flanders (Belgium). Flanders is the part of Belgium that borders on the Netherlands and is in many ways comparable to the Netherlands. Therefore, the results are relevant for comparison with and interpretation of the Dutch data obtained in the BOP programme. For a detailed description of the methodology and results from the Chemkar project we refer to (VMM, 2009). The VMM chose their monitoring locations in such a way as to obtain a reasonable spread in location and variety in type of location. The six locations were Houtem (rural background), Zelzate (industrial), Mechelen (suburban), Borgerhout (urban background), Aarschot (rural background) and Hasselt (suburban). The results from the Chemkar project indicated that the share of mineral dust in Flanders varies from 2.2 to 6.2 µg/m³ (Figure 3-1). The average contribution for all locations was 14%, but for some this went up to ~18% (Zelzate, Borgerhout). In the case of Zelzate, a specific industrial activity may have caused the elevated levels of mineral dust in PM_{10} (VMM, 2009).

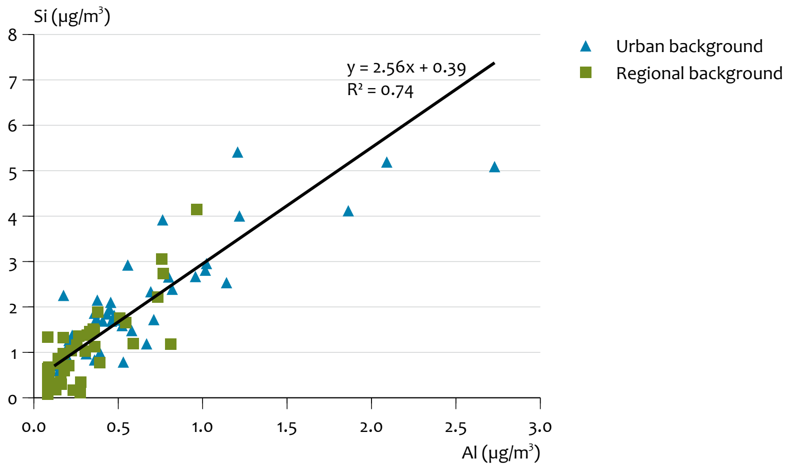
The elevated share of mineral dust at Borgerhout was clearly associated with road traffic. This can be deduced from Figure 3-1 by looking at the increase in elemental carbon (EC) in PM_{10} at Borgerhout; EC is a known indicator of (diesel) traffic.. Moreover, the detailed chemical analysis presented in VMM (2009) showed that, at this location, Cu was also elevated. Cu

Average share of major component in PM_{10} for six stations in Flanders **Figure 3.1**



Average share of major components in PM_{10} at six locations in Flanders (VMM, 2009).

Si and Al concentration in PM_{10} **Figure 3.2**



Si and Al concentrations in PM_{10} for an urban and a rural background location in North Rhine-Westphalia (Data from Quass et al., 2009).

is a good indicator of brake wear and hence of traffic (Denier van der Gon *etal.*, 2007). Furthermore, the contribution of MD at location Borgerhout is a factor of ~2 higher than at the rural background locations Houtem and Aaeschot. Therefore, we concluded that the enhanced mineral dust which was found in PM_{10} at traffic locations in Flanders, suggests that resuspension of PM caused by road traffic largely explains the observed elevation. At the industrial location of Zelzate, the increase in mineral dust may have been due to a specific industrial process, but also (partly) was associated with resuspension, since heavy-duty trucks have a much higher resuspension emission factor than passenger vehicles (Denier van der Gon *etal.*, 2008).

3.2 Germany

North Rhine-Westphalia is situated in the western part of Germany and shares borders with Belgium in the south-west and the Netherlands in the west and northwest. Recently, the unit 'Air Quality & Sustainable Nanotechnology' of the German Institute of Energy and Environmental Technology (IUTA) executed a one-year measurement programme, from April 2008 to March 2009, to investigate the chemical composition of PM in North Rhine-Westphalia (Quass *etal.*, 2009). The IUTA kindly provided the data that they collected during the overlapping period (April to Sept. 08) with the BOP programme. The Si and Al data from Quass *etal.* (2009) showed a good correlation, both for the rural and urban background locations (Figure 3-2), confirming that these elements mostly originate from the same type of material and/or the same source. Since we had access to the original data, we could analyse these data in the same way as the BOP data in Chapter 2. The average contribution of MD to PM_{10} over the period from April 2008 to March 2009 was 3.5 µg/m³ for the urban background locations and 1.6 µg/m³ for the rural background locations. The pattern of the mineral dust concentration in PM_{10} over time (Figure 3-3) showed a similar pattern as that of the Dutch BOP data in Figure 2-5, with episodes of clearly elevated MD levels.

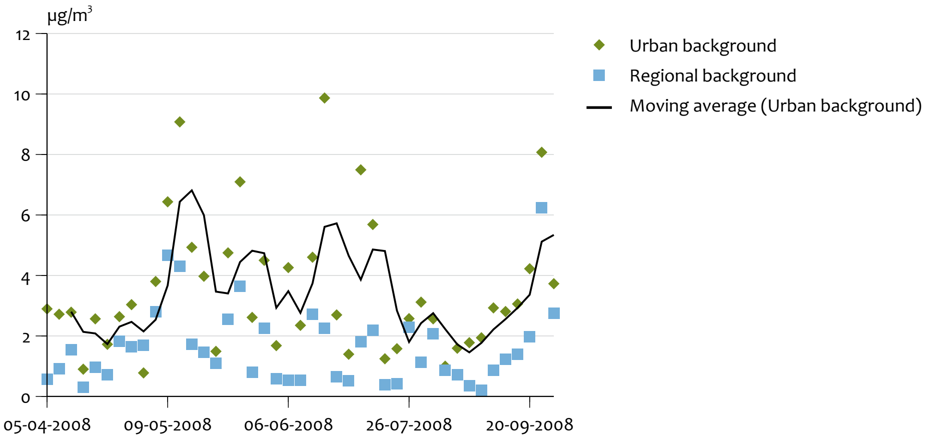
3.3 Comparison with the Dutch data and Conclusions

In the Flemish data, the contribution of mineral dust was higher than in the BOP data for 2001 to 2008. However, the sampling periods did not overlap. The sampling period of the North Rhine-Westphalia data partly overlapped and, although contributions were higher, they were closer to the BOP data. It is, therefore, not unlikely that the sampling period played a role, although this could not fully explain the discrepancy in the data. The amount of mineral dust observed in PM_{10} during the BOP campaign in 2007-2008 confirmed the increase in MD in the urban environment and at traffic locations. However, the relative increase as derived from the tracer elements was significantly lower than observed in Flanders (VMM, 2009), North Rhine-Westphalia (Quass *etal.*, 2009) and earlier in the Netherlands (Visser *etal.*, 2001). To further investigate this apparent contradiction, we plotted the coarse fraction of PM_{10} ($PM_{2.5\text{-}10}$ or PMc) that was measured during the BOP campaign, as a function of the type of location and the MD in the PMc fraction (Figure 3-4, left). Note that this was not feasible for the traffic location of Breda, as no $PM_{2.5}$ was monitored at this location. The PMc fraction at the urban background and traffic locations was more than double that of the rural background location. This is consistent with urban activities, such as road traffic, which is an important source of coarse PM, and the more general findings in neighbouring countries. However, Figure 3-4 also clearly illustrates that during the BOP campaign MD was only a minor fraction of the PMc. This is remarkable and contrary to the average observational data available for Europe (see also Chapter 5). Moreover, not only was this a small fraction, but also the relative increase at locations from rural to urban background and traffic, was considerably higher for total PMc than for MD in this fraction (Figure 3-4, right). For a discussion on the complete composition of PMc and mass closure we refer to Schaap *et al.* (2010).

Is it possible that the BOP campaign did not properly capture the increase in road traffic, for example, because of a sampling artefact? No, this is not likely. Although the

Estimated mineral dust in PM_{10} in North Rhine-Westphalia

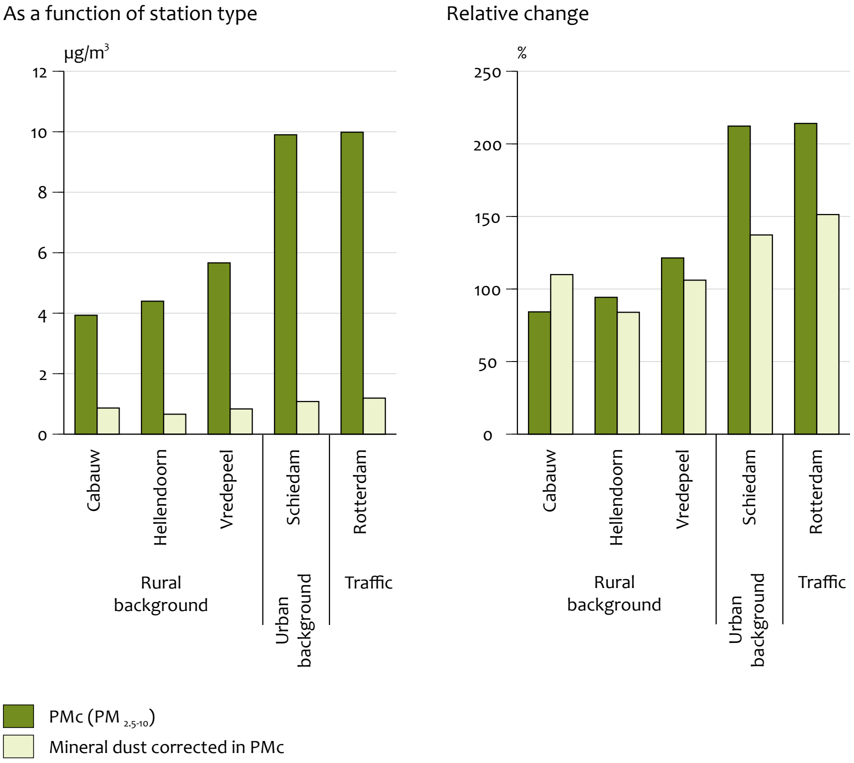
Figure 3.3



Estimated mineral dust in PM_{10} in North Rhine-Westphalia in 2008-2009, based on data from Quass et al. (2009).

Concentration of $PM_{2.5\text{-}10}$ (PMc) and mineral dust in PMc

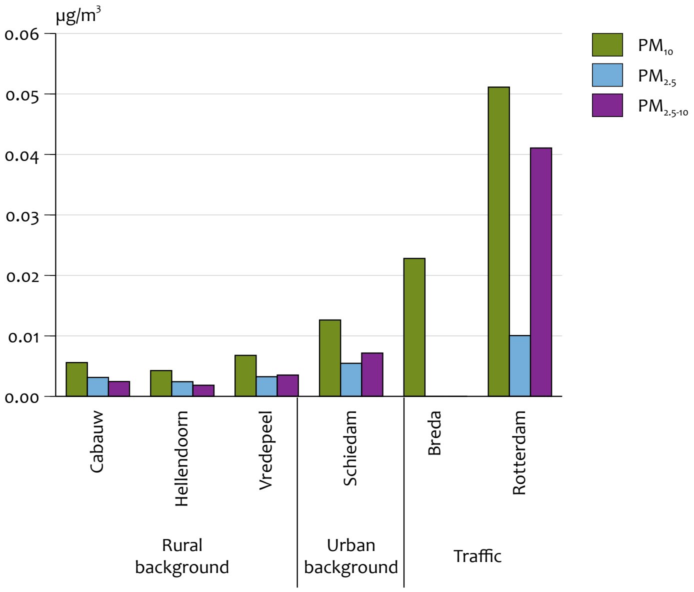
Figure 3.4



Concentrations of $PM_{2.5\text{-}10}$ (PMc) and MD in PMc as a function of location type (left) and the relative change of PMc and MD in PMc as a function of location type (right).

MD contribution was smaller than expected, the Al and Si analysis was very consistent (Figure 2-2, Figure 2-3). To further check the capturing of increments at traffic locations, we investigated the copper concentration in the PM fractions for the different locations (Figure 3-5). Copper is a reliable tracer for brake wear (Denier van der Gon *etal*., 2007). The increase observed at traffic locations clearly illustrates that the traffic was accurately characterised in the BOP chemical analysis data and, therefore, in the sampling. The increase in copper in the coarse fraction of PM_{10} was exactly in line with data for the Netherlands and Switzerland, reported earlier by Denier van der Gon *etal*. (2007). Hence, as there was no reason to attribute the relatively low MD share to a sampling artefact, we concluded that PMc played an important role at traffic

Concentration of Cu in various PM fractions by location type **Figure 3.5**



Concentration of Cu in various PM fractions per location type.

locations, although we do not yet sufficiently understand the processes and sources to effectively mitigate or reduce these concentrations. This lack of understanding is reflected in the lower MD share, compared to previous Dutch measurements, as well as recent chemical speciation data from neighbouring countries.

Although the MD contribution appeared to be relatively low in the data from the BOP campaign, the important conclusion can be drawn that, even if the contribution would double in other years, and match levels observed in Flanders or North Rhine-Westphalia, MD would only cover a small part of the large ‘non-modelled’ PM source, previously attributed to ‘soil material and other natural sources’ as stated in the Dutch benchmarking publication ‘Fijn stof nader bekeken’ (MNP, 2005)

4

Modelling mineral dust over Europe

In the previous chapters we have shown that mineral dust is an important constituent of PM_{10}. Mineral dust may originate from distinctly different sources, such as wind erosion of bare soils, agricultural land management, resuspension of road dust, road wear, driving on unpaved roads, handling of materials, and building and construction activities (see Chapter 0). This chapter describes the development of a methodology for checking first-order estimates of the various source strengths in Europe, in general, and the Netherlands, in particular.

For this purpose, we implemented simple and therefore transparent emission functions in the LOTOS-EUROS model for the processes *wind erosion of bare soils, agricultural land management, resuspension of road dust and road wear,* as these are thought to make up the highest source contributions in north-western Europe. To simplify the problem, a few assumptions were made beforehand.

- We established that primary (or direct) road wear, which also results in the emission of mineral dust (see Chapter 0), is a much less important source than resuspension (see below), and would not need to be treated separately. This simplified the traffic component, as road wear could not be distinguished based on its chemical composition from re-emitted MD due to resuspension.
- We included boundary conditions for desert dust from the global TM5 model. As the uncertainties concerning the dust emissions were very high, special attention was given to the identification of the controlling factors for the source estimates. We refer to Schaap *etal.* (2009) for an exact definition of the representation of desert dust in the current LOTOS-EUROS model.

Within this chapter the focus is on mineral dust share of PM_{10} as , which is mainly found in the coarse PM fraction. The emission modules implemented in the LOTOS-EUROS model for 1) wind erosion of bare soils, 2) agricultural land management, and 3) resuspension of road dust (including road wear), were developed in the BOP programme and have been extensively described in Schaap *et al.* (2009). Below, only a brief summary is provided of the most important characteristics, and we refer to Schaap *et al.* (2009) for a detailed description and underlying motivations of the choices that were made.

4.1 Emission module for wind-blown dust

When the wind blows over a surface, it exerts pressure on it, which may mobilise soil material. Such transportation of soil material can be divided in three regimes: creep, saltation and suspension (See Figure 4-1). The (type of) mobilisation of a particle is controlled by gravitational force, inter-particle cohesion and wind shear stress on the particle, all depending on the size of the particle. Each of these processes were discussed by Schaap *et al.* (2009). The parameterisation for the horizontal and vertical dust flux utilised in the BOP research programme has been based on the work by Marticorena and Bergametti (1995, 1997), Gomes *et al.* (2003) and Alfaro *et al.* (2004).The relation between the horizontal flux of saltating soil aggregates and its induced vertical flux of aerosol particles, depends on the size of the soil aggregate, that of the released aerosol and the kinetic and binding energies involved. Alfaro *et al.* (1997, 1998) developed a sandblasting model which takes these dependencies into account. This sandblasting model was adapted for the calculation of wind-blown dust fluxes by the LOTOS-EUROS model (Schaap *et al.*, 2009).

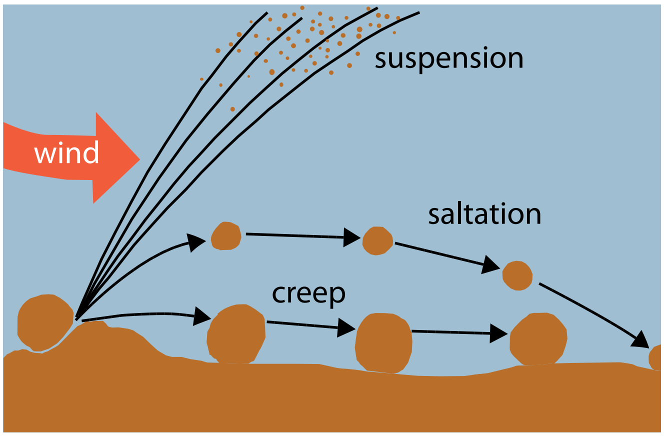
This wind-blown dust module in the LOTOS-EUROS model takes into account the spatial distribution of the main soil types in Europe, as soil particle size is an important parameter, next to soil moisture, which may vary considerably across Europe. Moist soil inhibits the release of soil particles, because it requires larger threshold friction velocities for the uplift of particles.

In our configuration of the LOTOS-EUROS model, the effect of precipitation (rain, snow) on dust emissions was represented by assuming the dust flux to be zero during precipitation events and up to 48 hours onwards. In case of a snow cover (aas derived from three-hourly ECMWF meteorological data, the dust flux was set to zero.

4.2 Agricultural activities

Soil erosion as a result of land management activities leads to soil degradation and loss of soil fertility. In addition, recently, it has also been acknowledged that (soil) dust generated during land management is not only causing soil degradation

Schematic of soil movement processes **Figure 4.1**



Schematic of soil movement processes.

$PM_{2.5}$ and PM_{10} emission factors applied in the LOTOS-EUROS model **Table 4.1**

Reference	**Location**	**Year**	**Method**	**Activity**	**Operation type**	**Emission Factor**	
						$PM_{2.5}$ ($mg.m^{-2}$)	**PM_{10} ($mg.m^{-2}$)**
Öttl et al. (2007)	Germany	2004	PM obs., disp. mod.	land prep.	Harrowing	29	82
					Disc harrowing	12	137
					Cultivating	6	186
					Ploughing / tillage (wet)	0.5	120
Gaffney et al. (2003)	California, US	1995-2003	Upwind-downwind	harvest	corn	21*	191

** derived from the average ratio in emission factors between $PM_{2.5}$ and PM_{10}*

but may also be a significant source of PM_{10}. Currently, questions are being raised about its possible contribution to fugitive PM_{10}. However, dedicated research on emission factors of PM_{10} and $PM_{2.5}$ due to land management has been very limited. This is not surprising as it is quite complicated to determine what would be a ‘representative’ situation; dust emissions from land management depend on many variables, such as soil type, meteorological conditions, crops, and type of machinery, to name but a few. There are only a handful of studies available in which emission potentials have been estimated for several land management activities. These studies were reviewed by Schaap *et al.* (2009). The ranges of reported emission factors were wide, and varied from 0.3 to 14 $kg.ha^{-1}$ and from 3.8 to 45.7 $kg.ha^{-1}$ for land preparation and harvesting, respectively. Clearly, the uncertainty associated with these numbers is large. Moreover, it is not known how many of the emitted particles were mineral dust. Some of these soil particles will have been organic, such as the particles released from crops during harvesting.

Despite the limited knowledge on and availability of observations, a first selection was made from the emission factors to calculate the emissions related to agricultural land management, using the LOTOS-EUROS model (Table 4-1). The emission factors were discussed in more detail by Schaap *et al.* (2009), and since their publication, the emission factor for ploughing or tillage were adjusted as they resulted in an overestimation of the mineral dust concentrations over Europe. Hence Table 4-1 is an updated version compared to Schaap *et al.* (2009).

Soil dust resulting from land management operations is characterised by high emission intensities, occurring in relatively short periods of time. In addition, the timing of all operations varies widely and is dependent on the local growing season, local weather and type of crop. This has been taken into account in the module developed for the LOTOS-EUROS model, with the use of a simple calendar of agricultural practices (Table 4-2) Winter activities on the fields are negligible and were not included. Land preparation, characteristically, is carried out in spring and late autumn, whereas harvesting takes place from August to October. During the growing season, when agricultural land management activities are minimal, we assumed emissions to be negligible. For more details on the agricultural calendar and its respective limitation on emissions, we refer to Schaap *et al.* (2009).

The module that we used in the LOTOS-EUROS model is a first approach to get a better grip on this source of MD in the PM_{10} size range. A first empirical correction was made by adjusting the emission factor for ploughing. Through

Calendar of agricultural activities

Table 4.2

Jan.	Feb.	Mar.	Apr.	May	Jun.	Jul.	Aug.	Sept.	Oct.	Nov.	Dec.
winter		land prep. & seeding		growing season			harvest			winter preparation	

$PM_{2.5\text{-}10}$ emission factor (mg.vkm-1) for traffic-related resuspension as a function of road type for light- and heavy-duty traffic, applied in the LOTOS-EUROS model

Table 4.3

	Road type		
	HW	**RUR**	**URB**
HDV	198	432	432
LDV	22	48	48

iteration and model evaluation the module still requires further adjustment, as it suffers from oversimplification. Some of the main points that would need to be addressed in this respect, in the near future, are the fraction of emitted soil organic material as opposed to pure MD, the differences between cropping calendars across Europe, and the influence of regional climate. Regarding the climatic influence, in our research we have now only omitted the emissions during rain events.

