A methodology to constrain the potential source strength of various soil dust sources contributing to atmospheric PM10 concentrations

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Abstract

Crustal material or soil particles typically make up 5 - 20 % of the mass of ambient PM10 samples. In certain regions and/or specific meteorological conditions the contribution may even be higher. Crustal material may originate from distinctly different sources e.g., 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 crustal material in total PM10 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., erosion) whilst others like re-suspension are not recognized as primary emission but a re-emission. Separating the source contributions is difficult because the unique tracers for crustal material in PM10 samples (e.g., Si, Ti) do not allow a distinction between the potential sources of this material. To make progress in our understanding of the crustal material source strengths we need a combination of flux measurements, emission estimates, chemical analysis of ambient PM10 samples and atmospheric transport modelling. In this paper we present a methodology to check first order estimates of the various source strengths. Simple and therefore transparent emission functions are combined with activity data or land use maps to make emission grids. The gridded emissions are used as input for the LOTOS-EUROS model to calculate the resulting concentrations. The predicted concentrations will be compared with observations in various parts of Europe derived from the literature. This will give an indication if the source strengths are in the right order of magnitude to explain observed crustal material contributions to ambient PM10. Next, by varying the source strengths of the individual categories and implementation of a meteorological dependency we will investigate if the patterns of the predicted concentrations are in line with observations.

Keywords: particulate matter, soil particles, resuspension, wind ersosion, crustal material, emissions, air quality

1 The relevance, chemical composition and sources of crustal material.

Particulate matter (PM) is the generic term used for a type of air pollution that consists of complex and varying mixtures of particles suspended in the atmosphere that has been found to present a serious danger to human health. Particulate pollution comes from such diverse sources as coal or oil fired power plants, vehicle exhaust, wood burning, mining, and agriculture. Airborne PM includes many different chemical constituents. PM10 samples collected on a filter can be analyzed for chemical composition. Sources contributing to the PM10 concentrations may emit particles with a unique chemical composition that are enriched, relative to particles from other sources, in certain elements. For example, vanadium and nickel have typically been used as tracers for emissions from fuel oil combustion. Based on the concentrations of the tracers an estimate of the total amount of PM10 emitted by the particular source can be made. Crustal material (mineral matter, soil particles) typically makes up 5 - 20 % of the mass of ambient PM10 (table 1).

In certain regions and/or specific meteorological conditions the contribution may be higher. This is illustrated in table 1 with the values for Northern EU and Southern EU where the contribution is elevated due to the use of studded tires and desert dust, respectively.

Crustal material may originate from distinctly different sources e.g., 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 (figure 1). Despite the importance of crustal material in total PM10 mass, the sources are still poorly understood as well as their relative contribution to total crustal PM10. In this paper we present a methodology to check first order estimates of the various source strengths (figure 1). Simple, transparent emission functions are combined with activity data or land use maps to make emission grids.

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Table 1:
Mean annual levels (µgm^{-3}) of PM10, PM2.5, mineral elements, and the equivalent contributions to bulk mass concentrations (% wt), recorded at regional background (RB), urban background (UB) and kerbside stations (RS) in Central EU (examples from Australia, Berlin, Switzerland, The Netherlands, UK), Northern EU (13 sites in Sweden) and Southern EU (10 sites in Spain).

	Central EU			Northern EU)			Southern EU		
	RB	UB	RS	RB	UB	RS	RB	UB	RS
PM10 (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 PM10	5 – 10	10 – 15	12 – 15	20 – 30	35 – 45	65 – 70	12 – 40	25 – 30	25 – 37
PM2.5 (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 PM2.5	2 – 8	2 – 8	5	15 – 25	25 – 30	30 – 40	8 – 20	10 – 20	10 – 15

Source: Querol X. et al (2004)