4.3 Resuspension due to road traffic

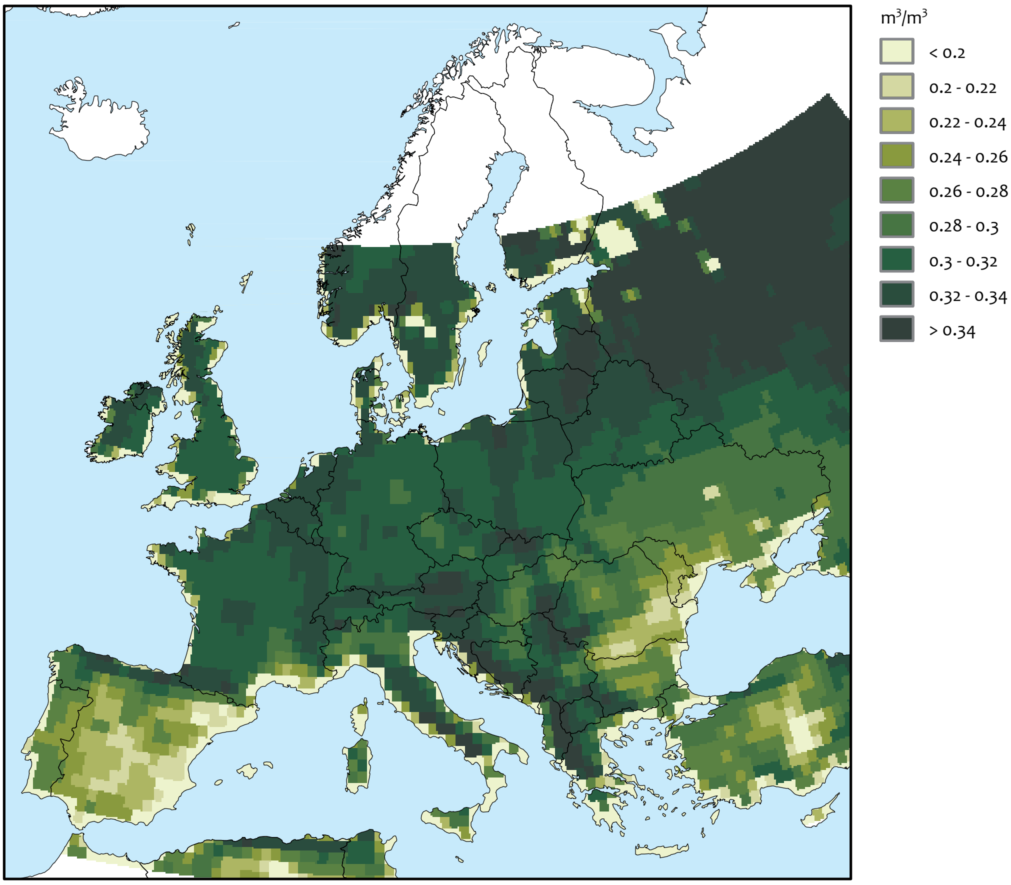
Resuspension of dust particles smaller than 10 microns due to vehicle-generated turbulence is widely recognised as a significant source of PM_{10}, especially in the urban environment. The material that is being resuspended varies with the nature of the local circumstances. It typically includes particles from vehicle tire wear, brake wear, primary exhaust emissions that have settled on road surfaces (perhaps adhering to larger particles), and environmental dust from many sources, such as pollen, sea salt, construction work and wind-blown soils. In addition, the contribution from resuspension to total PM also depends on meteorological conditions; during episodes of rain and when surfaces are wet, resuspension will be much less. The mix of particles with varying chemical composition, and effects from local and climatic conditions make resuspension a difficult source to generalise about and to quantify. Various studies have suggested that emissions from resuspension are of the same order of magnitude as primary emissions from road traffic, relating to vehicle exhaust fumes, tire wear and brake wear (Lenschow *et al.*, 2001; Harrison *et al.*, 2001; Gehrig *et al.*, 2004; Thorpe *et al.*, 2007). Furthermore, a number of studies have reported that resuspension due to heavy-duty vehicles (HDV) is higher by about a factor of 8 to 10, than that caused by light-duty vehicles (LDV) (e.g., Abu-Allaban, 2003; Gehrig *et al.*, 2004; Thorpe *et al.*, 2007). An extensive review of the literature on resuspension due to road traffic can be found in a study by Schaap *et al.* (2009). Based on this review, we made a selection of emission factors for resuspension expressed in mg/vkm (Table 4-3). Special correction factors were developed to adjust the emission factors in Table 4-3 for climate (southern Europe) and for non-skid measures, such as studded tires and road gritting (northern Europe) which are known to increase resuspension emissions.

4.3.1 Accounting for higher contribution from resuspension in southern Europe

Source apportionment studies (Putaud *et al.*, 2004; Querol *et al.*, 2004; Kuhlbush *et al.*, 2000; Beekmann *et al.*, 2007) have pointed out that mineral ambient PM_{10} concentrations in city background locations and near busy streets are higher in southern European countries (e.g. Spain) than in central Europe. Hence, there is a need to account for this increased flux of resuspended particles as a function of the climatic conditions in southern Europe. Schaap *et al.* (2009) provided a (more) detailed discussion on the various options to do such accounting. Here we have summarised our selected approach. An important prerequisite for modelling regional dependency on climate conditions is that it should yield a continuous relationship (without jumps) in all of Europe. One way to pursue this would be to use a proxy map for Europe, to which the emission potential is related. As soil aridity plays an important role, we chose to use the average annual soil moisture map for Europe as a proxy field (*Figure 4-2*). The next step was that of relating the existing – and poorly covered – emission factor estimates to the soil moisture map. This posed a challenge, as no emission factor estimates for southern Europe were available. Therefore, we based the emission sensitivity to soil moisture on the gradients in the more widely performed PM observations of material resuspended by road traffic. We assumed that the contribution from road traffic itself (deposited wear and exhaust particles) to the total amount of resuspended material within cities in central and southern Europe would be similar. Where resuspension caused by road traffic is the dominant source, this would obviously be the case. This approach was further justified by the observations studied by Querol *et al.*(2007). They observed a higher concentration of certain crustal elements (Al, Ca, Fe, Ti, Li, Rb) at urban locations, compared to rural sites, indicating that these elements originated mainly from road traffic. Hence, under the above assumption, the ratio of the increment in MD at urban locations between southern Europe and central Europe would be a measure of the relative increase in the resuspension source strength. Based on their review, Schaap *et al.* concluded that the ratio of this increment was ~ 2.5.

The concentration of a primary pollutant in a source area is, besides the emission strength, also (linearly) dependent on the height of the mixing layer. Hence, we needed to account for the differences in mixing layer heights (h_{mix}) to relate the

Annual average volumetric soil moisture **Figure 4.2**



Average annual volumetric soil moisture in 2005 (ECMWF).

differences in urban increment to emission estimates. To do so, we used the average annual mixing-layer height from ECMWF for Spain (750 m), and central Europe (550 m). To explain the observed Spanish urban increment in MD, the emissions would have had to be about 2.5 * 1.4 = 3.7 higher than in central Europe. Since we want to use the soil moisture map (*Figure 4-2*) as a proxy for the variation in the MD correction factor, a simple linear interpolation was carried out going from "3.7" for Spain (soil moisture = 0.225 $kg.kg^{-1}$) to "1" central Europe (soil moisture = 0.3 $kg.kg^{-1}$) (Figure 4-3). We did not extrapolate outside the indicated range in soil moisture content. Hence, within the model domain, a maximum emission estimate for light-duty vehicles (LDVs) was used: 3.7 * 22 = 80 $mg.vkm^{-1}$ for motorways and 3.7 * 48 = 176 $mg.vkm^{-1}$ for urban and rural roads.

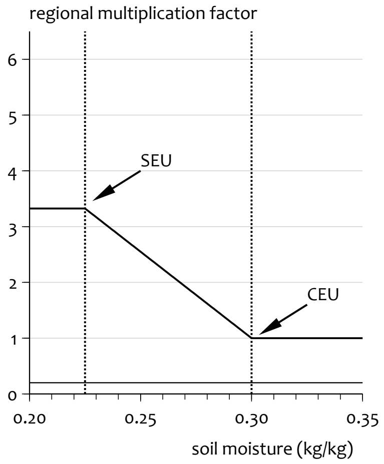
4.3.2 Accounting for higher contribution from resuspension in northern Europe

The use of studded tyres and road gritting in the winter season causes additional and high peaks in PM_{10} emissions, with a contribution of non-exhaust mineral material of up to 90% in Nordic and Alpine countries (Larssen *et al.*, 1994; Omstedt *et al.*, 2005; Kupiainen *et al.*, 2003). Omstedt *et al.* (2005) developed an empirical model for research into the influence of road gritting and studded tyre usage on PM emissions from resuspension on Swedish roads. The model makes use of emission factors and traffic data, and takes into account the thickness of the on-road dust layer,. This model has been applied, specifically and successfully, to investigate the effects of road gritting and the use of studded tyres in Scandinavian winters on fugitive PM_{10} concentrations. For Scandinavian countries, based on a model study by Omstedt *et al.*(2005), a multiplication factor of 2 was applied for the springtime months of March, April and May, to standard emission factors for PM_{10} and $PM_{2.5}$, for light- and heavy-duty vehicles, to correct for additional resuspended material as a result of road gritting and tyre use. We did not account for road gritting in the Alpine region.

4.4 Road wear

Road surfaces consist mainly of stony materials and the contribution from road wear to the total PM_{10} emission factor is difficult to separate from the contribution from wind-blown or resuspended soils and other geological material. However, in road tunnels, the amount of wind-blown soil dust coming from sources other than road wear (e.g. the shoulders) would be limited. A first approximation would therefore be that all mineral dust in PM_{10} produced in these tunnels originates from road wear. Gillies *et al.* (2001) reported that ~12% of PM_{10} in the Sepulveda Tunnel (Los Angeles, the United States) was

Correction factor for MD resuspension by traffic **Figure 4.3**



Regional multiplication factor for mineral PM10 emission from road traffic that corrects for climatic differences throughout Europe, based on observed PM, average annual soil moisture and mixing layer height in reference areas in central Europe (CEU) and southern Europe (SEU).

of geological origin. Denier van der Gon *et al.* (2003) used these results in combination with a chemical analysis of PM_{10} formed in the Maas Tunnel (Rotterdam, the Netherlands), to derive an emission factor of 3 to 4 mg per vehicle kilometre (vkm) for mineral dust from road wear. Road wear depends not only on surface material, but also on vehicle braking frequencies and cornering, which cause additional friction. In a tunnel environment these situations are limited compared to those of average urban traffic. Hence, we have awarded the tunnel road wear emission factor a lower value. In the literature, emission factors for road wear often range between 8 and 10 mg/vkm, which is consistent with a 3 to 4 mg/vkm lower limit. When comparing these values to the estimates for resuspension (Table 4-3), it becomes clear that primary emissions from road wear (excluding the use of studded tires and road gritting) could modify total PM_{10} MD emissions with 10 to 20%, but that resuspension is by far the more relevant process.

4.5 First results

The modelled average annual concentration of mineral PM_{10} for Europe, came to 1 to 2 µg.m^{-3} (Figure 4-4). Concentrations exceeding 3 µg.m^{-3} have been modelled for densely populated areas and relatively dry and arid environments. Lower values of 0.2 µg.m^{-3} have been simulated for large forests, where no sources are located.

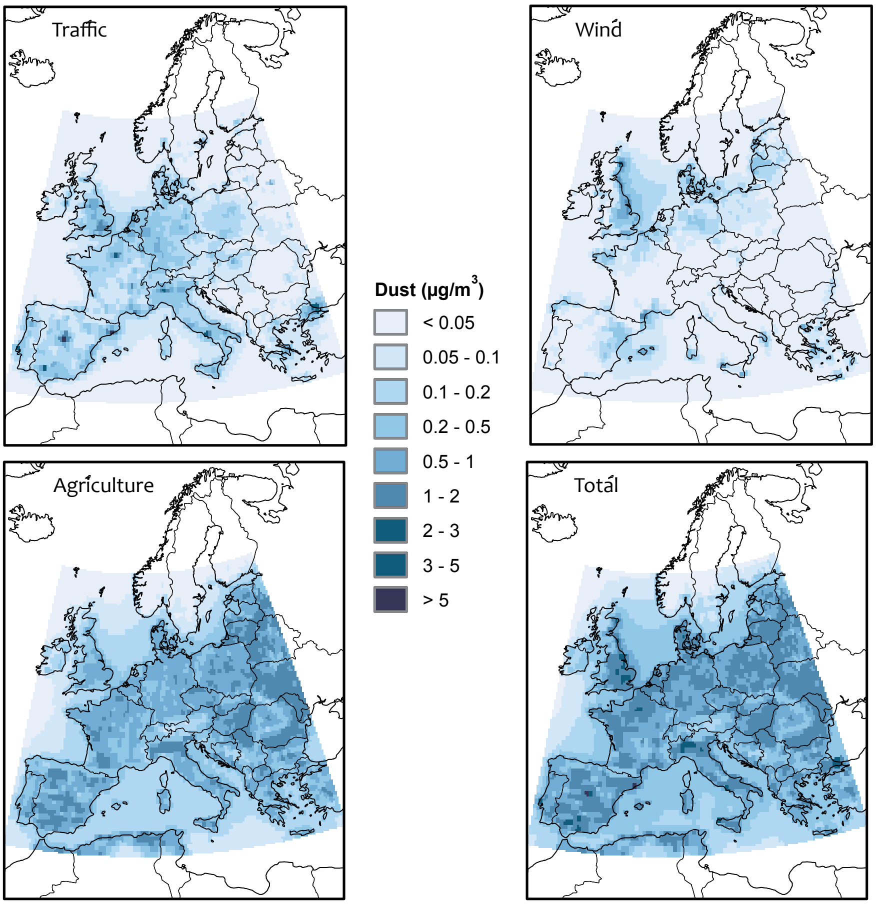
Wind-blown dust. The modelled average annual wind-blown dust emission showed that wind erosion from European soils contributes generally less than 0.1 µg.m^{-3}. In the modelling, the importance of this source compared to other mineral dust sources, although uncertain, appeared to be low. The highest contributions of wind-blown dust were found in areas with high average annual wind speeds, in combination with areas that hold a relatively high amount of the type of soil dust suitable for resuspension.

Agricultural land management. The model results indicated that annual contributions from agricultural land management activities for Europe are about 0.2 to 1 µg.m^{-3}, but with contributions as high as between 1 and 2 µg.m^{-3} for the arable areas. The latter is in line with the current understanding that, for arable land, land management is by far a larger source of dust than is wind erosion. The spatial distribution of ambient mineral matter from agricultural land management activities was very different than from road traffic or wind-blown dust. Compared to resuspension due to road traffic, the contribution showed a more regional dependence. Within the agricultural areas the highest concentrations were found in regions with low amounts of precipitation.

Resuspension due to road traffic. The annual contribution from resuspension was less than 0.3 µg.m^{-3} in rural areas, but varied to up to 5 µg.m^{-3} or more in densely populated regions. The spatial distribution of resuspension due to road traffic showed the highest levels in densely populated areas, reflecting vehicle use. . In Europe's main cities, such as London, Paris, Madrid, and Milan, this has led to high peaks of resuspended soil dust, compared to levels in nearby rural areas (Figure 4-4). It is only around cities that the contribution from resuspension due to road traffic exceeded that from agriculture.

Desert dust. The contribution of desert dust appeared only important in the Mediterranean Basin at latitudes below 44° N (Figure 4-5). In Spain, southern Italy, Greece, and Turkey, the average annual inflow of desert dust contributed an additional 0.2 to 1 µg.m^{-3}, and in some locations up to 2 µg.m^{-3}, to the total ambient mineral dust concentration. On occasion, significant amounts of African desert dust would reach higher latitudes. In most of these cases desert-dust enriched air masses were first being transported over part of the Atlantic Ocean before reaching central Europe. Sporadically, there was inflow of desert-enriched air mass at the south-eastern boundary of the model domain, with dust originating from south-eastern and Asian desert areas, rather than from the

Modelled mineral dust contributions to PM_{10} **Figure 4.4**

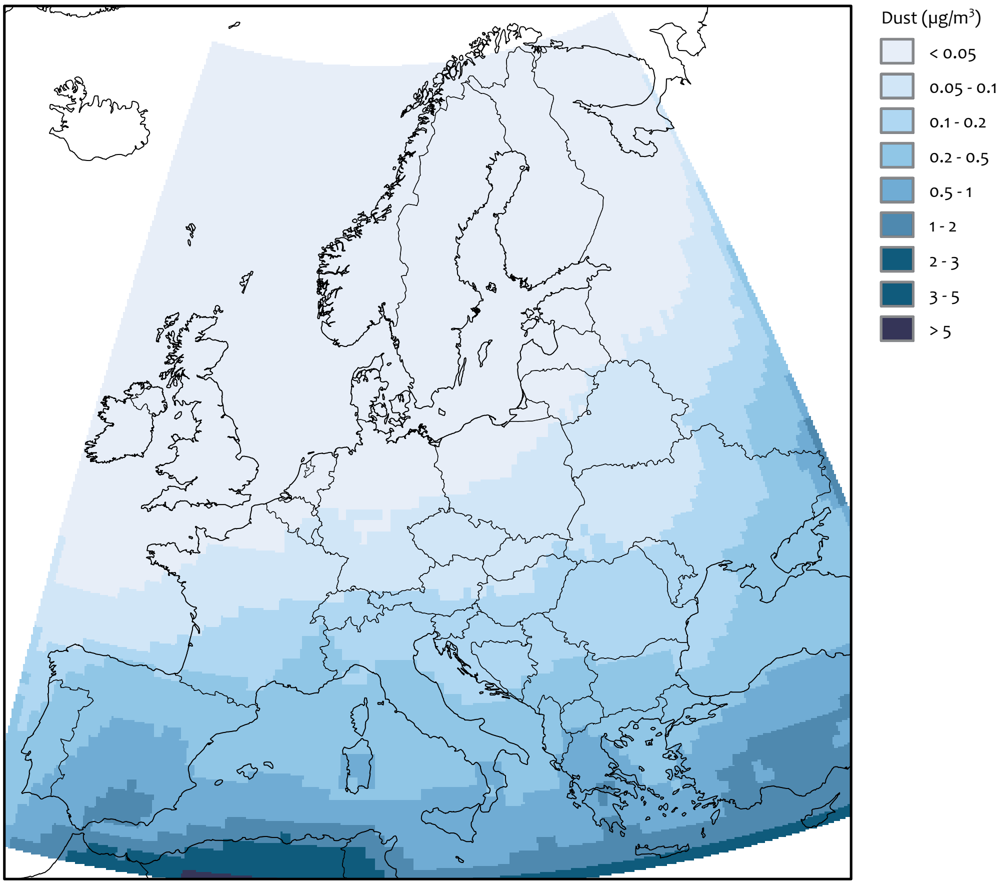


Modelled mineral dust contributions to PM_{10} as a result of resuspension caused by traffic (top left), from wind (top right), from agriculture (bottom left), and the total modelled resuspension of mineral dust contributing to PM_{10} (bottom right) for 2005.

Sahara. In the Netherlands, 2005 was the only year that the level of desert dust reached a concentration of 2.5 µg.m^{-3} (Schaap *et al*., 2009).

Modeled mineral dust contribution to PM_{10} of desert sources

Figure 4.5



Modelled mineral dust contribution to PM_{10} from desert sources outside the LOTOS-EUROS model domain (input at domain boundaries from TM5 transport model).

Mineral dust in PM_{10} and $PM_{2.5}$ in Europe

5

Analysis of PM_{10} and $PM_{2.5}$ samples has shown that mineral dust (or soil dust) contributes significantly to ambient PM_{10} and $PM_{2.5}$ concentrations in Europe (Putaud *et al*., 2003; Querol *et al.,* 2004; see also Chapter 1). The sources of fine mineral dust are poorly understood, may vary widely in time and space and, therefore, are not well-represented in emission inventories. Moreover, for some sources, such as resuspension of road dust, inclusion in an inventory of primary PM_{10} is arbitrary, as the dust particles are a re-emission of previously emitted material. To get a better grip on the significance of MD as a component of total PM, we investigated reported MD contributions for Europe or estimated contribution levels using tracer elements derived from chemical elemental analysis of data on PM samples. Furthermore, to verify the results from modelling MD in ambient PM_{10} and $PM_{2.5}$ (see Chapter *Error! Reference source not found.*), a database of observational data for PM mineral dust in Europe would be needed. This would allow for comparison of predicted crustal PM with actual observations and for correction of the source functions, if needed. The objectives of the work described in this chapter, therefore, were to:

- Obtain a balanced view on the significance of mineral dust as a component of PM_{10} and $PM_{2.5}$ across Europe and across all location types.
- Collect a set of observational data, which can be used to validate source functions implemented in the LOTOS-EUROS model for mineral dust

First, this chapter describes our approach for investigating mineral dust contributions to PM_{10} and $PM_{2.5}$ over Europe, including their spatial distribution and a compilation of daily data on MD to validate modelled concentrations (Section 5.1). Relationships that can be used to derive the mineral dust component in PM_{10} and $PM_{2.5}$ are discussed in Section 5.1.2. Section 5.2 describes collected data, analysed in a number of steps, which started with limited data sets to address simple questions. Gradually more data were brought in to work towards coverage of the European domain.

Next, Section 5.3 discusses two studies (Putaud *et al*., 2003 and Querol *et al*., 2004) that each compiled data from different locations. We paid additional attention to these studies because the data they collected were measured following rather similar protocols. The data sets were used to derive preliminary guidance on MD contributions across different types of locations and geographical patterns. In Section 5.4 we brought all studies together, identified the geographical location of each study and the location types for which data were reported.

Having taken data together from many different studies may also have obscured the true relationships because, for example, different analytical techniques and sampling strategies were used. Therefore, the conclusions from the data compilation in the previous sections were crosschecked using crustal PM in $PM_{2.5}$ data derived in an independent study which measured elemental composition of ambient fine particles at 21 European locations (Section 5.5).

5.1 Compilation of data on Mineral Dust in PM across Europe

Most observational PM data only provided the mass of PM_{10} and/or $PM_{2.5}$. Mineral dust in PM_{10} and $PM_{2.5}$ was never directly measured as such. If an estimate of MD is given, it was usually based on the use of certain elements (e.g. Si, Ca) as tracers of mineral dust. However, samples of PM_{10} and $PM_{2.5}$ with complete chemical composition were scarce and often not all elements were available. Since MD in PM_{10}/$PM_{2.5}$ was only available for a limited set of environments, years and locations, we needed to develop general relationships to be able to extrapolate to non-covered areas. Therefore, we set out to answer a number of key questions;

1. Is the contribution of MD to PM_{10}/$PM_{2.5}$ dependent on location type? Location types are defined as natural, rural, urban, industrial and traffic locations.
2. Is the contribution of MD to PM_{10}/$PM_{2.5}$ dependent on geographical location, influenced by, for instance, meteorology or sources in the vicinity (e.g., Saharan dust)?
3. Is the relative contribution of MD to PM_{10} different from its relative contribution to $PM_{2.5}$?
4. Is the level of comparability of different studies hampering a generalisation of the findings?

5.1.1 Collection of data on chemical composition of PM_{10} and $PM_{2.5}$

Observational data on PM_{10}/$PM_{2.5}$ together with analyses on the chemical composition were collected in a first step in the strategy of investigating the contribution and distribution of mineral dust (MD) in PM_{10}/$PM_{2.5}$ over Europe (see also Chapter 1, Figure 1-1). From the literature, data were compiled that included analyses of one or more of the following elements; aluminium (Al), silicon (Si), titanium (Ti), calcium (Ca), potassium (K) and iron (Fe). Data that did not provide chemical speciation but presented a direct estimation of mineral dust were also collected. An overview of the collected data and references to their origins are presented in Annex 3. In many cases, authors were contacted to supply us with additional data if these were available (see Section 5.2).

5.1.2 Estimation of mineral dust in PM_{10}/$PM_{2.5}$ from elemental analysis

Unlike, for example, elemental carbon, mineral dust is not measured as a separate property or component in PM. It needs to be estimated on the basis of the contribution of (tracer) elements that originate from mineral dust. A number of elements are known to be present in mineral dust, the most important being Si, Al, Ca, K, Fe and Ti. The strength of the correlation between these elements in PM samples, especially in the coarse fraction, confirm whether these elements may originate from the same source (e.g., Figure 2-2 and Figure 2-6). In this study, we used an estimation approach for calculating the contribution of mineral dust (MD) to PM_{10}, based on concentrations of Al and Si, as discussed in detail in Chapters 2 and 3. The MD mass that was estimated by using Equation 2.1 included the contribution of minerals other than Al silicates and quartz, which are also present in soils.