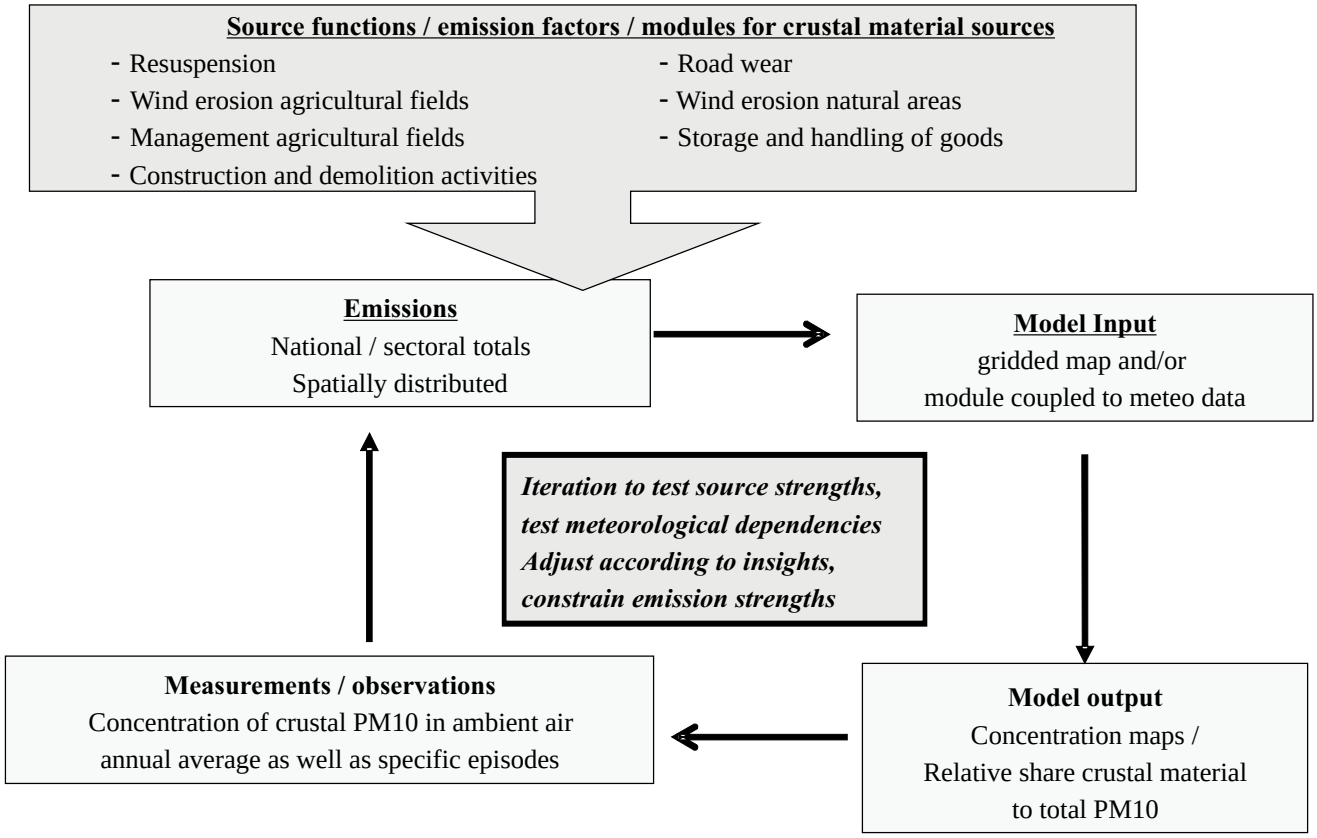

Figure 1:
Outline of the methodology to constrain the source strength of various crustal material sources contributing to PM10 using an atmospheric chemistry and transport model and observational data

2 Emission functions and source strengths

Eventually all different sources of crustal material (figure 1) should contribute their own unique emission pattern and strength to the gridded data going into the atmospheric transport model. So, for each of these emission causes an emission estimate and/or module that calculates emission within the model needs to be constructed. To illustrate the methodology we present three examples.

- Windblown dust from bare soils; A wind velocity driven factor is applied to all bare soils following the parameterization of van Loon M. et al. (2005). This is a simplified version of paramterizations developed to predict wind blown dust from deserts. As a first approximation we applied this function to all arable lands according to the CORINE land use map (CORINE, 2003) but at only 1/50 of the source strength because arable lands will have a lower emission than continuous bare soils or deserts. Furthermore we assume no emissions occur on rainy days. Next, sensitivity studies will be done by increasing the bare soil area and adjusting the source function. Obviously our first assumptions may easily be an order of magnitude wrong and comparisons with measurement data in particular regions will be used to tune the source strength.
- Road wear or pavement wear; Pavements consist mainly from rock material and the contribution of pavement wear to the total PM10 emission factor is difficult to separate from the contribution of windblown- or resuspended soil and other geological material. However in a road tunnel the amount of windblown soil dust coming from the shoulders or elsewhere will be limited and a first approximation can be that all the crustal material in PM10 produced in the tunnel originates from road wear. Gillies J. A. et al. (2001) reported that ~12 % of the PM10 in the Sulpeveda tunnel was of geological origin. Denier van der Gon H. A. C. et al. (2003) used this observation in combination with the chemical analysis of PM10 formed in the the Maas tunnel (Rotterdam, The Netherlands) to derive an emission factor of 3 - 4 mg per vehicle kilometre (vkm) for crustal material from road wear. Road wear will be dependent on the pavement material but also on the frequency of braking and cornering causing additional friction. In the tunnel environment this will be limited compared to the average urban traffic. Hence we see the tunnel road wear emission factor as a lower value. In the literature road wear emission factors are often in the range of 8 - 10 mg/vkm which is consistent with 3 - 4 mg/vkm being a lower limit.
- Resuspension by traffic; The material that is being resuspended will vary with the nature of the local circumstances but it is typically likely to include particles from vehicle tyre and brake dusts, primary exhaust emissions that have settled out of the atmosphere (perhaps adhering to larger particles) and environmental dusts from many sources eg pollen, sea salt, construction work and wind blown soils. Furthermore the contribution of resuspension to total PM will be depedendent on the meteorological conditions; during rain and/or while the surfaces are wet the resuspension will be much lower. The mix of particles with varying chemical composition, dependencies on local and climatic conditions make this a difficult source to quantify and generalize. However, it is widely acknowledged that resuspesion is potentially a very important source in the same order of magnitude as traffic exhaust emissions. In our first, explorative approach we take an emission factor of 80 mg/vkm and than selectively switch this off on rainy days.

It should be noted that road wear and resuspension will both be coupled vehicle kilometres driven, hence they cannot be separated. Furthermore, as discussed above, not all resuspension will be crustal material. Therefore the 80 mg/vkm that we use as a first approximation for crustal PM10 from traffic contains only the crustal fraction of resuspension emissions and includes road wear.

3 Spatially distributed emission maps

The basic procedure to come to a high resolution gridded emission map that can be used as input for a model is to redistribute total national sectoral emissions using high resolution distribution patterns, also called proxi data. The TNO proxi data consist of recent geographical distribution data for point sources and area sources and are described by Visschedijk A. H. J. and Denier van der Gon H. A. C. (2005). For example, in the present study the emission maps for crustal material from road wear and resuspension are made by calculating the total amount road wear and/or resuspension from annual amount of kilometres driven by vehicle category (e.g. heavy duty vehicles, light duty vehicles and passenger cars) within a country taken from the CAFE base line scenario (Amman M. et al., 2005) and split in a highway, urban and rural part. For the spatial distribution of the emissions the highway part is distributed according to digitized maps of highways, using traffic intensities by highway section for LDV and HDV from Eurostat (Eurostat, 2003). The rural part of the road transport emission is distributed according to the digital maps of the European rural road network. The urban part of the road transport emissions is distributed according to a high resolution population density map (CIESIN, 2001).