Our estimation would only be valid if the Al and Si concentrations indeed originated from MD. This hypothesis was checked by studying the correlations between these elements at different locations. A good correlation between potential tracers of MD is important because, in actual practice, the number of data analyses that include a complete elemental analysis of PM is rather limited. To bypass this problem of limited data availability, the strength of the correlation between tracers can be used for deriving an MD estimation that uses just two and sometimes only one element. Further in this chapter, a more elaborate overview is given of correlations between elements contributing to MD, according to various studies.

5.2 Collection of measurement data

An extended literature review was executed to collect observational data for mineral dust in PM in Europe. The list of articles used in our analysis consisted of 54 papers and/or reports (see Annex 3). Several authors were approached to supply us with any further or more detailed data, if these were available. The requested information focused on concentrations of PM_{10}, $PM_{2.5}$, MD, Al, Si, and Ti, and on additional information about the locations, such as type, longitude, latitude, altitude, and the analytical techniques applied for chemical analyses. The following authors kindly supplied us with their observational PM data and chemical speciation data:

- Xavier Querol (Institute of Earth Sciences Jaume Almera, ICTJA-CSIC, Barcelona, Spain),
- Jean-P Putaud (European Commission, Joint Research Centre, Institute for Environment and Sustainability, Ispra, Italy),
- Federico Mazzei (Physics Department, University of Genoa and Instituto Nazionale di Fisica Nucleare-Milano, Milan, Italy),
- Mustafa Koçak (Institute of Marine Sciences, Middle East Technical University, Erdemli-Mersin, Turkey),
- Jianxin Yin (Division of Environmental Health and Risk Management, School of Geography, Earth and Environmental Sciences, University of Birmingham, Edgbaston, Birmingham, the United Kingdom),

The other sources of information included:

- EMEP measurement data online (http://tarantula.nilu.no/projects/ccc/emepdata.html) with additional support by Wenche Aas (Norwegian Institute for Air Research NILU, Atmosphere and Climate Change Department)
- World Data Centre for Aerosols (http://wdca.jrc.ec.europa.eu/)
- The studies by VMM (2009); Quass *et al.*, 2009 and Visser *et al.*, (2001) as discussed in Chapters 2 and 3, were also included in the European analysis.

5.2.1 Limitations

Many studies that report on PM_{10} or $PM_{2.5}$ concentrations do not provide chemical composition of the PM mass. Especially information on the preferred tracer (Si) for MD is scarce. This severely limited the number of studies suitable for our purpose. However, even when information on a tracer was available, using a compilation of reported data had a number of important limitations:

- Some studies only presented aggregated results (e.g. estimated MD) without reference to the original data or how the MD estimate was derived. Some studies presented data for only one possible tracer. However, these data were also collected, since complete data reported over longer periods of time are scarce.
- Often the measurement periods would be short, campaign-like and would cover less than a year or even two seasons. Frequently, studies measured chemical speciation for only 1 to 2 weeks. However, to derive average annual contributions of MD to PM_{10}/$PM_{2.5}$ we needed representative data.
- Interannual variation in the contribution of MD to PM_{10}/$PM_{2.5}$ could be substantial, for example, because of extremely dry or wet years. However, because of the scarcity of suitable data all years were pooled.
- The studies were not uniform, and would use different approaches, methods and techniques. There was also a lack of consistency in nomenclature and samples size fractions.

5.3 Regional contribution of mineral dust to PM_{10} and $PM_{2.5}$

A first analysis of the factors controlling mineral dust contribution to PM_{10} and $PM_{2.5}$ was made by using earlier compilations by Querol *et al.* (2004) and Putaud *et al.* (2004) (Table 5-1). The data were sorted according to increasing contributions of MD and showed the coarse fraction ($PM_{2.5\text{-}10}$)

Contribution of mineral dust to $PM_{2.5\text{-}10}$ and $PM_{2.5}$ in Europe **Table 5.1**

Location type	Urban background			Rural background			Urban back ground			Rural background		
fraction	$PM_{2.5\text{-}10}$	MD	MD	$PM_{2.5\text{-}10}$	MD	MD	$PM_{2.5}$	MD	MD	$PM_{2.5}$	MD	MD
Country	[µg m^{-3}]		%	[µg m^{-3}]		%	[µg m^{-3}]		%	[µg m^{-3}]		%
Austria[a)]	-	-		4.0	0.6	15	-	-		20.0	0.4	2
Switzerland[b)]	6.7	1.7	25	2.4	1.1	46	18.5	1.3	7	8.1	0.8	10
Netherlands [a)]	8.4	2.0	24	-	-		35.8	2.9	8	-	-	
Belgium [b)]	7.0	2.0	29	-	-		26.0	3.0	12	-	-	
Italy [b)]	5.0	2.4	48	8.0	1.4	18	20.0	0.6	3	14.0	0.6	4
Germany [a)]	13.4	2.6	19	-	-		26.4	1.1	4	-	-	
United Kingdom [a)]	9.0	3.0	33	-	-		16.0	2.0	13	-	-	
Sweden	8.0	5.0	63	2.0	1.0	50	12.0	3.0	25	10.0	2.0	20
Spain [a)]	15.0	7.0	47	7.0	4.0	57	22.0	3.0	14	14.0	2.0	14
Average	9.1	3.2	36	4.7	1.6	37	22.1	2.1	11	13.2	1.2	10

[a)] Querol *et al.* (2004)
[b)] Putaud *et al.* (2004)

and fine fraction ($PM_{2.5}$) which, together, form PM_{10}. Rules of thumb for grouping the total collected data set are derived from Table 5-1.

- The average contribution of MD to PM_{10} was ~ 18% at both urban and rural locations. An important conclusion is that the contribution to $PM_{2.5}$ was also relevant and certainly not negligible.
- MD may dominate the coarse fraction of PM_{10}; on average, a third of coarse PM is mineral dust.
- The fraction of MD in coarse PM_{10} ($PM_{2.5\text{-}10}$ or PMc) was ~ 36% at both urban and rural locations.
- The fraction of MD in $PM_{2.5}$ was ~ 10% at both urban and rural locations.
- At urban background locations, the concentration of MD in PMc ranged from 1.7 to 7 µg/m³, and for $PM_{2.5}$ the range was from 0.6 to 3 µg/m³.
- Concentrations of MD in PM at urban locations were about double those at rural locations.
- There were also regional differences in MD. High contributions were observed in northern countries (caused by road gritting and use of studded tires), as well as in southern countries (due to relatively dry conditions and contribution of Saharan dust.

5.4 Results from literature review

A first set of data retrieved from the literature were the reported measurements on mineral dust (Table 5-2). These data were then aggregated further, as presented in Table 5-3. This section deals briefly with the aggregated data in Table 5-3; the country-specific data in Table 5-2 is discussed in one of the following sections.

The table shows that contributions of mineral dust to PM in the natural regions of northern Europe were low, both in absolute and relative terms. For rural locations, the concentration of MD were rather low in countries bordering the Atlantic (the United Kingdom, Ireland) and increased from 2 to 5 µg/m³ from northern Europe to southern Europe. Mineral dust in PM_{10} at urban background and traffic locations was significantly elevated compared to that in the rural background. Although the rural background concentrations in the United Kingdom and Ireland were somewhat lower, a difference for concentrations at urban and traffic locations was not observed, when compared to central Europe.
The contribution of MD to PM increased further at traffic locations, to as much as 17 µg/m³. In extreme situations, MD could dominate the PM_{10} concentration. The most remarkable increase in concentration levels was seen at northern European traffic locations during wintertime and/or the spring. This increase was related to road gritting and driving with studded tires (Larssen *et al.*, 1004).

5.4.1 Size fractionation of mineral dust in PM_{10}

Size fractionation of mineral dust is important because of the possible health relevance of the fine fraction. It is also important for separate $PM_{2.5}$ air-quality guidelines and reduction targets, and for predictive models because the smaller size fractions are transported over longer distances and have a longer lifetime due to a slower deposition rate. Therefore, a general relationship was derived for the amount of MD in $PM_{2.5}$ as a fraction of the MD in PM_{10}. This allowed estimation of the fraction of MD in $PM_{2.5}$ even when no measurement data on $PM_{2.5}$ were available. From the data collected from the literature (see Annex 3), we have taken all data on location for which mineral dust was reported in PM_{10} and $PM_{2.5}$ (Figure 5-1). The results indicated that the amount of MD in $PM_{2.5}$ at urban and rural locations was ~28 and ~34%, respectively. Especially for the urban locations, percentages varied considerably, but one should bear in mind that the data cover the entire European continent.

Variation in the size fractionation of mineral dust occurred not only geographically, but also over time. Looking at data for one location over a year, the fraction of MD in the $PM_{2.5}$ size range varied by as much as between 10 and 80 %, as was also the case for two locations in Spain, in Figure 5-2. In general, high MD concentrations in PM_{10} tended to have relatively low fractions in $PM_{2.5}$. So, the more important the MD contribution to PM_{10}, the less relevant the $PM_{2.5}$ fraction would be. This indicates that events of elevated MD contributions probably originated from nearby sources. In cases of no or little contribution from local sources, the MD in PM_{10} predominately originates from long-range transport. The relative contribution of MD to $PM_{2.5}$ than increased as this has a longer lifetime than MD in the coarser $PM_{2.5\text{-}10}$ size range. A compilation of the average annual data for Europe

Average PM_{10}/$PM_{2.5}$ and the absolute and relative contributions of mineral dust to PM_{10}, per country and location type, collected form the literature (data and references presented in Annex 3)

Table 5.2

		fraction	PM_{10}	MD in PM_{10}		$PM_{2.5}$	MD in $PM_{2.5}$		$MD_{2.5}$/ MD_{10}
ID	**Country**	**Location_type**	**[µg/m^3]**	**[µg/m^3]**	**[%]**	**[µg/m^3]**	**[µg/m^3]**	**[%]**	**[%]**
1	FIN	urban	21.10	6.62	31.39	8.30	0.53	6.40	8.01
		natural	4.50	0.30	6.64	3.41	0.15	4.28	50.00
2	SWE	urban	17.00	7.00	41.18	8.00	2.00	25.00	28.57
		rural	8.00	2.00	25.00	7.00	1.00	14.29	50.00
		traffic	26.00	17.00	65.38	13.00	4.00	30.77	23.53
3	NOR	natural	7.98	0.60	7.44	4.99	0.22	4.39	36.36
4	GBR	urban	25.00	5.00	20.00	23.50	1.30	7.22	40.00
		traffic	35.00	7.00	20.00	24.00	2.00	8.33	28.57
5	IRL	urban	23.00	4.10	17.96	12.10	0.50	4.14	12.46
		rural	10.40	1.00	9.62	6.00	0.20	3.33	20.00
		traffic	37.30	6.50	17.43	22.40	0.60	2.68	9.23
6	NLD	urban	27.85	3.75	13.79	20.40	0.69	3.49	19.93
		rural	23.23	3.93	16.55	15.50	1.05	6.62	27.80
		traffic	30.00	6.00	20.00	20.00	0.70	3.50	11.67
7	BEL	urban	32.03	2.94	9.15	22.45	0.83	3.61	27.94
		rural	39.55	3.76	9.51	25.58	0.98	3.82	26.06
8	CHE	urban	24.77	2.73	10.96	18.68	1.20	6.42	44.31
		rural	12.26	1.77	14.67	11.26	0.82	8.06	44.90
		traffic	37.96	5.31	13.69	22.31	1.26	5.72	29.38
9	DEU	urban	24.95	3.92	15.39	18.35	1.91	9.62	44.49
		rural	20.85	2.61	12.45	15.24	1.44	9.26	62.61
		traffic	37.00	14.00	37.84	25.00	3.00	12.00	21.43
10	AUT	rural	23.96	1.01	4.21	19.97	0.33	1.65	32.75
		traffic	53.79	4.94	9.16	38.26	1.54	4.00	28.90
11	CZE	urban	35.00	2.85	8.13	29.60	0.74	2.50	25.96
12	FRA	rural	---	---	---	9.52	1.05	11.00	---
13	GRC	urban	54.00	19.95	36.94	25.30	1.29	5.10	6.47
14	ITA	urban	44.95	9.05	20.57	39.42	2.46	6.99	31.19
		rural	27.17	5.80	21.78	22.60	1.90	8.46	29.66
		urban-industrial	41.70	3.30	7.91	19.00	3.20	16.84	96.97
		traffic	43.55	12.45	28.97	20.60	2.00	9.71	15.87
15	ESP	urban	34.90	9.07	25.92	22.40	2.80	12.58	30.73
		rural	18.65	4.28	22.96	13.82	1.39	9.89	38.98
		urban-industrial	36.95	11.29	31.03	25.72	3.64	13.72	29.41
		traffic	48.77	14.06	29.01	32.50	4.28	13.36	30.37

PM_{10} and $PM_{2.5}$ data from Table 5 2, aggregated per location type and region

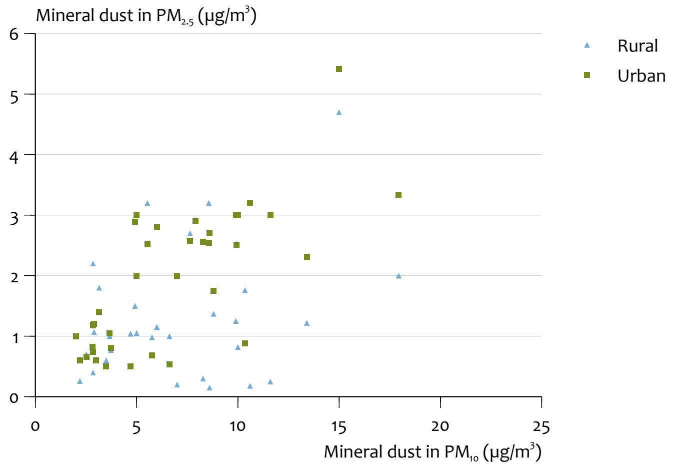
Table 5.3

Location type	Region[a)]	PM_{10} [µg/m^3]	CM_{10} [µg/m^3]	MD in PM_{10} [%]	$PM_{2.5}$ [µg/m^3]	$MD_{2.5}$ [µg/m^3]	MD in $PM_{2.5}$ [%]	CM2.5/CM10 [%]
NATURAL	North	6.2	0.5	7.0	4.2	0.2	4.3	43
	Atlantic	18.6	1.2	6.5	7.4	0.3	4.1	25
RURAL	North	8.0	2.0	25.0	7.0	1.0	14.3	50
	Central	24.0	2.6	11.5	16.2	0.9	6.7	39
	South	22.9	5.0	22.4	18.2	1.7	9.2	34
	Atlantic	10.4	1.0	9.6	6.0	0.2	3.3	20
URBAN	North	19.1	6.8	36.3	8.2	1.3	15.7	18
	Central	28.9	3.2	11.5	21.9	1.1	5.1	33
	South	44.6	12.7	27.8	29.0	2.2	8.2	23
	Atlantic	24.0	4.6	19.0	17.8	0.9	5.7	26
TRAFFIC	North	26.0	17.0	65.4	13.0	4.0	30.8	24
	Central	39.7	7.6	20.2	26.4	1.6	6.3	23
	South	46.2	13.3	29.0	26.6	3.1	11.5	23
	Atlantic	36.2	6.8	18.7	23.2	1.3	5.5	19

[a)] North = FIN, SWE, NOR; Atlantic = GBR, IRL; Central = NLD, BEL, CHE, DEU, AUT, CZE, FRA; South = GRC, ITA, ESP.

Relation between mineral dust in PM_{10} and $PM_{2.5}$ for rural and urban locations across Europe

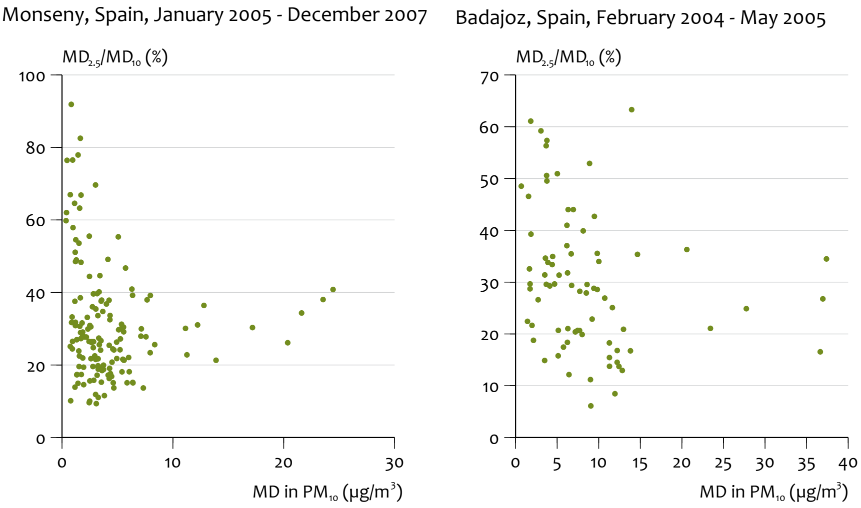
Figure 5.1



Relation between mineral dust in PM_{10} and $PM_{2.5}$ for rural and urban locations across Europe (one outlier removed from the urban data set) For specifics on the locations, see Annex 3.

The fraction of $PM_{2.5}$ in mineral dust PM_{10}

Figure 5.2


Monseny, Spain, January 2005 - December 2007

Badajoz, Spain, February 2004 - May 2005

The fraction of $PM_{2.5}$ in crustal PM_{10} at two Spanish locations; urban (left), rural (right). Data from Querol et al. (X. Querol, pers. comm., 2009)

(Figure 5-3) presents a comparable pattern of the Dutch data analysed in Chapter 2 (Figure 2-7, Figure 2-8). However, the results are more obscure, because of averaging over time and locations. It is difficult to distil an average MD $PM_{2.5}$ fraction from Figure 5-3, but it is thought to be ~ 30%.

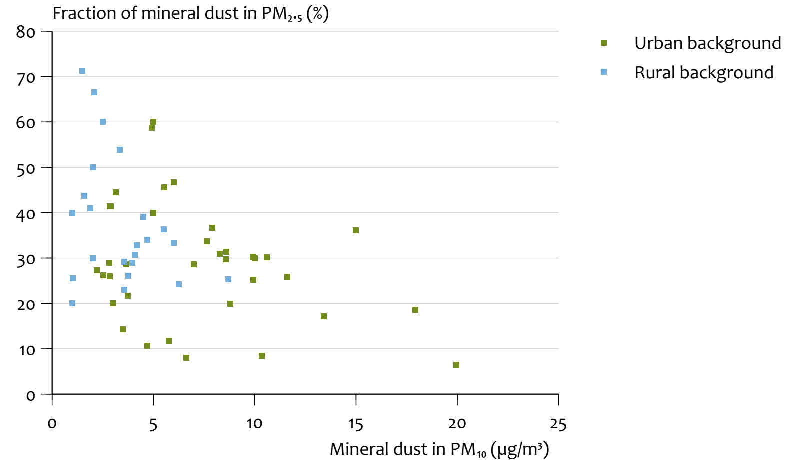
5.4.2 Validity of using tracers to calculate mineral dust

Among the characteristic elements for mineral dust are Al, Si, Ti, Fe and Ca, and we specifically searched for PM measurements with chemical analysis including one or more of these elements. To verify if the elements indeed represent mineral dust, we looked at correlations between the tracer elements. The assumption was that if these elements overwhelmingly would come from mineral dust, they would occur at a constant ratio. However, this ratio may vary geographically as the composition of mineral dust varies geographically. A striking good correlation between Si and Al in PM_{10} could be found all over Europe, for example, for Italy (Figure 5-4) and Belgium (Figure 5-5). Titanium is also strongly correlated with Al and Si and seems to have the same crustal origin. This was also shown in earlier BOP data (Figure 2-2) and in the 1998-1999 Dutch data from Visser *et al.*, (2001; data not shown). For particular locations, we subsequently observed good correlations between other constituents of mineral dust, such as Ti in Spain

The above data from individual locations have been shown to illustrate that, for individual locations, good correlations could be observed between elements associated with mineral dust, especially Si, Al, Ti, Fe and Ca (not shown). However, when all collected data were put together, it became evident that Ca and Fe are less reliable tracers. This could be explained by other sources which emit PM containing these elements, or by a high variability of the element in mineral

The fraction of mineral dust $PM_{2.5}$ in average mineral dust PM_{10} for ~60 locations in Europe

Figure 5.3

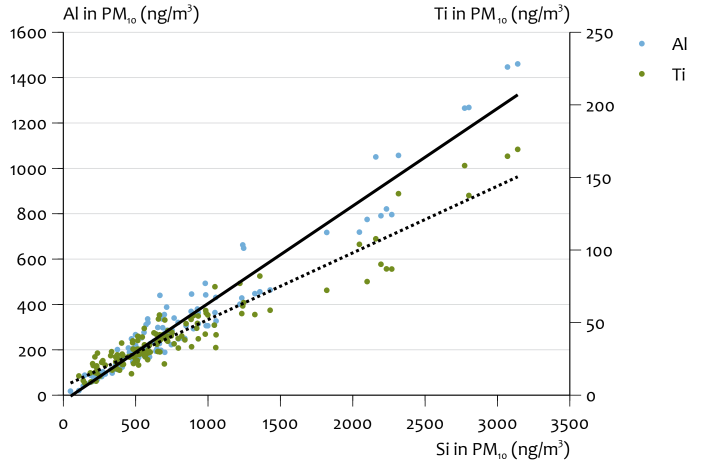


The fraction of crustal PM2.5 in average crustal PM10 for ~60 locations in Europe.

Al and Ti as a function of Si in PM_{10} for an urban site in Italy

Figure 5.4

Brignole, Italy, May 2002 - June 2006



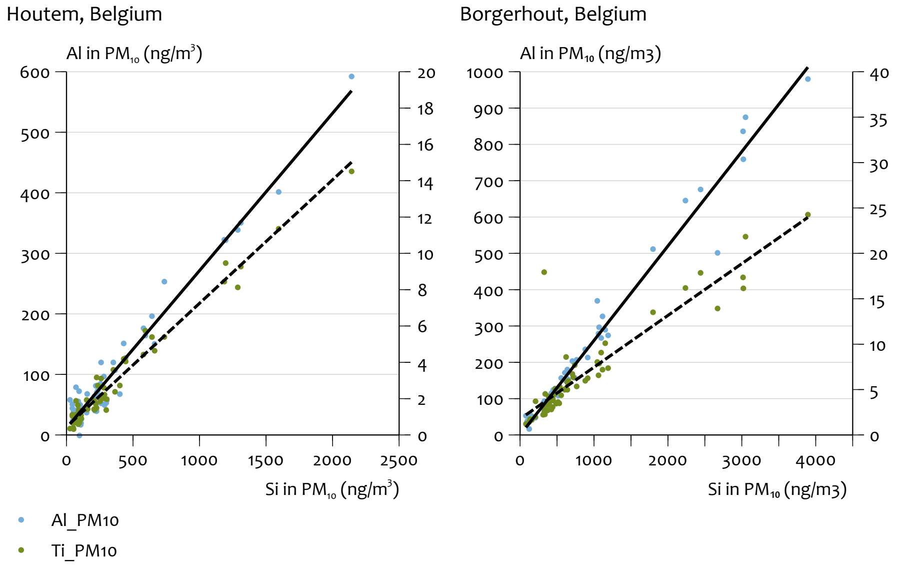
PM10 Al and Ti concentrations as a function of Si for a urban location in Italy (data from F. Mazzei, pers.comm, 2009).

dust. For example, Ca concentrations are high in calcareous materials, the levels of which may have been locally elevated, for example, due to liming of fields and/or the presence of cement factories. Fe is also related to the wear of metal parts, such as in engines and breaks, and is therefore less unique to mineral dust (see also Section 2.1).