4 Model description

We have used the LOTOS-EUROS model (Schaap M. et al., 2007) to calculate the crustal material concentrations in ambient air over Europe using meteorological data from 1998 and 1999. The LOTOS-EUROS model is a 3D-chemistry transport model developed to study the formation, transport and sinks of oxidants, particulate matter and heavy metals. LOTOS-EUROS is applied for the region that spans from 10 °W to 40 °E and from 35 °N to 70 °N with a spatial resolution of 0.5 x 0.25 degrees lon-lat, roughly corresponding to 25 by 25 km. The model

has been applied in numerous studies for particulate matter (Schaap M. et al., 2004; Schaap M. et al., 2007). The modelled PM10 concentration over Europe with the LOTOS-EUROS model in the year 2003 is shown in figure 2. The modelled concentrations can be compared with measured concentrations and showed that the model follows the temporal patterns correctly but that the absolute concentrations are systematically underestimated. This is explained by the omission of natural sources other than sea salt and resuspension emissions in the model input. When a comparison is made for an almost exclusively anthropogenic component like PM2.5 sulphate aerosol, both absolute concentrations and temporal patterns are predicted correctly by the model.

The model input in this study are gridded maps of crustal material emitted as PM10 with 80 % of the particles in coarse mode (PM2.5 - 10) and 20 % of the particles belong to the fine mode (PM2.5). The tracers for crustal material in the model are chemically inert and deposition is modelled as fine and coarse mode particles. For a full model description we refer to the model documentation by Schaap M. et al. (2007).

5 Mining observational data to get additional information

In our approach (figure 1) we have too many unknowns to be directly successful. However we can derive important hints from the observational data. For example, a remarkable feature of the data compiled in table 1 is the strong elevation of crustal material at the kerbside stations, suggesting an important contribution of (resuspended) crustal material by traffic. This implies that if we intend to constrain e.g., the source strength of crustal material emissions due to wind erosion using models and observational data, this cannot be done without including traffic emissions.

5.1 Ambient PM10 concentration of crustal material in the Netherlands

For the Netherlands additional information about crustal material in ambient PM is derived from the data collected in the bronstof study (Visser H. et al., 2001). Visser H. et al. (2001) investigated the composition and origin of airborne particulate matter in the Netherlands by collecting samples of the fine fraction (PM2.5) and the coarse fraction (PM2.5 - 10) at six sites during one year (1998 - 99). The samples were analysed for chemical composition, for a description of the measurement procedures and analytical specifications we refer to Visser H. et al. (2001). These data will be used to gain information about the nature of

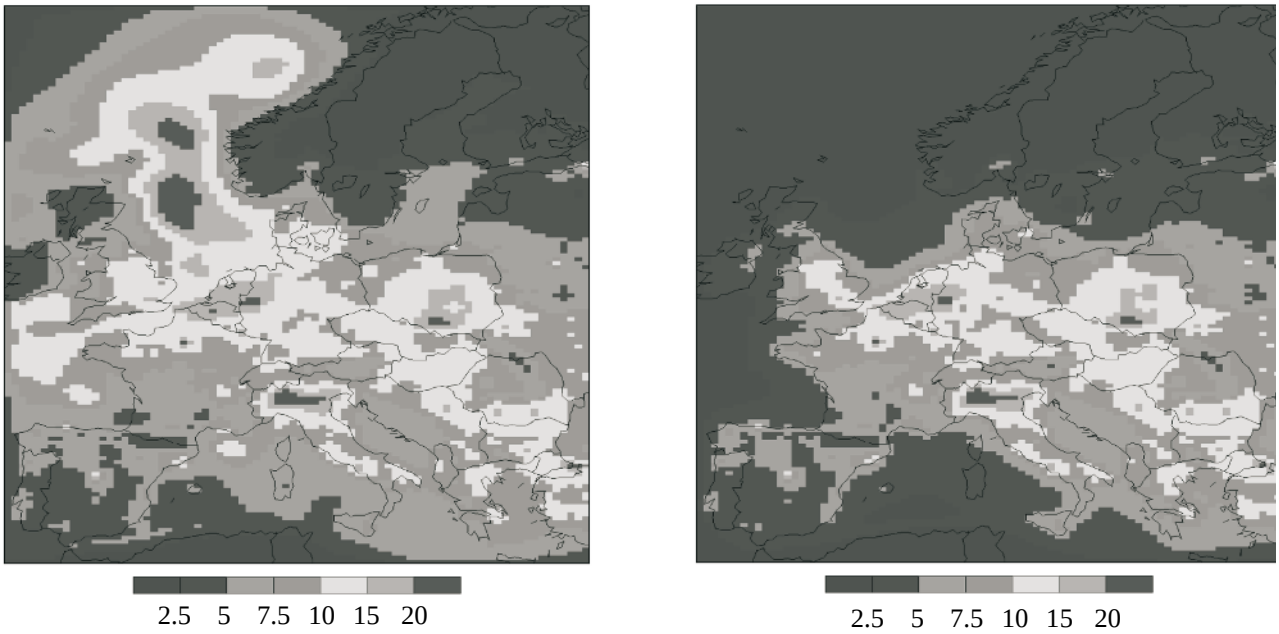

Figure 2:

Modelled PM10 concentration over Europe in 2003, Left PM10 including sea salt aerosol but excluding other natural soil dust emissions and resuspension; right anthropogenic PM10

the CM sources contributing to ambient PM10 in the Netherlands. A number of elements are known to be present in crustal material, the most important being Si, Al, Ca, K, Fe and Ti The strong correlations between these elements in PM samples analyzed by Visser H. et al. (2001) suggest they originate from the same source, especially Al, Si and Ti (data not shown). Clay minerals in soils and rock have complex aluminosilicate structures 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. Si and Al are the most abundant elements in crustal material and would be the most robust tracers. A formula [1] was derived, based on the chemical composition of the fine fraction of Dutch top soils to calculate the contribution of crustal material (CM) using the concentrations of Al and Si.

$$\text{Mass CM} = 0.49*[Si] + (2.36*[Si]+2.70*[Al]) \qquad [1]$$

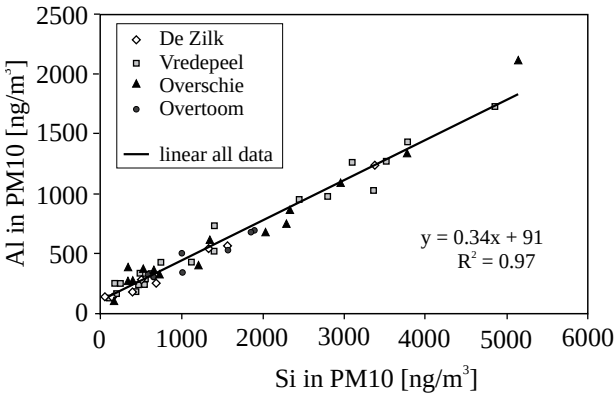

Figure 3:
Aluminum (Al) concentration as a function of Silicium (Si) in PM10 at four locations in the Netherlands (based on data from Visser H. et al., 2001)

The number of analysis days in the study of Visser H. 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 3) is used to calculate soil material on the basis of Si alone if no Al data where available (table 2). The average concentration of CM in PM10 for the Netherlands is ~2.5 at the coast to ~ 5.5 µg/m³ at the inland sites.

The contribution of crustal material to ambient PM10 at the Dutch sites is wind direction dependent (data not shown). However, the number of observations available to calculate a wind direction specific contribution is limited. To approximate the wind direction dependency and at the same time cover a reasonable number of observations the samples have been split into a continental/land and sea derived origin (table 3). The air masses with a more continental/land origin show an elevated crustal material component compared to the sea derived air masses (2.9 vs 1.8 µg/m³ for de Zilk and 8.6 vs 3.0 µg/m³ for Vredepeel, respectively). This may help us to identify source regions using trajectories in the models for the particular days with elevated concentrations as well as investigating the dependency on meteorological conditions.