The average PM_{10} concentrations of Si, Al, Ti and Fe across Europe and all locations have been compiled in Figure 5-8. Even at this aggregated level, Al and Si showed a strong correlation.. The correlation with Ti was less robust, but for PM_{10} still present. However, when looking at $PM_{2.5}$ (Figure 5-9), the Ti concentrations did not correlate well with Si. This could be explained by the fact that Ti in $PM_{2.5}$ could also have originated from the combustion of heavy fuel oil in shipping and refineries. Furthermore, some Ti may also have been released from mechanical wear of metal engine parts and brakes. The patterns depicted in Figure 5-8 and Figure 5-9 further proved that Si and Al could be considered the tracers of choice for mineral dust. At individual locations other elements could be useful and especially in the coarse fraction Ti may be an alternative. The other important consideration was that Al and Si would dominate the mineral dust weight, hence, lessening the uncertainty in their analysis than if we had used elements present in much lower concentrations.

To derive mineral dust from the concentrations of the tracer elements we used Equation 2.1 (see Section 2.1). An alternative approach would have been to take all studies that have reported MD in PM_{10} and also reported an analysis on Al and/or Si (and Ti for comparison). Obviously, some of these studies indeed used Al and/or Si to estimate MD, but they may have used locally derived relationships and may have included other elements that locally showed a good correlation with MD, such as Fe, Ca, Mg, or Ti. The relationship between MD

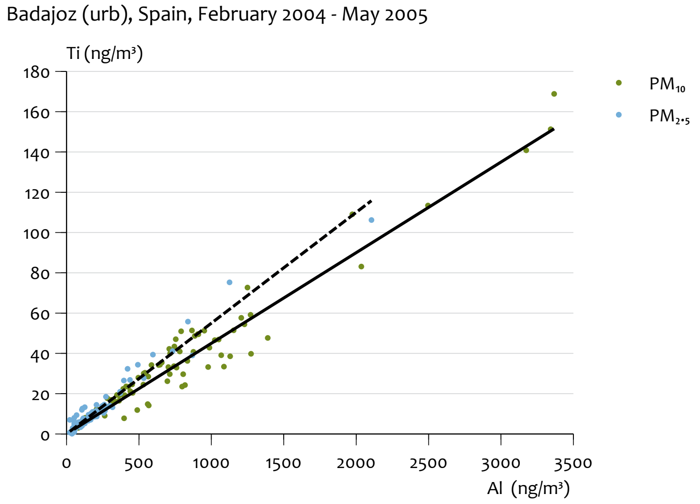
Al and Ti as a function of Si in PM_{10} for a rural site and street site in Belgium, september 2006-september 2007 Figure 5.5


Houtem, Belgium

Borgerhout, Belgium

PM_{10} Al and Ti concentrations as a function of Si for a rural location (Houtem) and a traffic location (Borgerhout) in Belgium (VMM, 2009)

Ti as a function of Al in PM_{10} and $PM_{2.5}$ for an urban site in Spain Figure 5.6


Badajoz (urb), Spain, February 2004 - May 2005

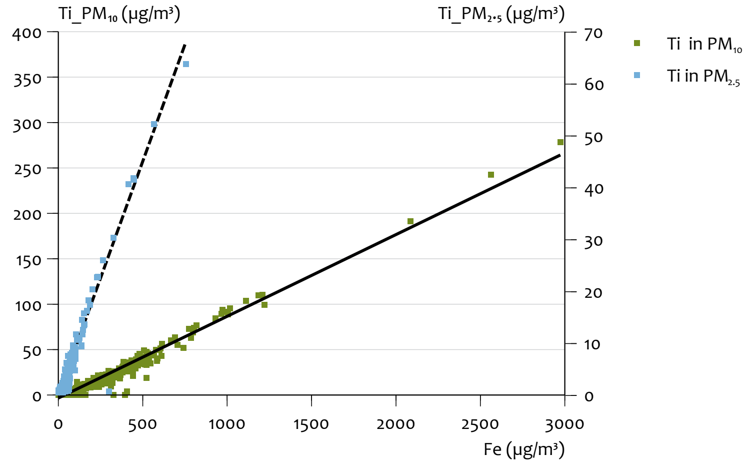
PM_{10} and $PM_{2.5}$ Ti concentrations as a function of Al for an urban location in Spain (data X. Querol, pers comm., 2009).

in PM_{10} and Al and Si concentrations as reported by various European studies are depicted in Figure 5-10.

Correlation between Fe and Ti at a rural site in Turkey

Figure 5.7

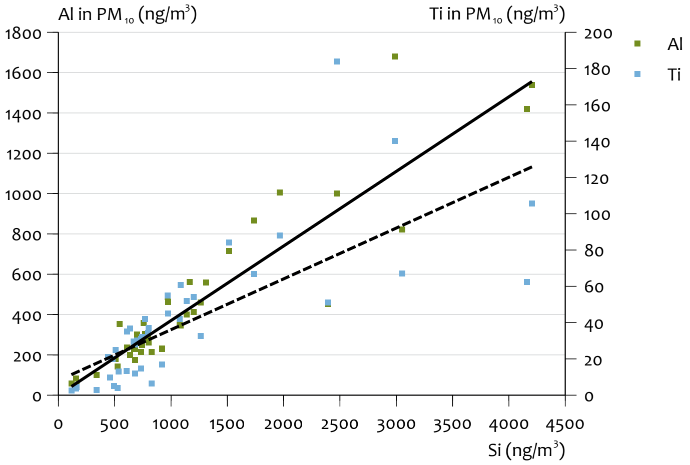
Erdemeli, Turkey, April 2001 - May 2002



Correlation between Fe and Ti at a rural location in Turkey (data from M. Kocak, pers. comm.).

Correlation between Si, Al and Ti in PM$_{10}$ for all location types across Europe and accross years

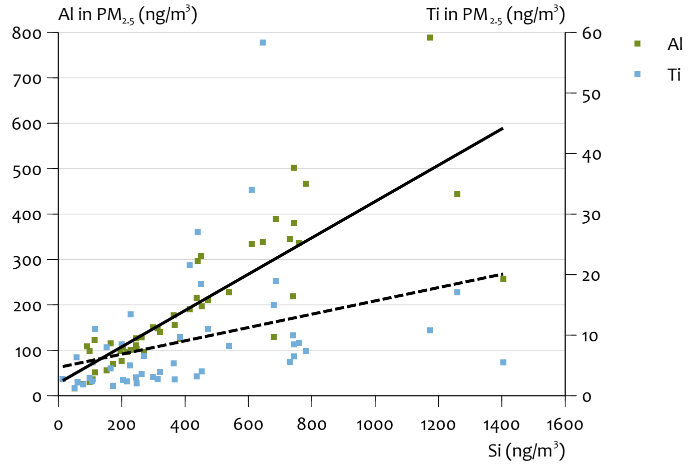
Figure 5.8



PM$_{10}$ Al and Ti concentrations as a function of Si for all location types across Europe and over time.

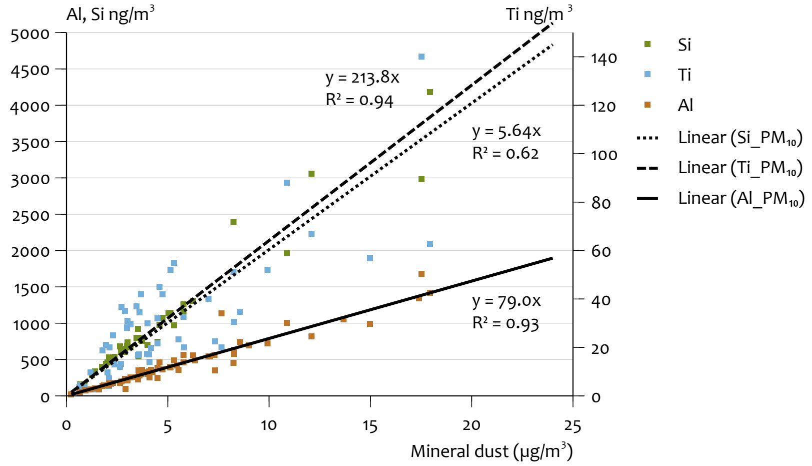
Correlation between Si, Al and Ti in PM$_{2.5}$ for all location types across Europe and across years

Figure 5.9



PM$_{2.5}$ Al and Ti concentrations as a function of Si for all location types across Europe and over time.

Correlation between reported Mineral dust and concentrations of Al, Si, and Ti in PM_{10} **Figure 5.10**



Correlation between reported MD in PM_{10} and concentrations of Al, Si and Ti in PM_{10}. (Note the different units along the X and Y axes.).

5.5 Mineral dust in $PM_{2.5}$ in European cities

An independent source of information is a dataset of ambient $PM_{2.5}$ samples collected at 21 European cities, analysed for elemental composition by Götschi *et al.* (2005). These data were collected in the framework of the European Community Respiratory Health Survey (ECRHS) in a coordinated effort ensuring that sample collection, methodology and timing would be comparable between the locations. Fine particles ($PM_{2.5}$) were sampled over a year (June 2000-December 2001) at 21 central urban monitoring locations (20 cities) over all of Europe (Figure 5-11).

This study offered the possibility to derive comparable estimates for MD in $PM_{2.5}$ in urban centres across Europe. Furthermore, we could derive crustal PM in $PM_{2.5}$ and PM_{10} by using the relationships described in the previous sections to estimate urban MD in PM_{10}/$PM_{2.5}$ for our study, and compare the results with patterns derived from the literature review in Section 5.4. Götschi *et al.* (2005) analysed $PM_{2.5}$ filter samples for elemental composition using energy dispersive X-ray fluorescence spectrometry and reflectance. The average annual concentration levels for $PM_{2.5}$, Al, Si, Ti, Ca, K, and Fe, for all ECRHS locations are presented in Table 5-4.

A considerable range in the average annual concentrations was found for all indicators (Table 5-4; $PM_{2.5}$, Al, Si and Ti). However, similar pollution levels were observed for locations within the same country or regions. The highest concentrations of $PM_{2.5}$ were observed in Italy (Turin, Verona, Pavia) and the lowest in northern Europe (Reykjavik, Umea). The contribution of mineral dust elements to $PM_{2.5}$ showed a seasonal pattern at some locations. For example, the levels at Spanish locations were elevated in the summer, whereas in for instance Reykjavik this occurred in the winter. This is most likely related to seasonal source contributions from desert dust and road gritting, respectively. For a more detailed description of the data we refer to Götschi *et al.* (2005).

To estimate MD in $PM_{2.5}$ for urban centres from the ECRHS data, we first verified the relationship between Si, Al and Ti (Figure 5-12). This confirmed that also in $PM_{2.5}$ Al and Si were highly correlated and supported our approach of basing MD estimates on the concentrations of Si and Al. The data for Grenoble were an outlier in the dataset: with an extremely high concentration of Si (1404 ng/m³) but the Al concentration did not differ much from the reported overall average (Table 5-4). The Si–Al ratio for Grenoble was 10 where the average for all others locations was 2.3 (ECRH II, 2004). Therefore, Grenoble was excluded from the correlation in Figure 5-12. The data may have been correct for Grenoble, but did not seem suitable for deriving general relationships for MD in urban centres.

The mineral dust mass in $PM_{2.5}$ for the 21 European study centres, was calculated by using Equation 2.1. The mineral dust in $PM_{2.5}$ was calculated by using both trace elements (Al, Si) and only one of them (Al or Si) (data not shown). The calculations for which only one element was used, showed slightly different results, but most notably (and predictably) different for Grenoble, where Si and Al+Si resulted in extremely high concentrations of ~ 5 ug MD/m³ in $PM_{2.5}$, and when using Al estimates this was ~ 2 µg MD/m³ in $PM_{2.5}$. Since Al and Si were available for all sites and well-correlated, the most robust approximation was that from using both elements (Table 5-5). The data in Table 5-5 have been sorted according to increasing concentrations of MD in $PM_{2.5}$. Estimated average annual MD in $PM_{2.5}$ ranged between 0.7 and 4.8 µg/m³. The highest concentrations were calculated for southern Europe (Spain, Italy and France), the lowest for northern and Atlantic Europe (UK and Ireland). The relative contribution of MD to $PM_{2.5}$ ranged from ~5% (UK) to almost 30% (Spain). This indicated that, although most MD is in the coarse fraction, the contribution of MD to urban $PM_{2.5}$ can be significant.

The estimated contributions to $PM_{2.5}$ were extrapolated to PM_{10} by using three related approaches developed in Sections 5.3 and 5.4.1 of this report:.

Map of locations of the 21 study centres of ECRHS **Figure 5.11**

Map of locations of the 21 study centres of ECRHS, reported by Götschi et al. (2005).

1. The estimation of MD in PM_{10} that was based on the general assumption that ~ 27% of mineral dust was in the $PM_{2.5}$ fraction. This assumption is a crude summary of Figure 5-1.
2. The estimation of MD in PM_{10} that was based on country/region-specific information from Table 5-2.
3. The estimation of MD in PM_{10} using the trend line equation from Figure 5-1

Estimated MD in PM_{10} for the 21 European urban centres ranged between 2 and 18 µg/m³. This was slightly more extreme than was derived from the overall literature study (3-13 µg/m³), but was generally quite consistent. The data from the ECRHS study, therefore, could be added to our total set of data, significantly increasing the coverage of urban locations. In this manner, we compiled estimates of MD in PM_{10} for 19 European countries, often for more than one location within each country.

Average annual concentrations of $PM_{2.5}$ and selected trace elements at 21 European locations (data from Götschi et al., 2005)

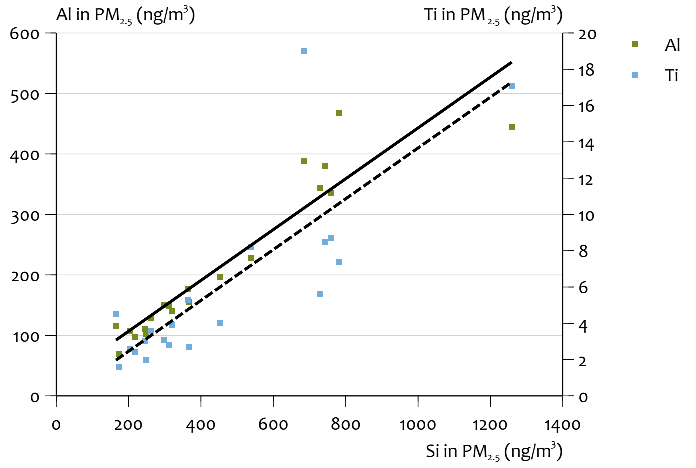
Table 5.4

Country	Location	$PM_{2.5}$ [µg/m3]	Al [ng/m^3]	Si [ng/m^3]	Ti [ng/m3]	Ca [ng/m^3]	K [ng/m^3]	Fe [ng/m^3]
BEL	Antwerp City *	24.1	177	363	5.3	87	182	127
BEL	Antwerp South	20.8	128	263	3.6	46	182	66
CHE	Basel	17.4	151	299	3.1	60	255	78
DEU	Erfurt	16.3	148	313	2.8	52	157	72
ESP	Albacete	13.1	344	730	5.6	277	350	49
ESP	Barcelona	22.2	389	686	19	226	430	145
ESP	Galdakao *	16.3	197	453	4	199	191	166
ESP	Huelva	17.3	444	1259	17.1	168	297	76
ESP	Oviedo	15.9	467	781	7.4	281	232	138
EST	Tartu	14.8	156	367	2.7	85	386	32
FRA	Grenoble	19	257	1404	5.5	139	326	125
FRA	Paris	17.8	141	321	3.9	79	180	98
GBR	Ipswich	16.5	115	165	4.5	37	201	41
GBR	Norwich *	16.2	108	204	2.6	92	116	42
ISL	Reykjavik	3.7	111	245	3	41	29	23
ITA	Pavia	35.3	228	539	8.2	85	364	124
ITA	Turin	44.9	380	744	8.5	116	471	262
ITA	Verona **	41.5	336	759	8.7	257	411	302
SWE	Gothenburg	12.6	97	217	2.4	38	113	52
SWE	Umea	5.6	70	172	1.6	22	63	25
SWE	Uppsala	10.4	103	247	2	34	116	54

* Winter mean based on < 80% of scheduled sampling time (Antwerp City: 60%, Galdako: 66%, Norwich: 64%).
* * Winter and summer mean based on less than 80% of scheduled sampling time (43 and 18%, respectively).

Correlation between Al, Ti and Si in $PM_{2.5}$

Figure 5.12



Al–Si and Si–Ti correlations (without the Grenoble location), data from Götschi et al. (2005).

5.6 Data for model validation

Source functions for MD sources were implemented in the LOTOS-EUROS model (Schaap *et al.*, 2009). To validate the model-predicted MD concentrations, we also collected daily measurement data on elements in PM_{10} and $PM_{2.5}$. A compilation was made with coordinates of the sample locations and estimated MD concentrations. The coverage of the data points is shown in Figure 5-13. The use of this data set is presented in Chapter 6.

Table 5.5

Calculated concentrations of MD in $PM_{2.5}$, based on measured Al and Si concentrations in $PM_{2.5}$, and estimated MD in PM_{10} according to the use of three different methods.

			Mass MD = 0.49*[Si]+(2.36*[Si]+2.70*[Al])		Based on the graph result, that 27% of mineral dust is in $PM_{2.5}$	Based on country-specific information	Based on the graph result, on the function $MD_{2.5}=0.22[CM_{10}]+0.46$
Country	**Location**	**$PM_{2.5}$ [μg/m³]**	**Mass MD (Al&Si) [μg/m³]**	**MD in $PM_{2.5}$ [%]**	**MD in PM_{10} [μg/m³]**	**MD in PM_{10} [μg/m³**	**MD in PM_{10} [μg/m³]**
SWE	Umea	5.6	0.68	12.13	2.52	2.38	1.00
GBR	Ipswich	16.5	0.78	4.73	2.89	1.95	1.46
GBR	Norwich *	16.2	0.87	5.39	3.23	2.18	1.88
SWE	Gothenburg	12.6	0.88	6.99	3.26	3.08	1.91
SWE	Uppsala	10.4	0.98	9.44	3.64	3.44	2.37
ISL	Reykjavik	3.7	1.00	26.97	3.70	3.80	2.45
BEL	Antwerp South	20.8	1.10	5.27	4.06	3.92	2.89
CHE	Basel	17.4	1.26	7.24	4.67	2.84	3.64
DEU	Erfurt	16.3	1.29	7.92	4.78	2.90	3.78
FRA	Paris	17.8	1.30	7.28	4.80	3.91	3.80
EST	Tartu	14.8	1.47	9.91	5.43	4.42	4.58
BEL	Antwerp City *	24.1	1.51	6.28	5.60	5.41	4.78
ESP	Galdakao *	16.3	1.82	11.18	6.75	5.93	6.20
ITA	Pavia	35.3	2.15	6.10	7.97	6.90	7.69
ESP	Albacete	13.1	3.01	22.97	11.15	9.79	11.59
ESP	Barcelona	22.2	3.01	13.54	11.13	9.78	11.57
ITA	Verona * *	41.5	3.07	7.40	11.37	9.84	11.87
ITA	Turin	44.9	3.15	7.01	11.65	10.09	12.21
ESP	Oviedo	15.9	3.49	21.93	12.91	11.34	13.76
FRA	Grenoble	19.0	4.70	24.71	17.39	14.16	19.25
ESP	Huelva	17.3	4.79	27.67	17.73	15.58	19.67

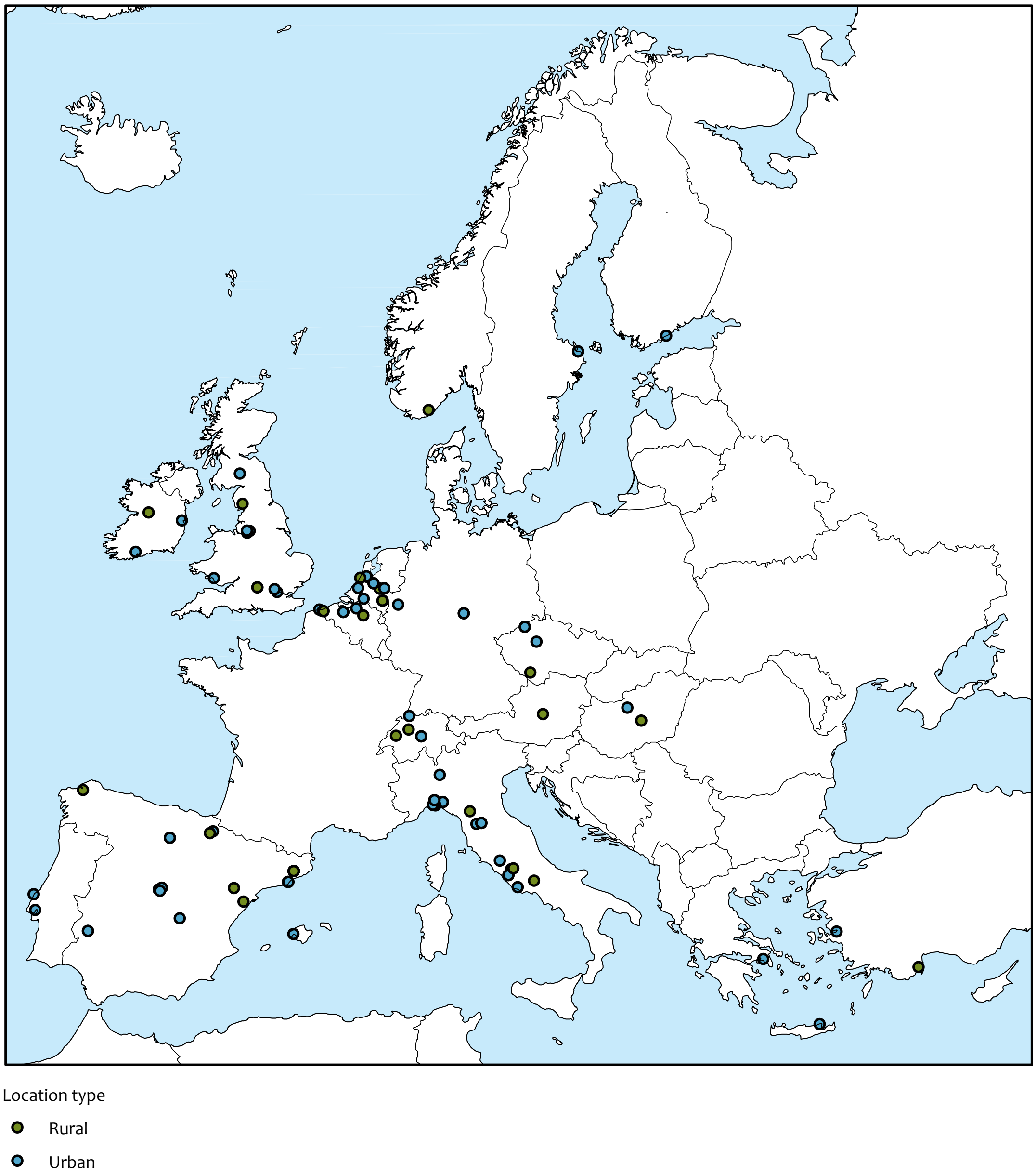
* Winter mean based on less than 80% of scheduled sampling time (Antwerp City 60%, Galdako 66%, Norwich 64%.