Table 2:
Average annual contribution of CM to ambient PM10 concentrations at four locations

Location method	DeZilk CM_alt	Vredepeel CM_alt	Overschie CM_alt	Overtoom CM_alt
number observations	31	45	42	49
avg (µg/m3)	2.4	5.4	5.0	3.3
sd	2.5	4.7	3.9	2.2

CM_alt = like CM eq[1] but missing Al data are estimated based on the Al to Si correlation (figure 3)

5.2 Size fractionation of crustal material in PM10

Another important feature that is needed for the models to predict the contribution of particulate emissions to ambient concentrations is the size fractionation. The smaller size fractions are transported of longer distances and have a longer life time due to a slower deposition rate. We have started our investigations assuming that 20 % of the CM is in the fine fraction (PM2.5) of PM10. These assumptions

Table 3:
The crustal material contribution to PM10 at de Zilk and Vredepeel at different wind directions

	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 (µg/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

can be refined using information from measurements. The average fraction of PM2.5 at the inland site (Vredepeel) and coastal site (De Zilk) is 15 % and 21 %, respectively. However, a closer look reveals that this fraction is not stable but changes as a function of the total CM content (figure 4). So, the more important the CM contribution, the less relevant is the PM2.5 fraction. This indicates that events of elevated CM contributions are probably highly controlled by sources in the vicinity. In future sensitivity studies we will vary the size fraction to see how this influences the sources contributing to our receptor points. We need to further analyze the data to see if we can use this information to exclude certain sources as being important for the Netherlands.

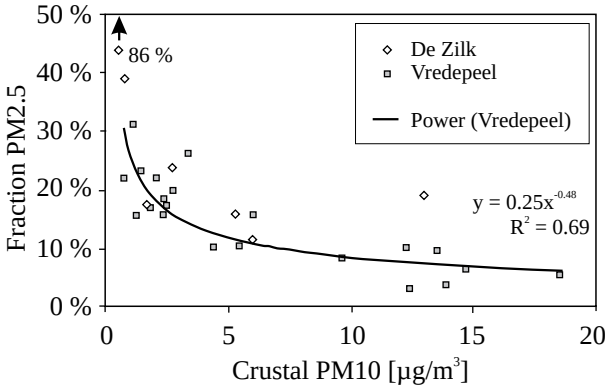

Figure 4:
The fraction of PM2.5 in crustal PM10 for two Dutch background stations

6 First results and further steps

The predicted concentration of crustal PM10 due to windblown dust from arable fields is presented in figure 5. This first approximation results in annual contributions to PM10 of ~ 0.2 µg/m^3 in the Netherlands. This is small compared to annual observed concentrations of 2 - 5 µg/m^3. However, it should be noted that our emission function does not yet include emission due to land management and, as was explained earlier, is highly uncertain. The patterns in the map reflect a combination of where arable lands are and arid regions because the emission is switched off on rainy days.

The predicted concentration of crustal PM10 due to resuspension and roadwear by traffic is presented in figure 6. Here the patterns reflect where most kilometres are driven (big cities and densely populated regions) and predict a contribution of traffic to crustal PM10 in the Netherlands of 0.3 - 1 µg/m^3, again modest compared to the observed annual concentrations. Further investigations of published road side measurements may help us to better define realistic estimates of the resuspension emission factor and the fraction of crustal material therein. Furthermore we may vary the resuspension emission factor between highways, urban areas and rural, secondary roads. It is to be expected that a car or lorry driving over a rural road is causing more resuspending of dust than the same vehicle being one of the 4.000 vehicles / hr passing a busy highway. It should be investigated how sensitive the patterns of the predicted concentrations are to such modifications and how sensitive they are to certain climatic conditions.

7 Conclusions and Outlook

Crustal material is an important component of the PM10 and especially PM2.5 - 10 concentrations observed in the Netherlands. It originates from natural and anthropogenic sources. A better understanding of the sources contributing to the crustal fraction in PM10 is important for understanding the ambient PM concentrations because the fraction of PM that can be influenced by abatement measures in future policy plans should include the anthropogenic sources of the crustal fraction such as resuspension of dust by traffic.

Despite the importance of crustal material in total PM10 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., erosion) whilst others like re-suspension are not recognized as primary emission but a re-emission. Separating the source contributions is difficult because the unique tracers for crustal material in PM10 samples (e.g., Si, Al, Ti) do not allow a distinction between the potential sources of this material. To make progress in our understanding of the crustal material source strengths we need to combine information from different disciplines. We present a methodology to combine the of information available from flux measurements, emission inventories, geographical information systems, atmospheric transport models,and chemical analysis of ambient PM10 samples to explore and constrain source strengths of the various sources of crustal PM10. Crustal material is dominantly found in the coarse fraction of PM10 (figure 4). Therefore, the coarse fraction (PM2.5 - 10) is the best fraction to study using the model. Even more so, because the coarse fraction is transported over shorter distances implying that the distance of the receptor site (where concentrations are measured) will be relatively close to source of origin.

Source functions and emission maps for other crustal PM10 sources need to be made and the predicted concentrations can be superimposed on each other. Given the uncertainty is our first approximations and that the sources will add up we can conclude that it is certainly feasible to come close to observed values. For most of the crustal

PM10 sources climatic conditions like precipitation and wind are controlling factors. The atmospheric dispersion models can be run with different years to match the moments of campaigns with sufficient chemical analytical data. For example, the maps in figure 5 and figure 6 are the result of using the meteorological data of 1998 and 1999 to match the sample dates of the Visser et al. (2001). In this way we may be able to use chemical composition data for different years from different regions.

The temporal and spatial patterns of observations and model predictions will give important clues to whether the balance between sources and the timing of there emissions is correct in our appraoch. Within the uncertainties surrounding this exploratory approach the balance in source contributions can be optimized to fit the observations in terms of absolute annual concentrations as well as the temporal patterns over the year. The result is a constraint on the total source strength of crustal material and an indicative ranking of the importance of contributing sources in various areas of Europe.

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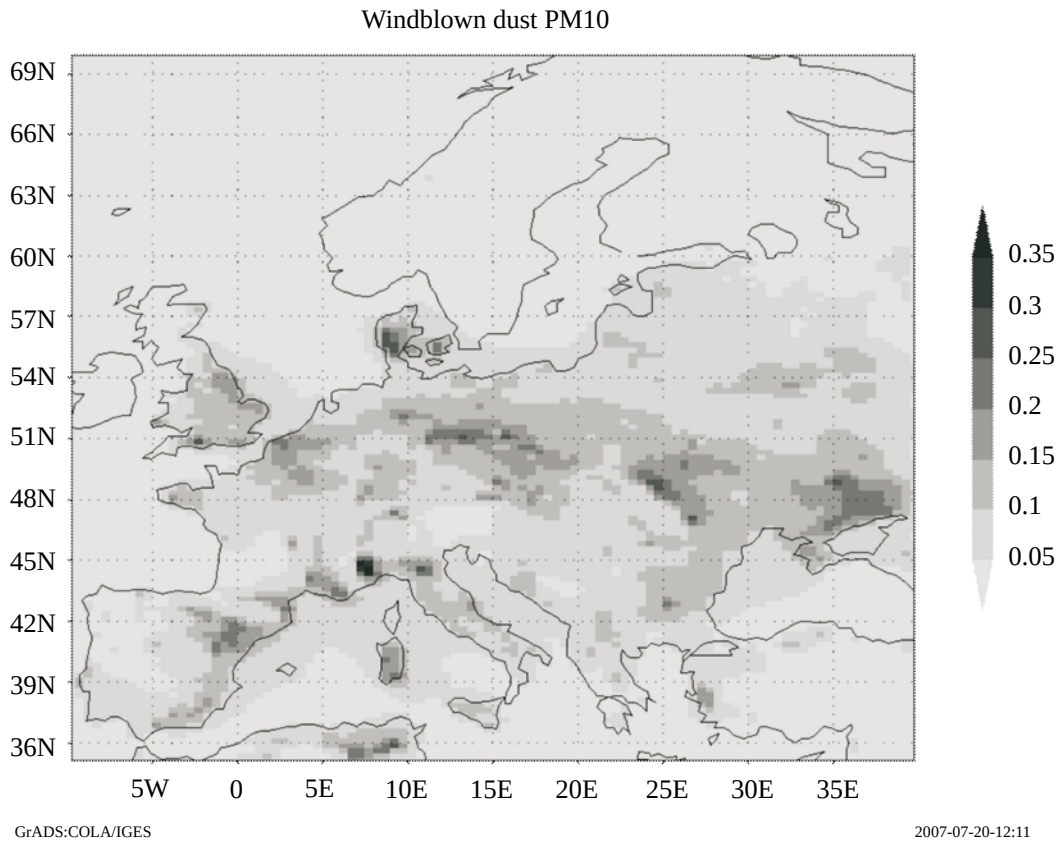