* * Winter and summer mean based on less than 80% of scheduled sampling time (43% and 18%, respectively).

FRA- the average for the central region (0.33) was used; ISL- the average for the UK and Ireland (0.26) was used; EST- the average for the central region (0.33) was used.

Rural and urban locations for which information on mineral dust in PM_{10} was available

Figure 5.13



Rural and urban locations for which information on mineral dust in PM_{10} was available. Data did not necessarily cover the same year or season.

6

Comparison between modelled MD and observational data, for Europe

The LOTOS-EUROS model is a regional model. Therefore, it was necessary to compare the results with observations at regional background locations, as urban locations would be influenced too much by local sources. To verify if the results from the modelling of MD in PM_{10} across Europe, as outlined in Chapter 4, approached realistic values, we compared the modelled concentrations with observed MD in PM_{10} for a number of well-established regional background locations across Europe (Table 6-1).

The contribution of various sources of MD to the total modelled MD at the locations listed in Table 6-1 is discussed in Section 6.1. This gives an impression of the relative importance of various sources across Europe. A first comparison of model results with observational data was published in Schaap *et al.* (2009). Based on these results, some modifications have been made to the source functions, as described in Section 6.2. The results from the improved source functions are discussed further in Section 6.3.

6.1 Seasonal variation

The seasonal variation in mineral dust emissions and the resulting ambient concentrations are specific to each source. The concentration of material resuspended due to road traffic (Figure 6-1), was relatively constant throughout the year, with <0.4 $\mu g.m^{-3}$ at rural locations and between 2.5 and 3.5 $\mu g.m^{-3}$ at Montseny in Spain. Although Montseny was classified as a rural/regional location, it is located close to Barcelona. As the grid resolution of the model was rather coarse, the modelled atmospheric mineral matter concentration, therefore, would have been strongly influenced by traffic emissions in Barcelona, and is not thought to present an accurate picture for this rural location. The seasonal variation in the traffic-induced mineral dust concentrations was mainly due to a variation in meteorological parameters (e.g. precipitation, boundary layer height, horizontal advection). The effect from the variation in mixing layer height seemed to be the most important. In summer, dust concentrations were lower than in winter, when the mixing layer would be relatively shallow,

Average annual mineral dust contribution to PM10 for selected locations in Europe, as calculated with the LOTOS-EUROS model, and as found on basis of observations **Table 6.1**

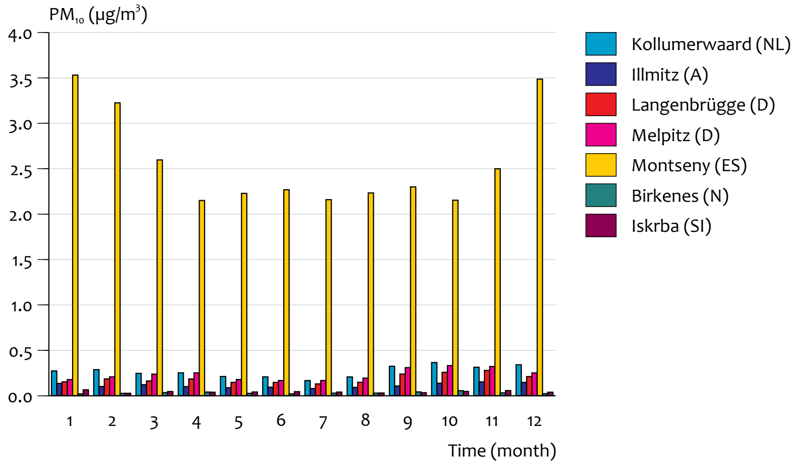
				Average annual mineral dust contribution to PM_{10}					
				Model				**Observations[1]**	
Location name	**Longitude (deg. E)**	**Latitude (deg. N)**	**Location type**	**Traffic ($\mu g.m^{-3}$)**	**Wind ($\mu g.m^{-3}$)**	**Agriculture ($\mu g.m^{-3}$)**	**Total ($\mu g.m^{-3}$)**	**Ca ($\mu g.m^{-3}$)**	**Total ($\mu g.m^{-3}$)**
Kollumerwaard (NL)	6.28	53.33	rural	0.27	0.06	0.10	0.42	0.09	0.71[2]
Illmitz (A)	16.36	48.23	rural	0.11	0.07	0.85	1.03	0.19	1.39[2]
Langenbrügge (D)	10.76	52.8	rural	0.19	0.13	0.71	1.01	0.14	1.07[2]
Melpitz (D)	12.93	52.53	rural	0.23	0.09	0.78	1.09	0.14	1.03[2]
Montseny (ES)	2.38	41.78	rural	2.55	0.07	0.53	3.17	0.38	3.57
Birkenes (N)	8.25	58.38	rural	0.03	0.01	0.05	0.10	0.06	0.46[2]
Ikrba (SI)	14.57	45.57	rural	0.04	0.00	0.16	0.21	0.13	0.97[2]

[1] the observations were taken from the EMEP database (http://emep.int), with the exception of the observations of Montseny in Spain, which were obtained from Querol *et al.* (pers.comm.) and corresponded to the observations discussed in Querol *et al.* (2007).

[2] total MD in PM_{10} inferred from the elemental Ca concentration by assuming a ratio of 7.5 between the Ca and the mineral dust concentration in PM_{10}.

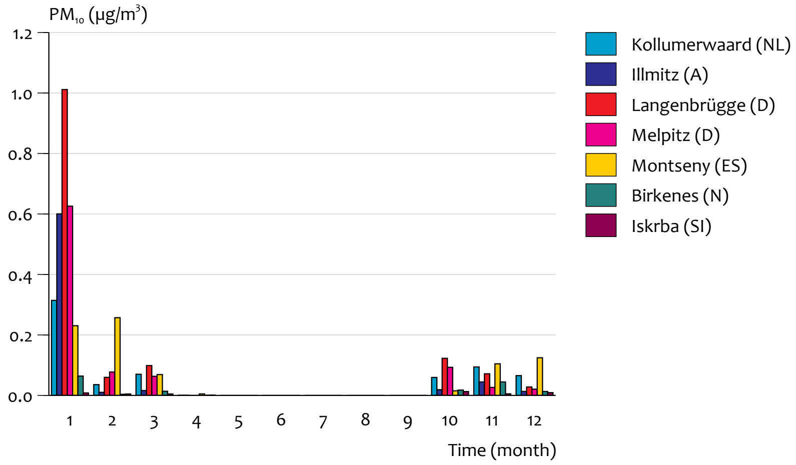
Note – values in this table are different from those in the related BOP report Schaap *et al.* (2009).

Traffic resuspended mineral dust **Figure 6.1**



Seasonal variation for 2005 in the modelled contribution of resuspended dust caused by road traffic, to mineral PM_{10} in Kollumerwaard (the Netherlands), Illmitz (Austria), Langenbrügge (Germany), Melpitz (Germany), Montseny (Spain), Birkenes (Norway), and Iskrba (Slovenia).

Wind-blown mineral dust **Figure 6.2**



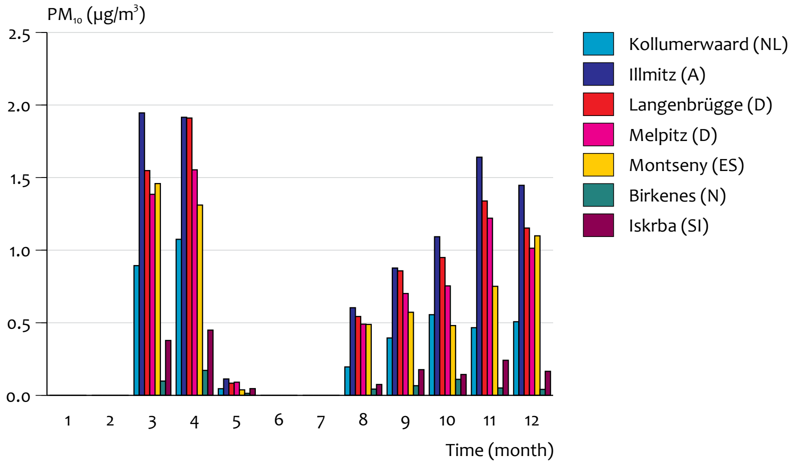
Seasonal variation for 2005 in the modelled contribution of wind-blown dust to mineral PM_{10} in Kollumerwaard (the Netherlands), Illmitz (Austria), Langenbrügge (Germany), Melpitz (Germany), Montseny (Spain), Birkenes (Norway), and Iskrba (Slovenia).

despite the fact that increased precipitation during the autumn and early winter would limit the dust production and increase the removal of ambient particles. For traffic-induced resuspension, the seasonal dependence was less pronounced than for wind or agricultural activities, which also showed a different timing of enhanced dust concentration.

The modelled concentrations of soil dust resuspended by wind (Figure 6-2) revealed the occurrence of high average wind speeds (in January 2005) which caused the dust production to increase tenfold or more, compared to the average annual production. In addition, seasonal variation would cause low concentrations of soil dust during the growing season, when most of the arable land would be covered by vegetation. Therefore resuspension from arable land in spring and summer was neglected in the present model setup and ambient soil dust concentrations dropped to zero in the spring and remain there throughout the summer.

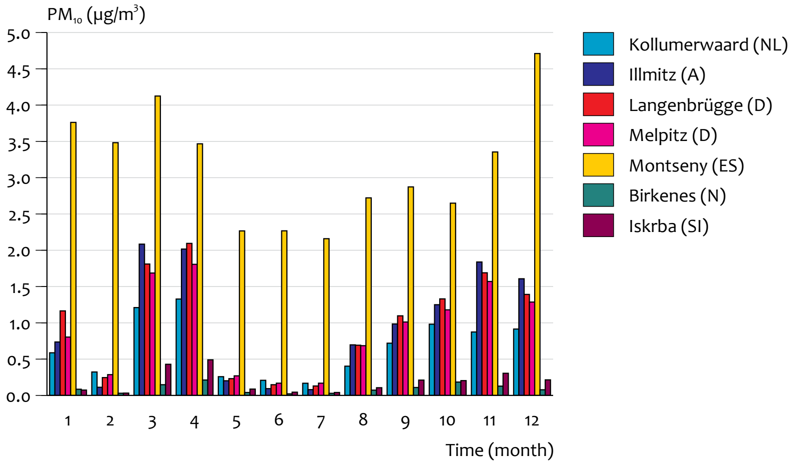
For land management induced resuspension, the emission times and associated periods with high concentrations were linked to the agricultural activity calendar (Table 4-2).. Concentrations peaked in the spring months, with values of up to 1.5 and 2 µg.m^{-3} in the German and Austrian locations (see Tables 5 and 13) and are also of significant magnitude in the autumn and early winter. However, in the growing season and in the middle of winter, when there would be no land management activity, concentration levels dropped to zero.

Agricultural dust **Figure 6.3**



Seasonal variation for 2005, in the modelled contribution of dust from agricultural land management activities to mineral PM$_{10}$ in Kollumerwaard (the Netherlands), Illmitz (Austria), Langenbrügge (Germany), Melpitz (Germany), Montseny (Spain), Birkenes (Norway), and Iskrba (Slovenia).

Total modelled mineral dust **Figure 6.4**



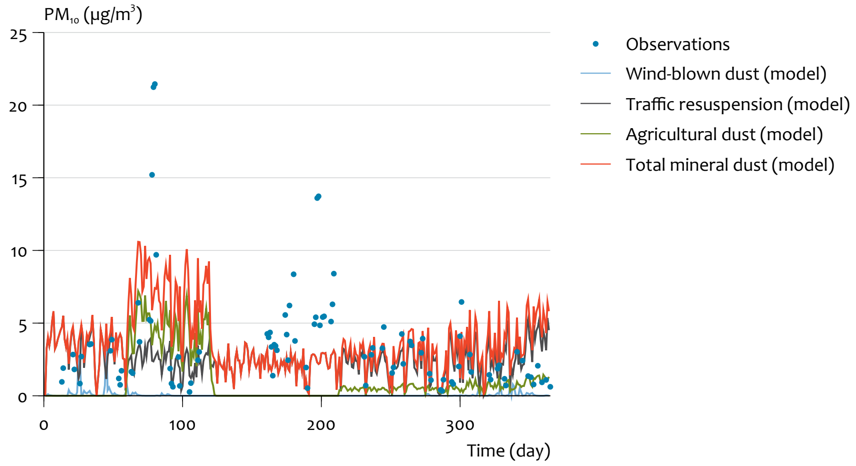
Seasonal variation for 2005 in the total modelled mineral PM$_{10}$ concentration levels in Kollumerwaard (the Netherlands), Illmitz (Austria), Langenbrügge (Germany), Melpitz (Germany), Montseny (Spain), Birkenes (Norway) , and Iskrba (Slovenia).

6.2 Verification

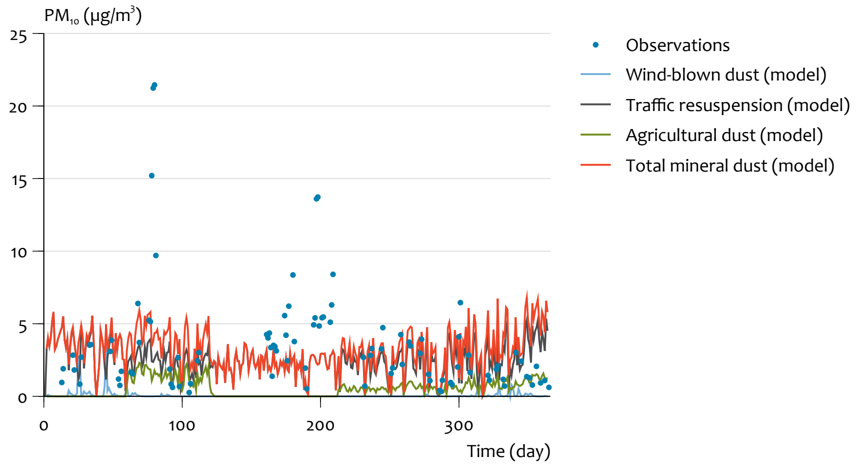
The model calculations were compared to the available data on observed mineral PM$_{10}$ concentrations. An extensive verification has been presented in Schaap *et al.* (2009) and for a discussion on verification we refer to this report. However, an important modification on Schaap *et al.* (2009) is the revision of the source function for MD emissions from agricultural land management. This significantly improved the match between modelled and observed concentrations (Figure 6-5 and Figure 6-6).

One of the existing data sets of time series on mineral dust, includes data on observations at a few locations in Spain (Querol *et al.*, 2007). Figure 6-5 shows a comparison between observed mineral dust and calculations with the LOTOS-EUROS model, for the rural location of Montseny, which is close to Barcelona. The reported mineral dust concentrations were calculated by the authors, based on the elemental composition of PM$_{10}$. In Montseny, model concentration levels equalled the levels of observed mineral dust. The average annual concentrations from both the model and observations was ~3.5 µg.m^{-3} (see Table 6-1). Furthermore, the values and the time of the variations in the modelled concentration levels were in agreement with those observed for the month of February and of September to November. For December to January, the modelled concentration levels were overestimated by the model, which was mainly due to an overestimation of the resuspension due to road traffic (see Figure 6-5). However, this may have been caused by the fact that Montseny is very close to Barcelona, and by the relatively coarse grid-cell resolution of the model (approximately

Mineral dust Montseny 2005 **Figure 6.5**



with new agricultural emission factor

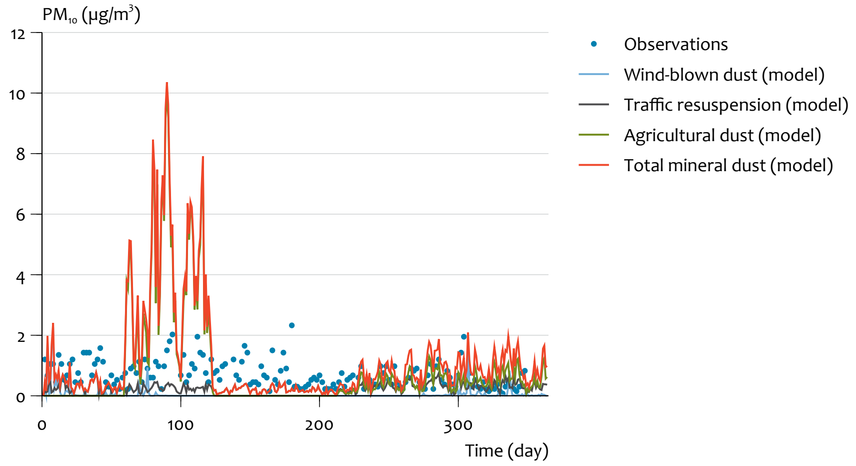


Time series of the observed and modelled contribution of mineral matter to PM_{10} in Montseny (Spain) in 2005, before (upper figure) and after adjustment of the emission factor for agricultural management (lower figure).

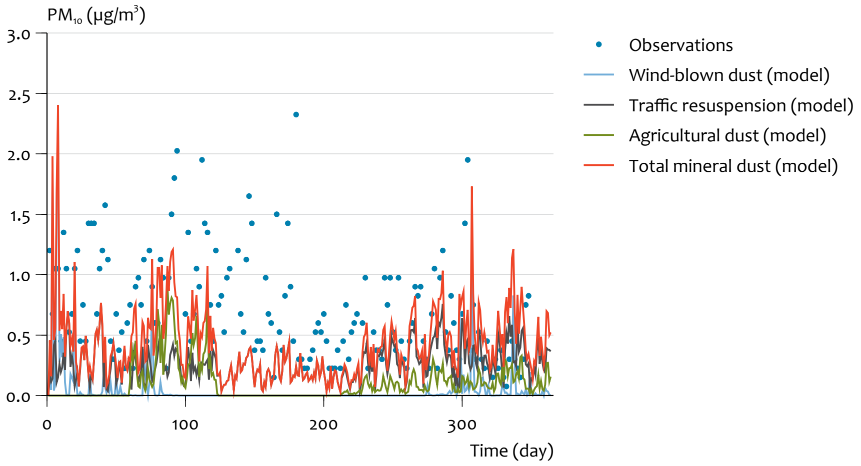
30 x 30 km at this latitude). The concentrations modelled for Montseny may have been too heavily influenced by resuspension emissions from the road traffic in Barcelona. Moreover, in the month of March, and towards the end of June and at the beginning of July, very high concentration levels were observed, which were not captured in the model calculations. These so-called dust events corresponded to an inflow of desert dust originating from the Sahara, the time of occurrence of which was evidenced by the model calculations carried out solely with the boundary input of mineral dust from the TM5 model (Schaap *et al.*, 2009). Furthermore, the contribution of agricultural emissions, as calculated with the adjusted emission factor for ploughing activity, for the spring season may still appear slightly overestimated, but the unrealistic increase in model-predicted concentration levels of mineral dust in the spring as reported by Schaap et al. (2009) was no longer present (compare Figure 6-5 upper and lower panel).

For the Netherlands we investigated the time series of the modelled mineral dust concentration at Kollumerwaard (Figure 6-6). The mineral dust concentration in PM_{10} was inferred from calcium (Ca) observations by deriving an empirical factor from the ratio between ambient concentrations of mineral dust and elemental Ca (Schaap *et al.*, 2009). This empirical factor with a value of 7.5 was derived from observations of both MD and Ca concentrations in De Zilk collected by Visser *et al.* (2001) for the 1998-1999 period. We assumed the average annual ratio between ambient Ca and mineral PM concentrations at Kollumerwaard to be the same as in De Zilk. The mineral PM_{10} time series obtained through the application of this multiplication factor to the Ca observations, again showed an overestimation by the model. This could mainly be attributed to the modelled contribution from agricultural land management, that is, the peak of agricultural dust in the spring shown by the model, was not seen in the observations. However, after a downward adjustment of the ploughing emission factor

Mineral dust Kollumerwaard 2005 **Figure 6.6**



with new agricultural emission factor



*Time series of the mineral dust contribution to PM_{10} for 2005, inferred from Ca observations by assuming a PM_{10}_MD=7.5*PM_{10}_Ca in Kollumerwaard (the Netherlands), before (upper figure) and after adjustment of the emission factor for agricultural activity (lower figure) (Note the different scales of the Y axes).*

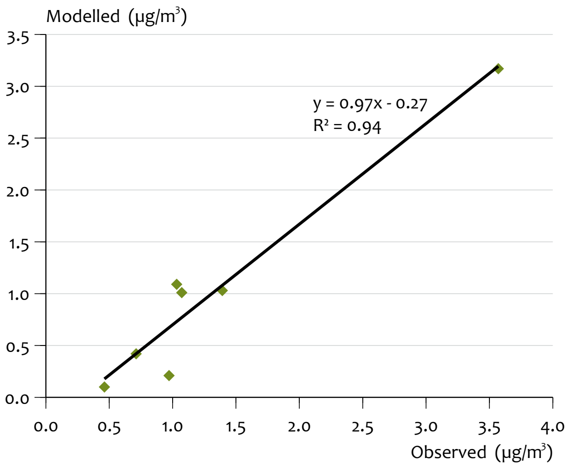
(of emission under moist soil conditions), the modelled mineral dust concentrations improved significantly towards more realistic concentration levels (Figure 6-6 lower figure). The modelled mineral dust contribution showed a broad agreement with the observations, although the spring season concentration was still slightly overestimated (note the different scales of the y axes in the upper and lower part of Figure 6-6. The latest results as shown in Figure 6-6 suggest that we have underestimated the contribution from diffuse sources in the drier periods of the year (summer) when there is no a agricultural management activity, however, we lack an alternative source causing some additional mineral dust release in the rural environment.

6.3 Comparison with observational data across Europe

The model-predicted average annual concentrations of MD in PM_{10} for seven locations across Europe showed a good correlation, when we (empirically motivated) replaced the emission factor for dry ploughing used in Schaap *et al.* (2009) with the lower emission factor for wet ploughing (Figure 6-7). The result indicated that between approximately 0.3 and 0.5 µg/m³ needed to be added to the modelled concentrations (the intercept in Figure 6-7), which is close to the general underestimation seen in Figure 6-6 b.

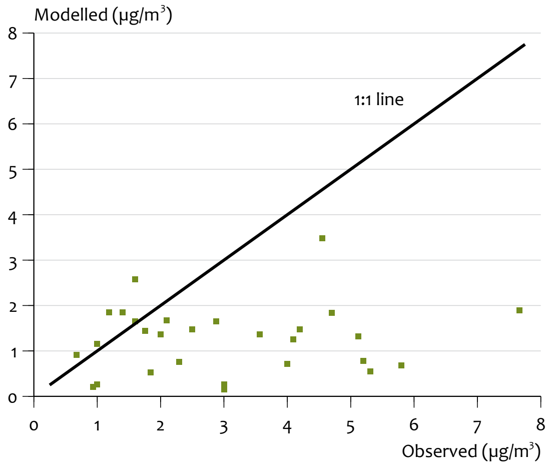
In Chapter 4, a compilation of MD in PM_{10} across Europe is presented. This compilation was meant to further validate the model predictions of mineral dust. The model was run at a 25 x 25 km resolution. Unfortunately, this implied that a first comparison could only be made for rural locations. The size of the grid cells was too large to properly capture the spikes

Annual concentrations of modelled and observed MD in PM_{10} for 7 locations across Europe **Figure 6.7**



Annual concentrations of MD in PM_{10} for 2005, in seven locations across Europe (Kollumerwaard (the Netherlands), Illmitz (Austria), Langenbrügge (Germany), Melpitz (Germany), Montseny (Spain), Birkenes (Norway), and Ikrba (Slovenia)).

Annual concentrations of modelled and observed MD in PM_{10} for 30 rural sites across Europe **Figure 6.8**



Observed mineral dust in PM_{10} versus model-predicted MD contribution to PM_{10} for 30 rural locations across Europe.

in resuspension caused by heavy road traffic, whereas the urban measurement data were generally close or next to busy streets. Therefore, to see if the model would properly predict urban background concentrations, a zoom version needed to be run and a selection needed to be made of urban locations that truly would represent the urban background situation. Traffic locations could be used to indicate the importance of resuspension, but were deemed unsuited for verification of a regional scale model, even if such a model would be run at, for example, a 5 x 5 km resolution. A first comparison for 30 rural locations across Europe is shown in Figure 6-8.

The near perfect correlation shown in Figure 6-7 cannot be observed in Figure 6-8, in fact there seemed to be no correlation, only a general level of modelled MD. This discrepancy asks for further investigation.

- First, the model tended to underpredict observed levels of MD, especially when high MD contributions to PM_{10} were observed. Modelled concentrations of MD varied between 0.2 and 3.5 µg/m³, whereas observed concentrations ranged from 0.7 to 8 µg/m³. However, the model-predicted concentrations did not include any desert dust. Some of the higher observed values in Figure 6-8 were for southern European countries, and inclusion of desert dust may increase the predicted values with ~ 1 to 2 µg/m³.
- Even more importantly, the observed values belonged to different years and were sometimes only from short measurement campaigns. In other words, quite a few of the observational data points may not have been representative of a full year. The period of sampling can be very important, as is shown in Figure 6-1 to Figure 6-4.

The match between model-predicted and observed values can be expected to improve when a more severe selection would be made of representativeness for average annual data and desert dust contributions would be included. Nevertheless,

Mineral dust in PM10 in 2007-2008, for rural Dutch locations in the BOP campaign, and according to the model **Table 6.2**

Location	MD_corrected[a)] µg/m^3	MD model predicted
Cabauw	1.45	
Hellendoorn	1.18	
Vredepeel	1.57	1.65
Wageningen		1.44
Noordwijkerhout		1.36
De Zilk		1.36
Average	1.40	1.46

[a)] see Section 2.2

the model gave a rather flat signal of between 0.5 and 2 µg/m³, and concentration levels of between 5 and 10 µg/m³ are not expected to have been covered. Such high concentration levels were most likely a regional phenomenon with further increases caused by local circumstances.

6.4 Discussion

The calculated contributions of wind-blown dust to mineral PM_{10} was (very) low, compared to those from resuspension due to road traffic or from agricultural land management. The levels could be higher in reality, which should be detectable because of the strong episodic character of these emissions. To verify whether the contribution of wind-blown dust has been underestimated, one needs to look for data on a well-documented dust storm within the European domain. However, please note that we have quantified average annual contributions, and that extreme events may only be dominant for a few days and may not necessarily contribute much to average annual concentrations.

In comparison to resuspension due to traffic or from wind-blown dust, the most uncertain factor appeared to be the modelled mineral dust induced by agricultural activities. An added uncertainty on the emission from agricultural emissions is that the mineral dust organic carbon fractions in these emissions are unknown. A more thorough study of observations would point out whether the simple approach used here is reliable (within limits) for calculating the agricultural contribution to mineral dust emissions.

In a comparison of the model-predicted concentrations for the Dutch rural locations with the measured concentrations from the BOP 2007-2008 campaign, the match appeared surprisingly good (Table 6-2). Although, as shown in the previous section, the model may have underpredicted. However, the discrepancy for the Netherlands was expected to be limited, because the most severe underprediction in Figure 6-8 occurred for southern Europe. Yet another question is that of the importance of the year that was studied. The model predicted for 2005, the BOP campaign was held in the 2007-2008 period, whereas older data (see Chapter 2) have shown that MD at Vredepeel (close to the German border) could have been substantially higher than the 1.6 µg/m³ that was reported in Table 6-2.

The modelled contributions to mineral PM_{10} from the different mineral dust sources are a (crude) approximation of reality. In general, the emission factors were based on a limited data set, which was not representative of the full range of European conditions. For this reason, the band of uncertainty on the modelled concentrations is relatively large. However, the predicted concentrations were within a realistic range, although extremes appeared not to have been well-captured. This would need further study, by identifying areas and /or periods for which the underprediction occurred most, and by attributing them to certain processes or regional conditions. We recommend that this is pursued in the follow-up of the current study, and would also include data on desert dust for southern European locations, as well as a screening on the representativeness of average annual concentrations.

7

Discussion, conclusions and recommendations

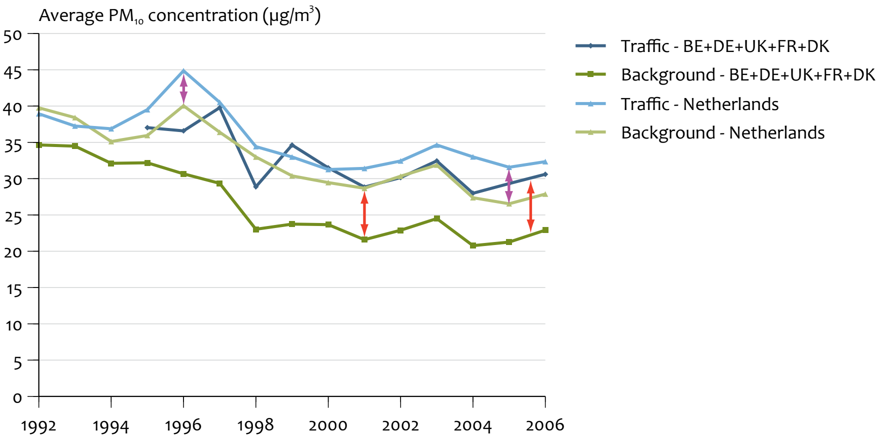
Mineral dust is generally thought to be an important component of particulate matter. Sources of this mineral dust (or soil dust) are, for example, wind-blown dust, agricultural activities, and dust resuspension caused by road traffic. Since these sources are mostly semi-natural and/or relate to re-emission, they are often not included in traditional emission inventories. Without an emission estimate, a source cannot be included in modelling of ambient PM concentrations. This is than often solved by applying a correction factor to the predicted concentrations. The consequence of this practice is that much of the 'missing' PM is attributed to 'mineral dust and similar sources', for example, in the past about 40% of ambient PM_{10} in the Netherlands was attributed to this category (MNP, 2005). However, since this concerns a substantial amount of total ambient PM_{10}, it has become an unsatisfactory solution. It inhibits the proper analysis of the origin of PM_{10} as well as the designing of effective abatement strategies, because too much of the PM is effectively classified as being from 'unknown sources'. Within the framework of the BOP Programme, this study was conducted to better understand the contribution of MD to PM in the Netherlands and in Europe.

The chemical speciation data that were collected in the BOP programme during the 2007-2008 period, at six locations in the Netherlands, have been used to estimate the contribution of mineral dust to Dutch ambient PM_{10}. Furthermore, we collected and interpreted earlier PM data (including elemental composition data) on the Netherlands and Europe, to relate the amount of mineral dust present in PM_{10} to certain geographical locations and location types (rural, urban or traffic). The amount of mineral dust was estimated using the elemental analysis of PM samples through the use of the two key tracer elements (Silicon and Aluminium). Overall, we estimated the contribution of mineral dust to PM_{10} in the Netherlands in the 2007-2008 period at no more than ~ 1.5 µg/m³. This is significantly lower than previously observed for 1998-1999 by Visser *et al.* (2001), and more recently for 2006 in Belgium (Flanders; VMM, 2009). In the neighbouring German province of North Rhine-Westphalia data were collected by Quass *et al.* (2009) in a partly overlapping period with the BOP campaign. Again, the estimates on MD based on the German data were lower than those derived from the BOP data, but the gap is smaller. This suggests that the discrepancy between the various estimates may be partly due to the period of sampling, with 2007 to 2008 having been a year with relatively low amounts of MD in PM_{10}. The measurement data confirmed that MD can be elevated during certain periods (e.g., Figure 2-5 and Figure 3-3) and annual variation is likely to occur especially in relatively wet or dry years.

However, it would be incorrect to simply say that annual variation could explain a threefold difference in average annual MD contributions, as would be needed to explain the differences between concentrations from the BOP data for 2007-2008, data on the Netherlands in the 1998-1999 period (Visser *et al.*, 2001; Denier van der Gon, 2007) and on Flanders in 2006 (VMM, 2009). Unfortunately, there are no chemical speciation data including MD tracers for PM available for background locations in the Netherlands to show the temporal variation over the past years. A further complication in a data comparison for surrounding countries is the definition of mineral dust. In some cases, for example, by the VMM (2009), it is defined to include 'road dust'. The consequence of this definition is that substantial contributions from metals, such as iron (Fe) and copper (Cu) originating from resuspended dust from mechanical wear of vehicles, are included for the urban locations. A similar complication may arise when MD calculated from tracer elements, such as Si and Al, is compared to MD contributions calculated from data using statistical source attribution techniques, such as Positive Matrix Factorisation (PMF). These techniques investigate correlations and 'groups' certain parts of the total ambient PM; here, too, soil dust and road dust (including the metals from mechanical wear) are likely to be added together. Clearly, the issue is not whether this is right or wrong, but we need to be aware of it and ensure that we make the correct comparison.

- It is strongly recommended to initiate a routine analysis of chemical speciation of PM including MD tracers, for at least one air quality measurement location in the Netherlands. Even when sampled and analysed at a low temporal resolution to limit expenses, it would greatly enhance our capacity to interpret more detailed campaign data and quantify the importance of inter-annual variation for semi-natural sources.
- A more in-depth comparison using results from surrounding countries should be initiated, including a more detailed breakdown of mineral dust and road dust, and

Average annual PM$_{10}$ concentration at background stations and street stations **Figure 7.1**



Average annual PM$_{10}$ concentrations at background and traffic locations in the Netherlands and surrounding countries (data source AirBase, 2008). Purple and red arrows show the increment at background and traffic locations for the Netherlands and its surrounding countries (Belgium, Germany, UK, France and Denmark), respectively.

of the assumptions made in the individual studies. This comparison should include at least the data from the BOP programme (this report and Schaap *et al*., 2010), and that from Belgium (VMM, 2009) and Germany (Quass *et al*., 2009)

The amount of mineral dust observed in PM$_{10}$ during the BOP campaign in the 2007-2008 period shows an increase in MD in the urban environment and at traffic locations. This is consistent with the increase in MD across the European urban environment, as could be deduced from the compilation of European data.. However, in the BOP data the increment in MD from rural background to traffic locations was ~40 to 50%. In a large number of European studies (see Chapter 5), MD in PM$_{10}$ in urban or traffic locations was found to be roughly double that of the levels at nearby rural background locations. To investigate if this discrepancy was caused by an improper sampling during the BOP campaign, we explored the rural-urban increment in the coarse fraction of PM$_{10}$ (PM$_{2.5\text{-}10}$ or PMc). The hypothesis was that although MD may have shown annual temporal patterns, this should not be the case for other sources of PMc, such as non-exhaust emissions from road traffic. Indeed, the BOP PMc data for urban/traffic locations were more than double those of the rural background which is consistent with observations elsewhere. Therefore, we concluded that the BOP data had correctly captured the increment at urban/traffic locations, with the surprising observation that this increase in PMc was not proportionally reflected in the mineral dust concentrations. So, from the BOP campaign data was concluded that MD was only a minor fraction in the observed PMc. This observation is quite remarkable, as it is contrary to the average observational data available on Europe, and contrary to what was expected for the Netherlands based on the previous source apportionment study by Visser *et al*. (2001).

The overall increment in PM from rural background to urban background concentrations in the Netherlands appeared smaller than in surrounding countries (Figure 7-1). Traffic and population may be more equally spread over large parts of the country than in other European countries, reducing the sharp gradients. Therefore, some of the more general European concepts of air quality morphology, such as the concentrations increasing from rural, to urban and traffic locations (Lenchow *et al.,* 2001), may need to be slightly modified for the Netherlands. Recently, Keuken *et al.* (2009) compared observational data from traffic locations in Amsterdam and Rotterdam, as part of a study to investigate the impact on PM of street sweeping and washing. Although this impact was limited, their results did confirm that PMc levels are significantly higher at traffic locations. The PMc found at these locations deserves further attention, as it is relevant to human health, being a mix of metals and heavy metals, organic carbon – including PAHs, and salts and mineral dust. Local policies could be effective here, especially because it mostly originates from nearby sources.

- It is recommended to focus further research on the increment in the total coarse fraction of PM$_{10}$ at traffic and urban locations by using chemical speciation to identify the various source contributions, and to subsequently propose the most effective measures. This research should include temporal variation in the composition of PMc throughout the year.

For this study, we compiled European data to estimate MD in PM$_{10}$ for 19 European countries, often from more than one location per country. We preferred to estimate MD levels by using available chemical analysis data, but we also included data from studies that reported on MD without exact chemical speciation. The compilation shows that MD in PM$_{10}$ in urban centres ranged between 2 and 18 $\mu g/m^3$, with the higher value being an extreme and probably an overestimation. The more likely range would be between 3 and 13 $\mu g/m^3$, confirming the importance of MD in PM$_{10}$. Mineral dust in PM$_{10}$ in the Netherlands appeared to be relatively low, compared to southern and northern Europe.

These regions appeared to be experiencing higher MD concentration levels due to a dryer climate (southern Europe) or the use of studded tires and road gritting in wintertime (northern Europe). In the context of this study, the Netherlands would rank with countries such as Belgium, Germany, and the United Kingdom.

- A more detailed comparison between observational MD data from surrounding central European countries is recommended, possibly with national laboratories measuring the same filters to solve any discrepancies.

Specific modules have been developed for estimating MD emissions from wind-blown dust, agricultural land management and resuspension, and have been implemented into the LOTOS-EUROS model. Explicit modelling of MD as a PM component is a novelty in Europe. The first results for the model-predicted MD show that emissions from agricultural land management and from resuspension due to road traffic, contributed most to the total modelled mineral dust concentration; wind-blown dust and wind erosion appeared to be relatively minor sources. The compiled European observational data on MD allowed a comparison with the model-predicted MD and observed concentrations. This comparison was rather crude, since the observational data did not necessarily come from the same year or period that was covered by the model runs. A first verification of the modelled seasonal variation in mineral PM_{10} concentrations described by Schaap *et al.* (2009) indicated that the model had overestimated the impact from agricultural activities by using very high emission factors for ploughing. These emission factors have since been adjusted, which resulted in a better match between modelled and observed MD in PM_{10}. Average annual concentrations for a limited number of European rural locations were predicted remarkably accurately. However, a comparison between modelled concentrations and a larger set of observations indicated that current model approaches underestimate total mineral dust contributions. This underestimation may well be related to incomplete coverage of the urban, anthropogenic activities that add to MD in ambient air, such as construction and demolition, material handling and other urban activities that may cause resuspension of dust.

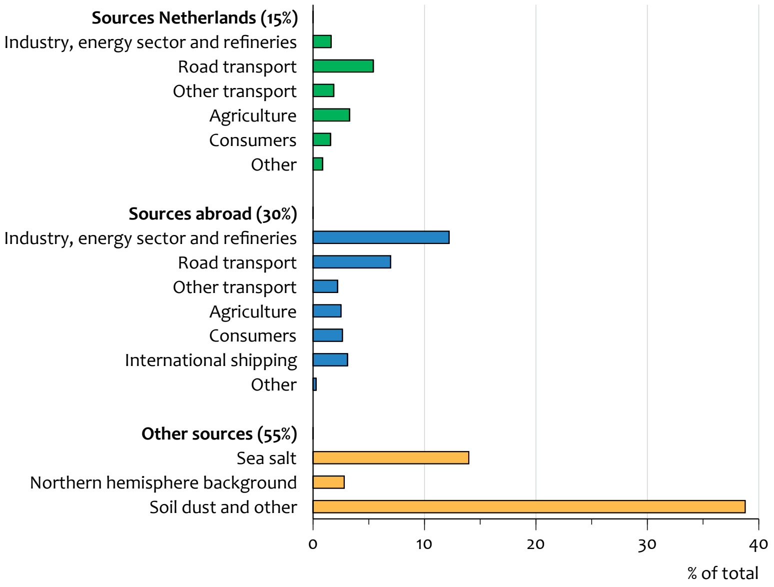
- The importance of urban, inner-city processes other than resuspension due to road traffic, should be quantified. The most remarkable feature of MD in PM_{10} still remains its increase within the urban environment.
- Further progress could be made by using all of the data collected in Europe (Chapter 4) and comparing them in more detail with the model-predicted concentrations (Chapter 5). In more detail, here, means investigating regional patterns and differences between rural and urban locations, as well as temporal variations. The last is important for checking certain hypotheses, but is also complicated because the observational data are collected over many different years.

Within the BOP project, we have been able to create a database of observational data and make modules to model MD in PM_{10} with the LOTOS-EUROS model. A tool has been created to investigate the different source contributions and is expected to enable further understanding by introducing the specific time of occurrence and spatial distribution for each source category. The match between model-predicted MD and some observational data on mineral dust at background locations across Europe, is excellent. Furthermore, the model-predicted MD concentrations in PM_{10} for Dutch rural locations matched with data from the BOP campaign (which had relatively low shares of MD.. However, it is suspected that the model results for certain conditions and regions were still underestimated, especially for urban background locations. Whether this was truly a regional or local effect, or partly caused by the lack of some relevant sources, is unknown. The temporal and spatial patterns of observations and model predictions could provide important clues as to whether the balance between sources and the time of occurrence of their emissions were correct in our approach. This would require rather time-consuming data mining, but seems the best way forward. Within the uncertainties surrounding such an exploratory approach, the balance between source contributions could be optimised to fit the observations in terms of absolute annual concentrations, as well as in temporal patterns over the year. The result could constrain the total source strength of mineral dust and an indicative ranking of the importance of contributing sources in various European regions. In short, the results achieved bring us much closer to understanding the role of MD in ambient PM10 but further analysis and improvement remains wanted.

- It is recommended that an empirical iteration is performed in a follow-up study partly based on process knowledge, to match the spatial and temporal patterns predicted by the model with the compiled observational data.

The final policy relevant conclusion of the present report would be that MD in PM_{10} in the Netherlands is less important than was previously thought. Before the start of the BOP programme, the general perception in the Netherlands was that mineral dust was one of the major contributors to ambient PM_{10}, as was shown in the overview publication by MNP (2005) and is summarised in Figure 7-2. This showed that, at the time, close to 40% of ambient PM_{10} was attributed to ‘mineral dust and other sources’ (Figure 7-2), with other sources not being sea salt or originating from the hemispheric background. Although other sources may still be important, we have concluded that mineral dust is not the dominant contributor it was previously thought to be. This conclusion is robust and would not change, even if the current BOP data would have underestimated the mineral dust contribution by a factor of 2, due to an unfavourable year or analytical recovery. This implies that the gap between the observed and the modelled concentration levels could not be closed by including emissions of mineral dust. This is consistent with another important discovery in the BOP programme, namely that secondary inorganic aerosol in the Netherlands is substantially higher than previously thought (Schaap *etal.*, 2010). Indeed, unravelling the ‘non-modelled’ part of PM, one of the aims of the BOP programme, has brought important new insights into the origin, composition and importance of (semi-natural) PM in the Netherlands.

Composition of ambient PM_{10} in The Netherlands before the start of the BOP programme **Figure 7.2**



The origin and source attribution of ambient PM_{10} in the Netherlands as understood in 2005 and 2006, before the start of the BOP programme (source MNP, 2005).

Acknowledgements

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Annex A Sources of mineral dust contributing to PM_{10}

Mineral dust may originate from distinctly different sources. Despite the importance of mineral dust in total PM_{10} mass, the sources are still poorly understood, as is the relative importance of individual mineral dust sources. This annex presents an overview of the most important sources of MD that may contribute to PM_{10}.

A.1 Wind-blown dust

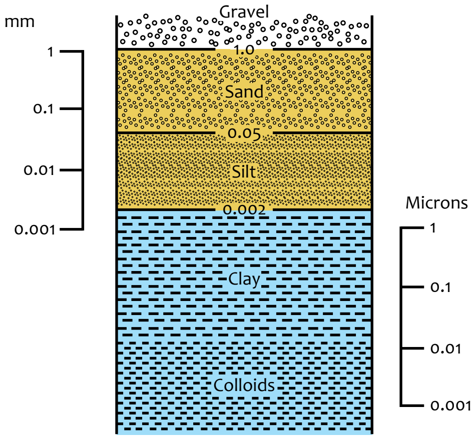
Wind action can mobilise dry, loose mineral dust on exposed ground surfaces and result in their suspension into the air. Mobilisation and the potential for suspension are dependent on particle size (Figure A-1). In order to become mobile, a particle would have to overcome the gravitational force, inter-particle cohesion and wind-shear stress. Particles of more than 0.5 mm (500 µm) are large and suppressed in their vertical motion because of gravitational force. These large particles roll or slide along the ground surface in a process called surface creep or surface traction, and do not become airborne. Medium-sized particles (D=50-500 µm) can be lifted from the surface and may reach a maximum altitude of one metre before falling back to the surface. This 'hopping' movement, known as saltation, occurs only if the wind friction velocity is above a certain threshold, such that the gravitational force is partly overcome. As the sallying particles impact the ground they can dislodge smaller particles (D<20 µm), either from the soil or from the falling particle itself, thus releasing them into the air. The impact may also induce surface creep and initiate abrasion. This impact action is the main process responsible for wind generated dust production and is also called sandblasting. Arid regions are the greatest source of suspended mineral dust, with the Sahara desert being the largest source of mineral dust on earth. Various parameterisations have been developed to predict wind-blown dust from deserts (e.g. Tegen et al., 1997), because at the global scale wind erosion or wind-blown dust is a major aerosol source. During dust storms, mineral dust spreads from the Sahara desert, across the Mediterranean and Caribbean seas, into Europe and the Americas, while dust from the Gobi Desert (southern Mongolia, northern China) affects eastern Asia and western North America. Chapter 6 presents a more detailed description of the source function that is used in the BOP simulations in this study.