Figure 5:
Predicted crustal PM10 over Europe using a first approximation of the windblown dust from arable lands

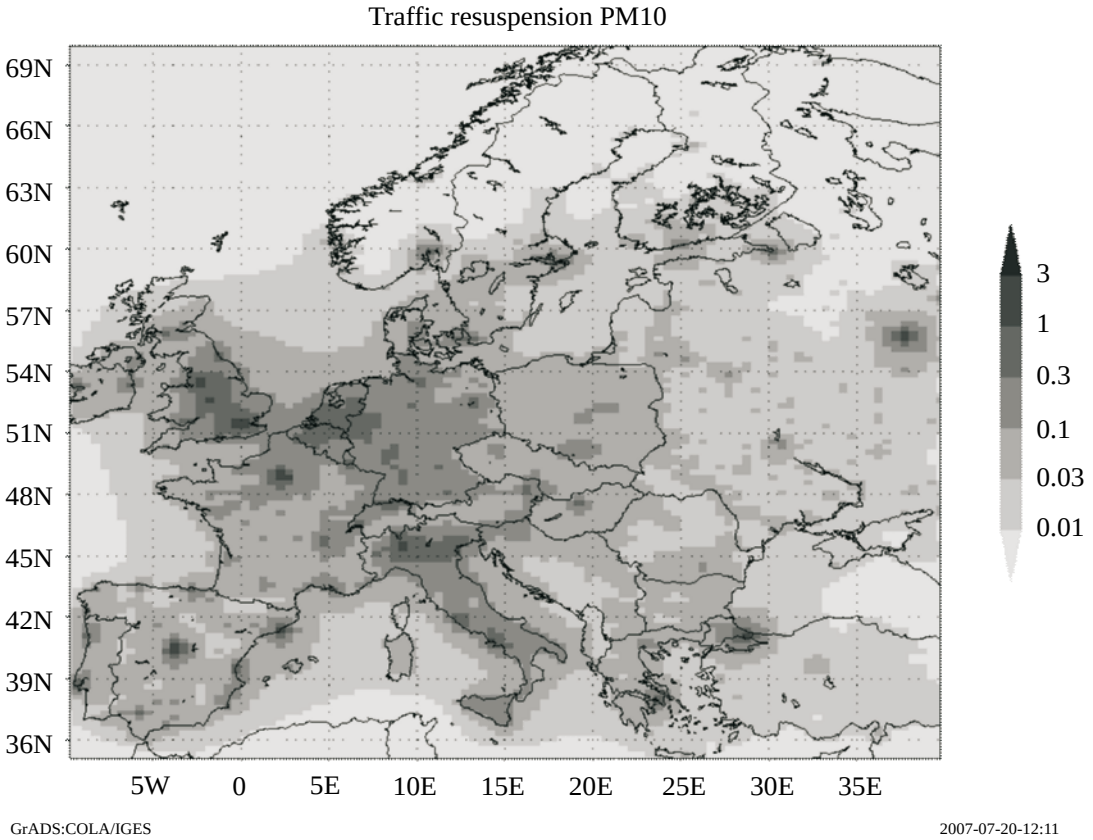

Figure 6

Predicted crustal PM10 over Europe using a first approximation of resuspended crustal material by traffic

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