A.2 Agricultural Activities

Mineral dust may be emitted because two major processes related to crop cultivation; 1) from wind erosion of bare fields, and 2) from land management and crop harvesting. The emission process is quite similar to that of wind-blown dust. The contribution from the second process – land management and harvesting – has been recognised for a long time. For example, already ~ 15 years ago the US Environment Protection Agency (EPA) presented emission factors to estimate contributions from agricultural tilling to PM inventories using emission factors based on surface soil silt content (U.S. EPA, 1995). However, it is widely recognised that a single parameter, such as soil silt content, cannot explain the observed emissions, as a myriad of factors seems to affect actual emissions during land and crop management (e.g., climate, soil wetness, type of machinery, crop type). To estimate the MD part of PM emissions from land and crop management is even more complicated as part of the emitted material tends to be organic (from crops, crop residues or soil organic matter). In the central region of the United States especially, PM emissions from agricultural fields are considered a major source, with total PM10 emissions from agricultural tilling operations estimated at >1.3 Mt yr-1, and PM2.5 emissions contributing ~270,000 tonnes to this total (Penfold et al., 2005). However, the situation in the Midwestern United States is not directly comparable to the European situation. However, Hinz et al. (2007) identified agricultural lands as a considerable source of PM emissions in Europe. Unfortunately, knowledge on PM10 and PM2.5 emission factors across the diverse range of agricultural activities is insufficient. This is mainly due to a poor availability of reliable observations. Currently, a handful of studies are available that have estimated the emission potentials for several land management activities. The estimates of PM emission factors for land preparation and harvesting vary widely, ranging from 0.3 to 14 kg ha-1 and 3.8 to 45.7 kg ha-1, respectively. The uncertainty associated with these numbers is also large. Most of the estimates have been based on PM measurements taken close to their points of origin, and emission factor estimates were inferred from PM observations by means of dispersion modelling. It has been said that these estimates perhaps do not reflect particle emissions on a larger scale (Hinz et al., 2007). Nonetheless, it was the only information available to work with, and the numbers presented in Table A-1 form the basis of the approach described

Sediment size grades used in the description of soil texture

Figure A.1



Grade Name	Diameter Limits
	2.0 mm
Very coarse sand	1.0 mm
Coarse sand	0.5 mm
Medium sand	0.25 mm
Fine sand	0.10 mm
Very fine sand	0.05 mm
Silt	0.002 mm (2 microns)
Clay noncolloidal	2 to 0.01 microns
Colloidal	<0.01 microns

Sediment size grades used in the description of soil texture (source: Strahler and Strahler, 1992).

Total PM_{10} and $PM_{2.5}$ emission factors for land preparation and harvesting activities

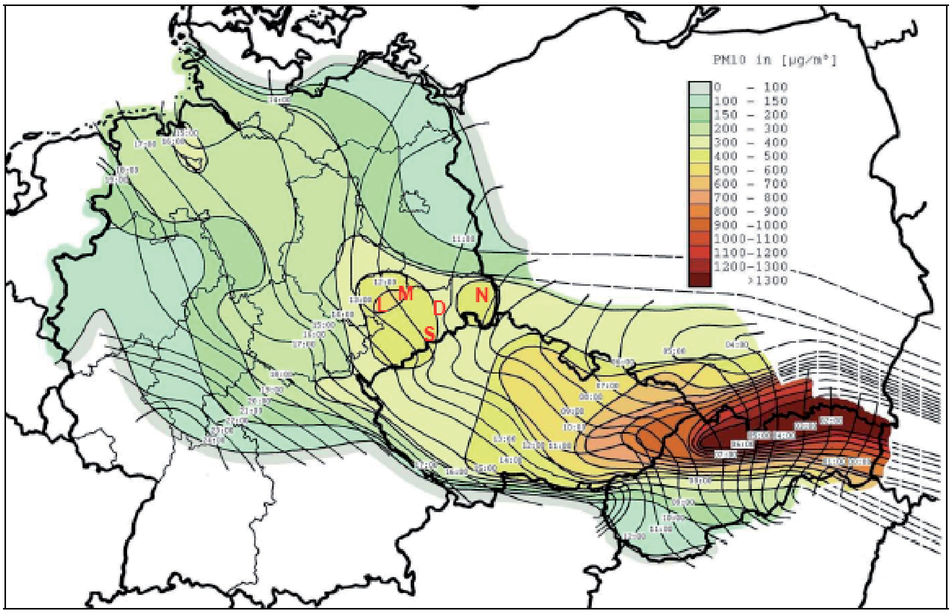
Table A.1

Reference	Location	Year	Method	Activity	Operation type	Emission factor $PM_{2.5}$ (kg ha^{-1})	PM_{10} (kg ha^{-1})
Öttl et al. (2007)	Germany	2004	PM obs., disp. mod.	land preparation	harrowing,	0.29	0.82
					discing,	0.12	1.37
					cultivating,	0.06	1.86
					ploughing (dry)	1.3	10.5
Funk et al. (2007)	Germany	2006	PM obs., disp. mod.	land preparation	ploughing (wet)	0.05	1.2
Gaffney et al. (2003)	, United States	1995-2003	Upwind-downwind	land preparation	root cutting,		0.3
					discing, tilling, chiselling,		1.3
					ripping, subsoiling,		5.2
					land planing and floating,		14.0
					weeding		0.9
				harvesting	almonds,		45.7
					corn,		1.7
					cotton,		3.8
					wheat		6.5

in Chapter 6, in which is described whether these numbers could result in a realistic representation of MD in PM10. Note that the data in Table A-1 represent the emission factors and not the share of MD in PM10 or PM2.5.

It has been established that agricultural dust has an impact on both local and regional scales. Even single events can have an impact on concentration levels in distant locations, especially under certain meteorological conditions. Birmili et al. (2008)

Spatio-temporal evolution of an agricultural dust plume in Central Europe on March 24, 2007 **Figure A.2**



Spatio-temporal evolution of an agricultural dust plume in central Europe on 24 March 2007.The dust plume presented in Figure 45 originated in the Ukraine and spread as far as to Germany. The map was based on time series of PM10 mass concentrations from over 360 government monitoring locations in Slovakia, the Czech Republic, Austria, Poland and Germany. Labelled frontal lines indicate peak values of PM10 and where these peaks occurred, simultaneously. The location of some monitoring sites is given: L=Leipzig, M=Melpitz, N=Niesky, S=Schwartenberg, D=Dresden. Dashed isolines are extrapolations along back trajectories over areas of missing data (Source: Birmili et al., 2008).

examined a dust cloud that formed over parched agricultural fields in southern Ukraine, which led to extremely high concentrations of particulate matter in central Europe. On 24 March 2007, the dust cloud spread across Slovakia, Poland and the Czech Republic to Germany. Peak concentrations of between 200 and 1400 µg m^{-3} of PM_{10} were measured (Figure A-2). By way of comparison, the average daily limit in the EU is 50 µg m^{-3}. Even if the prevailing meteorological conditions occur relatively infrequently, the unexpected scale of this phenomenon highlighted the potential strength of this source of mineral dust in the European region, especially within the context of increasing human-induced desertification and climate change (and the associated future changes in air circulation patterns).

Various methodologies exist to minimise the (PM) emissions from arable land. Often they are part of a strategy to minimise soil fertility loss. Examples are the planning of land preparation activities; these can be planned in such a way that emissions are minimised (e.g. when the soil moisture content is high, or by watering the soil before commencing activities), the type of tillage can be adjusted (Madden et al., 2008), land management can be adjusted, or an intermittent crop can be planted to keep the soil covered between crops.

A.3 Construction and Demolition

A growing population, growing urbanisation and the pressure on government to provide adequate housing and increase the capacity of transportation networks, means constant changes in the urban planning system, which in turn makes construction and demolition a growing and ever-present reality. Dust emissions from demolition and construction works can impact the air quality at construction sites (posing an occupational hazard to on-site workers), as well as on the indoor air quality in surrounding areas (posing public health impacts). Dust from demolition and construction works can also leave deposits on cars, windows and other property, reducing the quality of life. However, through careful planning and good management, these impacts can be reduced. In the United Kingdom, most local authorities have implemented a protocol to control dust emissions from construction and demolition works (e.g. London Councils and GLA, 2006). The protocols are used in the construction and demolition planning process, informing developers[1] about legislation requirements and the methods available to them for minimising dust emissions. For London, the protocol is reviewed regularly in order to update new best practice in dust emissions management.

Construction and demolition activities which generate dust are those of excavation, stock piling, cutting, grinding,

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[1] including architects, environmental consultants, local authority officers and any parties involved in the construction process.

Materials handling is a potential source of mineral dust PM_{10} **Figure A.3**

Material handling (loading, unloading, stockpiling, blending, transporting) is a potential source of mineral PM_{10}. (Foto: Wilco de Vries)

sawing, concrete crushing, cement batching, and vehicle movements in unpaved areas.

Dust emissions can be reduced by the following measures:

- Using water as a suppressant during dust generating activities (e.g. demolition, cutting, crushing and managing of stockpiles);
- Wrapping buildings to be demolished, covering stockpiles and ensuring that trucks carrying debris are properly covered before they leave the site;
- Dampening haul routes within and outside construction and demolition sites, especially during dry weather;
- Regularly vacuuming up dusty residue (rather than sweeping) to prevent the build up of dust;
- Erecting solid barriers around the site;
- No burning of any material on site.

A.4 Handling of Mineral Goods

Mineral dust may be generated and released into the atmosphere during industrial processes which use, produce, handle, store and transport mineral products. Cement production, lime production, limestone and dolomite use, soda ash production and use, asphalt roofing, road paving with asphalt, and mining (NFR sectors 2A and 1B1a) are major activities that may lead to mineral dust emission. With respect to mining operations, the activities of handling, conveying, transportation, storage, processing, crushing, classifying, milling, drying, and filtering, are a source of fugitive dust emissions (Figure A-3). The handling of goods is widely recognised as an emission cause. These emissions are reported in emission inventories, although often the chemical composition and dependence on type of goods handled are unknown. So, emissions reported under handling of goods may include mineral dust, but often other products such as coal, grain, and fertilisers are involved.

A.5 Road wear

Road surfaces consist mainly of stony materials. This is not only true for concrete or paved roads, but also for asphalted surfaces. Asphalt concrete is a high-quality, thoroughly controlled, engineered material which consists of two basic ingredients. The main ingredient is aggregates (stone, gravel and sand), which constitutes ~95% of the total weight of the paving mixture. The remaining 5% is bitumen, a derivative of crude oil refining that serves as a binder or ‘glue’ for the aggregates. The contribution from road wear to the total PM_{10} emission factor is difficult to separate from the contribution of wind-blown or resuspended soil and other geological material. However, in a road tunnel, the amount of wind-blown soil dust coming from the shoulders or from elsewhere is limited and a first approximation could be that all the mineral dust in PM_{10} produced inside the tunnel originates from road wear. Gillies *et al.* (2001) reported that ~12% of the PM_{10} in the Sepulveda Tunnel (Los Angeles, the United States) was of geological origin. Denier van der Gon *et al.* (2003) used these results in combination with a chemical analysis of PM_{10} formed in the Maas Tunnel (Rotterdam, the Netherlands) to derive an emission factor of 3 to 4 mg per vehicle kilometre (vkm) for mineral dust from road wear. Road wear depends not only on the road surface material but also on vehicle braking frequencies and cornering, which cause additional friction. In a tunnel environment these situations are limited compared to those in average urban traffic. Hence, we awarded a lower value to the tunnel road wear emission factor. In the literature, road wear emission factors often range between 8 and 10 mg/vkm, which is consistent with a 3 to 4 mg/vkm lower limit.

A.6 Road gritting and the use of studded tyres

Additional mineral dust, contributing to the particulate mass, may result from the use of anti-skid methods to enhance traction on snowy or icy road surfaces. In Europe, this applies especially to Nordic and Alpine countries. Such methods include gritting road surfaces and equipping tyres with metal studs or a special tread design. The use of studded tires strongly enhances road wear. In the early 1990s, it was recognised that the use of studded tyres was one of the major sources of PM_{10} in Scandinavian countries (Larssen, 1994). An alternative for studded tyres is the use of traction sand on road surfaces, which is crushed under vehicles' tyres into smaller particles, and the aggregate in asphalt concrete is worn by interaction with the tyres. Because the traction sand and the mineral matter from this aggregate can be of similar mineralogical composition, it has been difficult to determine the source of the mineral fraction in PM_{10}. Kupiainen *et al*. (2003) showed that ambient PM_{10} concentrations were higher when traction sand was used, regardless of whether tyres were studded or not. Surprisingly, the use of traction sand greatly increased the number of particles originating from the road surface. It was concluded that sand must contribute to Road wear. This phenomenon is called the sandpaper effect (Kupiainen *et al.,* 2003).

Emission factors for studded tyre use and road gritting

Emissions from studded tyre use and road gritting cannot be separated, they may occur at the same time in the same countries. The increase in springtime emissions because of these anti-skid measures taken in the winter in Nordic and Alpine countries, is well documented. Additional high springtime PM_{10} peaks with a contribution of non-exhaust mineral material of up to 90% have been reported, for instance by Omstedt *etal*. (2005) and Kupiainen *et al.* (2003). In some US studies, ~60% of the observed ambient PM_{10} could be attributed to road gritting (Gertler *et al.,* 2006; Wittorff *et al.,* 1996). Furthermore, about a 30% increase in PM_{10} and $PM_{2.5}$ was observed after roads that had been covered in a brine solution, had dried up (Gertler *et al.,* 2006). Derived emission factors for before and after application of road abrasives, increased from 229 to between 310 and 660 mg vkm^{-1} and from 76 to 133 mg vkm^{-1} for PM_{10} and $PM_{2.5}$, respectively. The increase in PM_{10} emission after street sweeping following a snow storm was not large, but for $PM_{2.5}$ a significant impact was observed. Kuhns *et al.* (2003) reported a short-term increase in PM_{10} of up to ~75%, due to the combined effect of winter gritting and street sweeping.

In recent years, much more detailed studies, showing the contribution from road gritting and studded tyre use, have become available (e..g. Gustafsson *et al.,* 2008; Hussein *et al.*, 2008). However, application in a regional scale model is still complicated because time of occurrence of emissions is important. Particles are deposited in snow, and after the snow melts and road surfaces dry up, a proportion of the dust is resuspended by road traffic. Therefore, high concentrations occur especially during the spring, but the emissions peak at varying times, from year to year and from country to country. Omstedt *etal*. (2005) developed an empirical model for investigating the influence of road gritting and studded tyre use on the resuspension of PM emission along Swedish roads. The model uses emission factors and traffic data, and takes into account the thickness of the on-road dust layer incorporating resuspension and run-off by precipitation or otherwise. This model is specifically and successfully applied to investigate the use of road gritting and studded tyre use in Scandinavian winters in relation to fugitive PM_{10} concentrations. Based on the findings by Omstedt *et al*. (2005), we applied a multiplication factor of 2 to the standard emission factors for heavy-duty vehicles (HDVs) and light-duty vehicles (LDVs) and for PM_{10} and $PM_{2.5}$ in the spring months, to incorporate additional resuspension which results from road gritting and studded tyre use in Scandinavian countries. Currently, road gritting in the Alpine region is not accounted for in the modelling study (see Chapter 4 for more details).

A.7 Driving over unpaved roads

Most unpaved roads consist of a compacted roadbed, usually created from the parent soil material. Both wind action and turbulent vehicle wake effects liberate fine particulate matter from the road surface leading to their resuspension in air. Dust emission rates depend on the fine particulate matter content of the road surface, soil moisture content, vehicle speed and vehicle weight. Resuspension emission factors (grams of PM_{10} liberated per travelled vehicle kilometre) on unpaved road surfaces, as well as the size of the turbulence created by a vehicle are directly and proportionally related to vehicle speed, weight and size (Gillies *et al*., 2005). Apart from contributing to PM concentrations, dust from unpaved roads also has other effects. To residents living along unpaved roads, the traffic-generated dust penetrates their homes, causing respiratory problems and other general nuisances. Crops and vegetation near unpaved roads can be covered in the airborne dust, stunting their growth due to the shading effect and clogging of the plants' pores. For motorists, the traffic-generated dust can reduce visibility and be a driving hazard. There are various mitigation options for limiting dust emissions. For roadbeds with soft and loose soil, the soil can be mixed with chemicals to stabilise soil properties and suppress dust formation. For example, the natural cementing sugars in lignin increase cohesion between soil particles, making it an effective soil stabiliser. Other dust suppression methods range from limiting vehicle speeds to applying water on the unpaved road surface.

PM_{10} emissions from traffic on unpaved roads are considered a significant source of PM within the United States (Gillies *et al*., 2005; Claiborn *et al.,* 1995). However, in most of Europe this road traffic is very limited and conditions seem less favourable for road dust to be resuspended (overall, a wetter climate). For these reasons, mineral dust emissions from unpaved roads have not been taken into account in the current BOP simulations. However, for some countries, especially the more dryer southern European countries, the importance of this source may be underestimated.

A.8 Resuspension due to road traffic

Moving vehicles cause atmospheric turbulence on a local scale, which in turn evokes the resuspension of particulate matter or dust from the road surface into the air. Traffic-generated fugitive dust is a predominant source of atmospheric particulate pollution at traffic locations (Thorpe *et al*., 2007) and makes up a large part of the PM_{10} emissions from road traffic. Some studies (Lenschow *et al.,* 2001; Harrison *et al.,* 2001; Gehrig *et al.,* 2004) suggest that resuspension of road dust is of the same order of magnitude as primary emissions from road traffic (e.g. vehicle exhaust, tyre abrasion, brake wear). The amount of resuspended PM per vehicle kilometre travelled is not the same at every location. This depends on the amount of dust that is susceptible to mobilisation (very fine fraction), humidity conditions (occurrence of precipitation), and surface wetness. Furthermore, in the turbulence caused by vehicles, downward air movement can also lead to re-deposition of particles as they impact the surface. Kuhns *et al.* (2003) suggested that the on-road PM_{10} matter exists in an equilibrium between emission and deposition, with short-term deviation periods lasting up to several hours. The amount of dust that is available for resuspension is limited because of road cleaning (within well-maintained city centres) or by run-off (Kuhns *et al.,* 2003). However, these sinks are currently not well quantified.

The resuspension emission factor is a function of traffic properties, such as the aerodynamic vehicle properties, vehicle type, vehicle speed, traffic volume, climate, and regional conditions. From an emission point of view, resuspension is a difficult phenomenon, as it is mostly re-emission and there is uncertainty about the correct activity data. That is, kilometres driven would seem to be the obvious choice, but after a certain amount of vehicle have passed by within say an hour, resuspension will slow down, as all the PM available will already be suspended and more vehicles will no longer increase the amount of resuspended material. Furthermore, although a substantial amount of the resuspended dust may be mineral dust, it is also composed of, for instance, material resulting from vehicle wear (e.g brakes and tyres) and deposited PM from other sources (combustion related emissions). Within the framework of the BOP programme, Schaap *et al.* (2009) have presented an extensive review of the data on resuspension. This is summarised in Chapter 5 of this report.

Annex B On the use of calcium to estimate mineral dust in PM_{10}

As an alternative to silicon and aluminium, calcium (Ca) is often used as a tracer for mineral dust. The main reason being that Ca is more often analysed in samples, is abundant in soils, and in PM samples often correlates well with Si and Al. However, the relative presence of Ca in soils, compared to other elements, is much less than in PM_{10}. In soil, calcium is fixed to the soil's cation exchange complex, clay and organic matter. Calcium is the most important exchangeable base in terms of physical, chemical and biological reactions in soil. Calcium is commonly present in soil, but concentration levels may vary. Soil science and agriculture often use five calcium content classes (Table B-1). For the Netherlands, the average content of Ca is 2 to 3% (Spijker *et al.,* 2008). In top soil, which is the most likely source of mineral dust entrainment into the atmosphere, there is no relation between the aluminium (or silicon) content and calcium (Figure 2-1; Chapter 2).

In the Netherlands, calcium has been measured for a long time, because it is part of the routine sampling and analysis by the Dutch National Air Quality Monitoring Network (LML) (see www.lml.rivm.nl). So, if calcium would be a reliable tracer, the added benefit would be the availability of a long time series for mineral dust. However, this has proven difficult. Calcium concentration levels that were measured during the same period by the LML network and in the BOP campaign, are shown in Figure B-1. Without going into much detail, it can be concluded that correlations do not look promising, and understanding the reasons for this difference in behaviour would require additional research. What can be learned form the LML series is that calcium is strongly influenced by anthropogenic activity, as its concentration pattern depends on the day of the week (Figure B-2; 433 = traffic location of Vlaardingen). Furthermore, no clear relationship between the LML calcium data and wind direction or wind speed could be found (data not shown); again an indication that calcium is more influenced by anthropogenic activities than by natural phenomena, such as storms or dry episodes.

In the BOP campaign, calcium concentrations correlated with the mineral dust markers Al and Si (Figure B-3). However, especially in episodes with higher mineral dust concentrations (Al and Si concentrations increase), calcium started to behave different and correlated less with Al and Si (Figure B-3).

We concluded that Ca often would correlate with the mineral dust content, but did not have a fixed correlation with Al and Si in soils. Hence, estimating soil dust in PM_{10} by using Ca data can be done, only when using empirically derived relationships between total soil dust and Ca concentrations in PM_{10}, and not by using the average Ca content in soils. The latter would imply multiplication of the Ca concentration in PM_{10} by 20 to 50 (based on a 2 to 3% soil content), resulting in unrealistic or even impossible shares of soil dust in total PM. The empirical factors are closer to a multiplication factor of 7, implying that Ca is significantly enriched from sources other than soil material, for example, from calcareous materials and concretes. Therefore, our tracers of choice for mineral dust were Si and Al and further checks on the consistency were done, using for instance Ti and Ca.

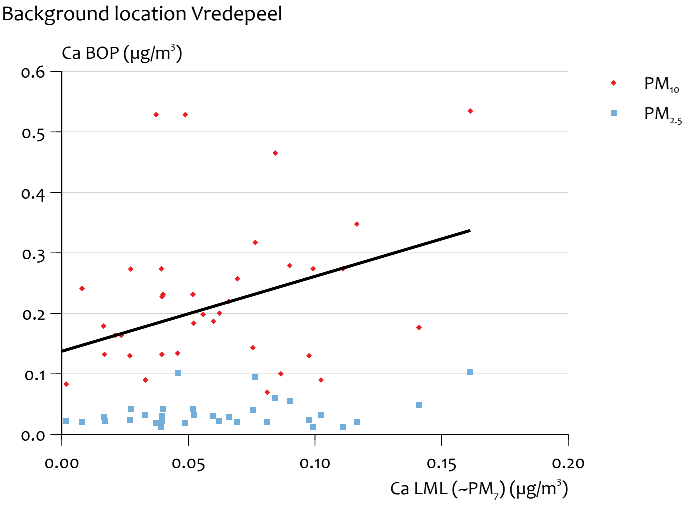
Definition of Ca content classes for soils **Table B.1**

Class[a]	Ca [a] (meq/100g)	Ca (mg/kg)	%Ca
Very low	> 2.5	< 5	< 0.5
Low	2.5-8.5	5	1.7
Moderate	8.5 – 14.5	17	2.9
High	14.5 – 20.5	29	4.1
Very high	< 20.5	41	> 4.1

[a] Martin and Nolin (1991)

Correlation between the calcium concentrations measured in LML and BOP

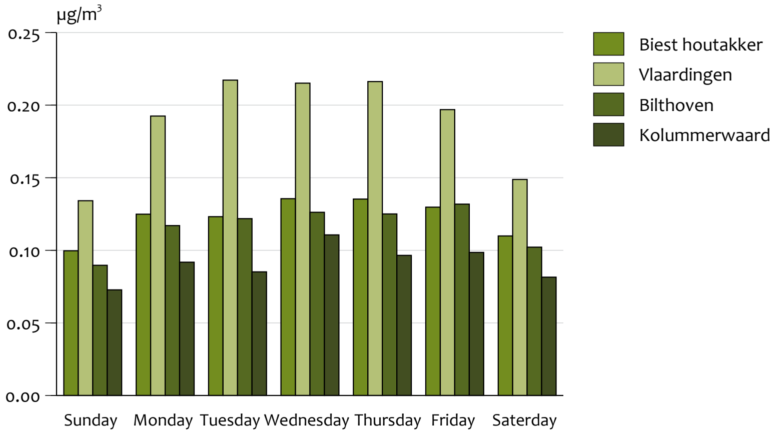
Figure B.1



Correlation between calcium concentrations measured in the LML network and the BOP campaign, at the background location of Vredepeel.

Calcium concentration at a number of Dutch measurement stations

Figure B.2

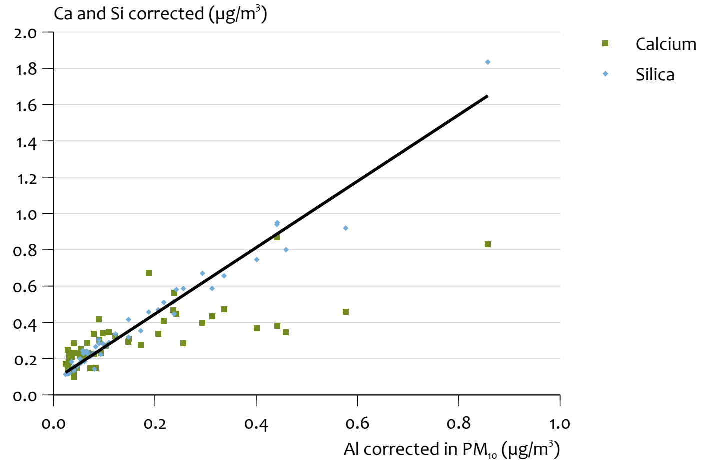


Calcium concentrations at a number of Dutch measuring locations, as a function of the day of the week.

Calcium and silica in PM_{10} as a function of the aluminium concentration

Figure B.3

Background location Vredepeel



Calcium and Silica concentrations in PM_{10}, as a function of the aluminium concentration at the background location of Vredepeel, during the BOP campaign in 2007-2008.

Annex C Collected observational data for mineral dust across Europe (annex to Chapter 5)

This annex provides the detailed data compilation described in Chapter 5, used for deriving generic rules for the contribution of mineral dust to PM_{10} across Europe. The data compiled in Table C-1 can be used for validating model results or – in the case of empirical tuning of source functions – for calibrating specific source functions for specific regions or location types.

The results described in Chapter 5 were based on a wide literature survey and sometimes personal communication with researchers active in this field. Not all literature that was reviewed was included in the Chapter 5 text, but a separate list of all the literature reviewed is provided in Annex C.1.

Collected data on PM10, MD_PM10, % MD in PM10, PM2.5, MD_PM2.5, % MD in PM2.5 at individual locations across Europe

Table C.1

ID	Ref.	Country	Name/Location	Location_type	year	PM_{10} [$\mu g/m^3$]	MD_{10} [$\mu g/m^3$]	MD in PM_{10} [%]	$PM_{2.5}$ [$\mu g/m^3$]	$MD_{2.5}$ [$\mu g/m^3$]	MD in $PM_{2.5}$ [%]	$MD_{2.5}/MD_{10}$ [%]
1	1	FIN	Helsinki	urban	2003-2003	21.10	6.62	31.39	8.30	0.53	6.40	8.01
2	2	SWE		urban		17.00	7.00	41.18	8.00	2.00	25.00	28.57
3	2	GBR		urban		25.00	5.00	20.00	16.00	2.00	12.50	40.00
4	3	GBR	Bloomsbury/London	urban	2004-2005	---	---	---	31.00	0.60	1.94	---
5	4	IRL	Coleraine Street/ Dublin	urban	2001-2002	22.10	4.70	21.27	11.70	0.50	4.27	10.64
6	4	IRL	Old Station Road/Cork	urban	2001-2002	23.90	3.50	14.64	12.50	0.50	4.00	14.29
7	5	NLD	Overschie / Rotterdam	urban	1998-1999	28.10	5.76	20.50	19.70	0.68	3.45	11.81
8	5	NLD	Overtoom / Amsterdam	urban	1998-1999	24.50	3.73	15.22	16.50	0.81	4.91	21.72
9	2	NLD		urban		25.00	3.00	12.00	20.00	0.60	3.00	20.00
10	1	NLD	Amsterdam	urban	2003-2003	33.80	2.52	7.45	25.40	0.66	2.60	26.19
11	6	BEL	Ghent	urban	1993-1994	39.80	3.67	9.23	26.39	1.05	3.98	28.61
12	7	BEL	Ghent	urban	2004-2005	24.25	2.20	9.07	18.50	0.60	3.24	27.27
13	6	CHE	Zurich	urban	1998-1999	24.50	2.85	11.63	18.98	1.18	6.23	41.40
14	6	CHE	Basel	urban	1998-1999	25.77	3.15	12.20	17.85	1.40	7.83	44.44
15	8	CHE	Basel	urban	1998-1999	24.80	2.90	11.69	18.90	1.20	6.35	41.38
16	2	CHE		urban		24.00	2.00	8.33	19.00	1.00	5.26	50.00
17	2	DEU		urban		28.00	5.00	17.86	22.00	3.00	13.64	60.00
18	1	DEU	Duisburg	urban	2002-2002	21.90	2.83	12.93	14.70	0.82	5.60	28.98
19	1	CZE	Prague	urban	2002-2003	35.00	2.85	8.13	29.60	0.74	2.50	25.96
20	1	GRC	Athens	urban	2003-2003	54.00	19.95	36.94	25.30	1.29	5.10	6.47
21	3	ITA	Via-Messina/Milan	urban	2003-2004	---	---	---	47.00	1.70	3.62	---
22	9	ITA	Milan	urban	1997-1998	92.00	17.94	19.50	60.50	3.33	5.50	18.56
23	10	ITA	C.so Firenze/Genoa	urban	2003-2004	25.00	4.50	18.00	---	---	---	---
24	10	ITA	Brignole/Genoa	urban	2002-2004	41.30	4.60	11.14	---	---	---	---
25	6	ITA	Bologna	urban	2000-2000	44.23	4.92	11.12	35.82	2.89	8.06	58.74
26	11	ITA	Milan	urban	2002-2002	---	---	---	42.90	1.55	3.61	
27	12	ITA	Villa Ada Park / Lazio region	urban	2004-2005	36.50	8.10	22.19	---	---	---	---
28	12	ITA	Latina/Lazio region	urban	2004-2005	39.10	9.90	25.32	24.70	3.00	12.15	30.30
29	12	ITA	Viterbo/Lazio region	urban	2004-2005	36.50	13.40	36.71	25.60	2.30	8.98	17.16
30	3	ESP	Jaume Almera-CSIC / Barcelona	urban	2003-2004	---	---	---	34.30	3.70	10.79	---
31	1	ESP	Barcelona	urban	2003-2003	46.30	10.35	22.35	20.00	0.88	4.40	8.50
32	2	ESP		urban		31.00	10.00	32.26	19.00	3.00	15.79	30.00
33	7	ESP	Barcelona	urban	2004-2004	31.75	8.80	27.72	21.10	1.75	8.29	19.89
34	13	ESP	Burgos	urban	2004-2004	30.00	6.00	20.00	22.00	2.80	12.73	46.67
35	13	ESP	Mallorca	urban	2004-2004	28.00	7.90	28.21	20.00	2.90	14.50	36.71
36	13	ESP	Alcobendas	urban	2001-2001	32.00	8.60	26.88	25.00	2.70	10.80	31.40
37	13	ESP	Badajoz	urban	2004-2004	38.00	10.60	27.89	19.00	3.20	16.84	30.19
38	13	ESP	Las Palmas	urban	2001-2001	44.00	11.60	26.36	18.00	3.00	16.67	25.86
39	14	ESP	Alcobendas	urban	2001-2001	32.23	8.28	25.68	24.95	2.56	10.26	30.92
40	14	ESP	Badajoz	urban	2004-2005	32.38	8.56	26.43	18.65	2.54	13.64	29.67
41	14	ESP	Burgos	urban	2004-2005	29.27	5.53	18.88	21.58	2.52	11.69	45.57
42	14	ESP	Iturrama (Pamplona)	urban	2003-2003	29.20	7.32	25.06	---	---	---	
43	14	ESP	Las Palmas	urban	2001-2001	42.93	9.93	23.13	18.40	2.50	13.60	25.18
44	14	ESP	Madrit Escuela Aguirre	urban	1999-2000	47.69	14.99	31.44	34.14	5.41	15.84	36.09
45	14	ESP	Palma de Mallorca	urban	2004-2005	28.79	7.64	26.53	19.94	2.57	12.87	33.64
46	2	SWE		rural		8.00	2.00	25.00	7.00	1.00	14.29	50.00
47	4	IRL	Ahascaragh/ County Galway	rural	2001-2002	10.40	1.00	9.62	6.00	0.20	3.33	20.00
48	5	NLD	Vredepeel	rural	1998-1999	25.30	6.24	24.66	18.40	1.51	8.21	24.20
49	5	NLD	De Zilk	rural	1998-1999	22.40	3.56	15.89	14.10	1.04	7.38	29.21
50	2	NLD		rural		22.00	2.00	9.09	14.00	0.60	4.29	30.00
51	6	BEL	Waasmunster	rural	1994-1995	39.55	3.76	9.51	25.58	0.98	3.82	26.06
52	6	CHE	Chaumont	rural	1998-1999	10.52	1.88	17.92	8.09	0.77	9.53	40.96
53	8	CHE	Payerne	rural	1998-1999	13.70	1.60	11.68	---	---	---	---
54	8	CHE	Chaumont	rural	1998-1999	10.80	1.60	14.81	7.70	0.70	9.09	43.75
55	2	CHE		rural		14.00	2.00	14.29	18.00	1.00	5.56	50.00

ID	Ref.	Country	Name/Location	Location_type	year	PM_{10} [µg/m³]	MD_{10} [µg/m³]	MD in PM_{10} [%]	$PM_{2.5}$ [µg/m³]	$MD_{2.5}$ [µg/m³]	MD in $PM_{2.5}$ [%]	$MD_{2.5}/MD_{10}$ [%]
56	6	AUT	Illmitz	rural	1999-2000	23.92	1.02	4.25	19.94	0.26	1.30	25.49
57	2	AUT		rural		24.00	1.00	4.17	20.00	0.40	2.00	40.00
58	6	DEU	Melpitz 96-99	rural	1996-1997	24.17	3.34	13.83	16.85	1.80	10.70	53.89
59	6	DEU	Melpitz 99-01	rural	1999-2001	19.39	1.50	7.73	13.63	1.07	7.82	71.33
60	2	DEU		rural		19.00	3.00	15.79	---	---	---	---
61	17	FRA	Plan d'Aups	rural	2001-2001	---	---	---	9.52	1.05	11.00	---
62	15	ITA	Mount Cimone	rural	2004-2004	16.10	4.00	24.84	---	---	---	---
63	12	ITA	Montelibretti / Lazio region	rural	2004-2005	34.20	8.70	25.44	21.90	2.20	10.05	25.29
64	12	ITA	Fontechiari/ Lazio region	rural	2004-2005	31.20	4.70	15.06	23.30	1.60	6.87	34.04
65	2	ESP		rural		21.00	6.00	28.57	14.00	2.00	14.29	33.33
66	6	ESP	Monagrega	rural	1999-2000	17.30	4.14	23.93	---	---	---	---
67	13	ESP	Monagrega	rural	1999-2000	21.00	5.20	24.76	12.00	---	---	---
68	13	ESP	Bemantes	rural	2001-2001	19.00	2.50	13.16	14.00	1.50	10.71	60.00
69	13	ESP	Montseny	rural	2002-2006	20.00	5.50	27.50	17.00	2.00	11.76	36.36
70	16	ESP	Monagrega/Ebro basin	rural	1999-2000	22.02	5.80	26.34	---	---	---	
71	14	ESP	Bemantes	rural	2001-2001	18.88	2.09	11.09	13.47	1.39	10.29	66.51
72	14	ESP	Montseny	rural	2004-2007	16.50	4.07	24.64	13.32	1.25	9.38	30.71
73	14	ESP	Montseny	rural	2002-2007	---	---	---	13.57	1.22	8.96	---
74	14	ESP	Montseny	rural	2002-2002	---	---	---	14.00	1.34	9.54	---
75	14	ESP	Montseny	rural	2003-2003	---	---	---	14.36	0.92	6.41	---
76	14	ESP	Montseny	rural	2004-2004	19.24	4.50	23.39	16.18	1.76	10.87	39.11
77	14	ESP	Montseny	rural	2005-2005	16.39	3.57	21.80	12.36	0.82	6.61	22.97
78	14	ESP	Montseny	rural	2006-2006	15.06	4.17	27.67	13.45	1.37	10.16	32.85
79	14	ESP	Montseny	rural	2007-2007	15.31	3.97	25.95	11.93	1.15	9.65	28.97
80	14	ESP	M.Perdon (Pamplona)	rural	2003-2003	20.76	4.09	19.71	---	---	---	---
81	6	FIN	Sevettijarvi	nature	1993-1996	4.50	0.30	6.64	3.41	0.15	4.28	50.00
82	6	NOR	Skreadalen	nature	1991-1996	8.01	0.48	5.95	5.05	0.18	3.64	37.50
83	6	NOR	Birkenes	nature	1991-1996	7.94	0.71	8.93	4.92	0.25	5.14	35.21
84	4	IRL		nature	1998-1999	18.60	1.20	6.45	7.40	0.30	4.05	25.00
85	10	ITA	Cornigliano/Genoa	urban-ind.	2004-2005	41.70	3.30	7.91	19.00	3.20	16.84	96.97
86	13	ESP	Cartagena-Santa Ana	urban-ind.	2004-2004	46.00	16.80	36.52	22.00	3.20	14.55	19.05
87	13	ESP	Onda	urban-ind.	2002-2006	28.00	9.50	33.93	20.00	---	---	---
88	13	ESP	L'Alcora	urban-ind.	2002-2006	35.00	16.40	46.86	24.00	---	---	---
89	13	ESP	Barcelona-CSIC	urban-ind.	2002-2006	46.40	11.70	25.22	32.10	4.70	14.64	40.17
90	13	ESP	Huelva	urban-ind.	2001-2001	37.00	11.80	31.89	19.00	2.70	14.21	22.88
91	13	ESP	La Linea	urban-ind.	2002-2004	42.00	10.50	25.00	24.00	2.50	10.42	23.81
92	13	ESP	Llodio	urban-ind.	2001-2001	32.00	7.30	22.81	24.00	2.30	9.58	31.51
93	13	ESP	Puertollano	urban-ind.	2004-2004	43.00	13.20	30.70	29.00	2.90	10.00	21.97
94	13	ESP	Santa Cruz	urban-ind.	2002-2006	38.00	---	---	23.00	5.10	22.17	
95	13	ESP	Tarragona	urban-ind.	2001-2001	37.00	9.20	24.86	22.00	1.90	8.64	20.65
96	18	ESP	Barcelona-CSIC	urban-ind.	2003-2006	47.00	14.50	30.85	31.00	5.10	16.45	35.17
97	18	ESP	Barcelona-CSIC	urban-ind.	2004-2004	32.00	8.80	27.50	21.00	1.80	8.57	20.45
98	14	ESP	Algeciras	urban-ind.	2003-2004	37.74	7.17	19.00	24.64	2.39	9.68	33.33
99	14	ESP	Barcelona (IJA-CSIC)	urban-ind.	2003-2007	45.24	14.32	31.65	30.17	4.83	16.02	33.73
100	14	ESP	Barcelona (IJA-CSIC)	urban-ind.	2003-2004	47.37	13.49	28.48	31.91	4.11	12.90	30.47
101	14	ESP	Barcelona (IJA-CSIC)	urban-ind.	2005-2005	45.26	12.75	28.16	30.97	5.20	16.79	40.78
102	14	ESP	Barcelona (IJA-CSIC)	urban-ind.	2006-2006	45.04	15.32	34.02	29.29	5.13	17.51	33.49
103	14	ESP	Barcelona (IJA-CSIC)	urban-ind.	2007-2007	43.43	15.38	35.41	28.19	5.17	18.34	33.62
104	14	ESP	Bastarrechte (Cartagena)	urban-ind.	2004-2005	47.01	9.95	21.17	---	---	---	---
105	14	ESP	Onda (Castellon)	urban-ind.	2002-2007	26.52	9.20	34.69	---	---	---	---
106	14	ESP	Onda (Castellon)	urban-ind.	2002-2002	25.88	8.46	32.69	---	---	---	---
107	14	ESP	Onda (Castellon)	urban-ind.	2003-2003	26.20	10.07	38.42	---	---	---	---
108	14	ESP	Onda (Castellon)	urban-ind.	2004-2004	28.97	7.81	26.94	---	---	---	---
109	14	ESP	Onda (Castellon)	urban-ind.	2005-2005	27.30	9.46	34.66	---	---	---	---
110	14	ESP	Onda (Castellon)	urban-ind.	2006-2006	24.68	8.89	36.03	---	---	---	---
111	14	ESP	Onda (Castellon)	urban-ind.	2007-2007	26.54	10.16	38.26	---	---	---	---
112	14	ESP	Santa Ana	urban-ind.	2004-2005	---	---	---	22.48	2.86	12.72	---
113	2	SWE		traffic		26.00	17.00	65.38	13.00	4.00	30.77	23.53
114	2	GBR		traffic		35.00	7.00	20.00	24.00	2.00	8.33	28.57

ID	Ref.	Country	Name/Location	Location_type	year	PM_{10} [µg/m^3]	MD_{10} [µg/m^3]	MD in PM_{10} [%]	$PM_{2.5}$ [µg/m^3]	$MD_{2.5}$ [µg/m^3]	MD in $PM_{2.5}$ [%]	$MD_{2.5}/MD_{10}$ [%]
115	4	IRL	College Street/Dublin	traffic	2001-2002	37.30	6.50	17.43	22.40	0.60	2.68	9.23
116	2	NLD		traffic		30.00	6.00	20.00	20.00	0.70	3.50	11.67
117	6	CHE	Bern	traffic	1998-1999	40.22	7.64	18.99	22.65	1.35	5.97	17.67
118	8	CHE	Bern	traffic	1998-1999	40.20	5.80	14.43	24.60	1.20	4.88	20.69
119	8	CHE	Zurich-Wiedikon	traffic	1998-1999	43.00	4.70	10.93	---	---	---	
120	8	CHE	Zurich-Kaserne	traffic	1998-1999	24.40	2.40	9.84	20.00	1.50	7.50	62.50
121	2	CHE		traffic		42.00	6.00	14.29	22.00	1.00	4.55	16.67
122	2	DEU		traffic		37.00	14.00	37.84	25.00	3.00	12.00	21.43
123	6	AUT	Wien	traffic	1999-2000	54.58	5.88	10.77	38.51	2.37	6.15	40.31
124	2	AUT		traffic		53.00	4.00	7.55	38.00	0.70	1.84	17.50
125	12	ITA	Largo di Montezemolo/ Lazio region	traffic	2004-2005	48.30	12.30	25.47	---	---	---	---
126	10	ITA	Multedo/Genoa	traffic	2004-2005	38.80	12.60	32.47	20.60	2.00	9.71	15.87
127	13	ESP	Madrid	traffic	1999-2000	48.00	15.20	31.67	34.00	5.60	16.47	36.84
128	13	ESP	L'Hospitalet	traffic	1999-2000	50.00	12.90	25.80	35.00	4.20	12.00	32.56
129	13	ESP	Barcelona-Sagrera	traffic	2001-2001	46.00	15.20	33.04	28.00	4.20	15.00	27.63
130	18	ESP	L'Hospitalet	traffic	1999-2000	50.00	12.90	25.80	34.00	3.80	11.18	29.46
131	16	ESP	L'Hospitalet/ Barcelona	traffic	1999-2000	49.51	12.50	25.25	33.92	3.40	10.02	27.20
132	2	ESP		traffic		45.00	16.00	35.56	28.00	5.00	17.86	31.25
133	6	ESP	Barcelona	traffic	1999-2000	52.85	13.71	25.95	34.61	3.79	10.96	27.64

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The contribution of mineral dust to ambient particulate matter

Mineral dust is a constituent of atmospheric particulate matter (PM_{10}). It originates from diverse sources, such as wind erosion, agricultural land management and resuspension by road transport. These sources are not included in emission inventories of primary PM_{10}. Until recently, mineral dust was assumed to be one of the major missing sources that would explain the gap between modelled and observed concentrations of PM_{10} in the Netherlands. This report assessed the importance of mineral dust in PM_{10} for the Netherlands and Europe, using chemical analyses of Dutch PM_{10} samples, a compilation of observational data from across Europe, and a regional atmospheric chemistry and transport model. Ambient particulate matter in Europe typically constituted of 5 to 20% mineral dust. The average concentration of mineral dust in PM_{10} at six locations in the Netherlands was approximately 1 to 1.5 µg/m^3. This is substantially less than reported by previous studies. The increased level of mineral dust in PM_{10} samples in the urban environment is still not properly understood, and nor are the differences between the Netherlands and its surrounding countries. An important conclusion of this report is that the contribution of mineral dust to PM_{10} in the Netherlands is less important than previously thought.

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