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A literature study of some anthropogenic and natural sources of particulate matter in the atmosphere

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1. Introduction

Recent studies have stressed the importance of atmospheric particulate matter in relation to human health and environmental issues like climatic change and acidification. On the purpose of outlining a policy by the Ministry for Housing, Regional Development and the Environment (VROM) on ambient particulate matter, a need exist for additional information on natural and anthropogenic particulate emissions on a European scale. The National Institute of Public Health and the Environment (RIVM) fulfils a prominent role with respect to supporting the ministry in realising its policy plan. For certain parts of this task RIVM, in turn, requires further assistance in the form of specific expertise and has required TNO to inventory and specify anthropogenic emissions of anthropogenic particulate matter in Europe. Results are presented in Berdowski et al. (1997). Abatement efficiencies and techniques for controlled particulate emissions in Europe are presented in a separate report (Visschedijk et al., 1997). In addition, a literature study was required on some expected important natural sources (sea salt and soil dust) and some specific anthropogenic sources (tyre wear and other traffic dust, wood stoves and fireplaces). This report presents the results of this literature study. The main focus is on the contribution of mentioned sources to ambient mass concentrations. Moreover, the size distribution and chemical composition of the emitted particles is assessed.

2. Pentagon parameters

2.1 Definitions and inter-relationships

Suspended particulate matter (also called aerosol), is a mixture of solid and/or liquid particles dispersed in air. The total suspended particulate matter (TSPM) is the total mass of all particulate matter per unit volume of air. Particles with diameters between approximately 0.01 and 100 μm are known as the total suspended particulates (TSP). Usually TSP can be considered a good measure of TSPM. Particulate matter with diameters less than 10 μm, 2.5 μm and 0.1 μm are referred to as PM_{10}, $PM_{2.5}$ and $PM_{0.1}$, respectively. PM_{10} can penetrate into the thoracic compartment of the human respiratory tract. $PM_{2.5}$ and $PM_{0.1}$ are also called fine particles and 'Aitken' particles, respectively. Particulate matter in ambient air consists of primary and secondary aerosols. Primary aerosols are emitted directly into the atmosphere as a result of combustion and mechanical processes. Secondary aerosol ($PM_{Secondary}$) is formed in the atmosphere mainly as a result of conversion of primary emissions. Carbonaceous aerosol ($PM_{Carbonaceous}$), also called black smoke, usually consist of particulate matter with diameters less than approximately 5 μm (Annema et al., 1996). PM_{10}, $PM_{2.5}$, $PM_{0.1}$, $PM_{Secondary}$ and $PM_{Carbonaceous}$ are called the Pentagon parameters.
Suburban and large scale air pollution situations in the Netherlands can be characterized by a PM_{10} fraction greater than 0.8 times TSP (EU, 1990), whereas in urban/industrial and/or very windy areas (the coastal area) the PM_{10} fraction approximates 0.55-0.65 times TSP. These findings are in agreement with results of earlier measurements made in the Netherlands when PM_{10} approximated 0.7 times TSP as an annual average (ref.) and with measurements made in urban, rural and alpine air in Switzerland (Monn et al., 1995). In the latter study the ratios between PM_{10} and TSP approximated 0.60-0.75, with the highest values in the most polluted urban areas. During the 1980s PM_{10} in the Netherlands was composed of $PM_{Secondary}$ and $PM_{Carbonaceous}$ in a ratio of approximately 1:1. On an annual basis $PM_{Secondary}$ and $PM_{Carbonaceous}$ each contribute 15-30% to TSP. During episodes of increased air pollution levels during summer and winter this contribution can be as large as 35-45% (Annema et al., 1996).

2.2 Sources

Particles in the coarse mode (larger than 2.5 μm) are primarily produced by mechanical processes, such as attrition or resuspension. Examples for vehicle traffic are wear of tyres, road surfaces and brakes, and examples in industry include storage and transhipment (e.g. coal) and manufacture of building materials. The principal natural sources are windblown soil, sea spray, pollen, and biomass burning (Annema et al., 1996). Usually crustal material and/or sea-salt aerosol make up the largest part of the mass within the coarse mode. Particles in the fine mode (smaller

than 2.5 μm) are generally of anthropogenic origin, and are generated in various high-temperature combustion processes (vehicular traffic, industry, space heating etc.) and welding and soldering, smelting of metals. A variety of natural sources including combustion (e.g. forest fires and volcanic eruptions) and non-combustion processes (e.g. erosion of the earth's crust, production of sea spray, and biological mobilization) may also be significant. Particles in the very fine mode (0.01-1 μm in diameter) are mainly formed by gas-to-particle conversion, the process by which vapour molecules diffuse to the surface of a particle and are subsequently incorporated into the particle (e.g. Seinfeld, 1986). Secondary aerosol is formed in the atmosphere mainly as a result of conversion of primary emissions of sulphur dioxide (SO_2), nitrogen oxides (NO_x) and ammonia (NH_3), and to a lesser extent of hydrocarbons. Carbonaceous particles are mainly emitted by fossil fuel combustion and biomass burning (Cachier, 1992; Penner, 1995). A recent exploratory study conducted by the RIVM in Dutch towns shows that the present carbonaceous aerosol is predominantly diesel exhaust rather than, as in the past, industrial and domestic coal smoke (Annema et al., 1996). The natural emissions of carbonaceous particles result from two sources: photochemical conversion of the gaseous emissions from vegetation to species with low vapor pressures and direct emission of particles from plants.

2.3 Physicochemical behaviour

The development of the particle size spectrum undergoes through successive stages, dominated by nucleation, coagulation, and heterogeneous condensation, in that order (Warneck, 1986). All three processes take place concurrently. Nucleation is the formation of molecular clusters due to weakly attractive forces between the molecules, the Van der Waals forces. Aerosol particles tend to coalesce when they collide with each other. Collision between particles lead to the formation of a new particle of larger size. This process, called coagulation, causes the size distribution to change in favor of larger particles. Coagulation must be distinguished from condensation, which describes the deposition of vapor-phase material onto particulate matter, a process called heterogeneous condensation. In the absence of preexisting particles, condensation leads to the formation of new (Aitken) particles, provided the vapor pressure of the condensing substance is sufficiently high. This process is usually termed homogeneous nucleation or gas-to-particle conversion.

Individual particles interact with water vapor. This interaction may range from the partial wetting of an insoluble dust particle to the complete dissolution of a salt crystral, such as sodium chloride derived from sea salt. In the course of time, however, coagulation, condensation and in-cloud modification processes cause even an insoluble siliceous particle to acquire a certain share of water-soluble material. Although there will always be particles that have retained their source characteristics, one may assume that in general the aerosol comprises a mixture of both water-soluble and insoluble matter. At sufficiently high humidities, water soluble parti-

cles turn into concentrated-solution droplets. Aerosol particles are of great importance for the formation of fogs and clouds by acting as condensation nucleï (Warneck, 1986).
Particles in the coarse mode are neutral to (slightly) alkaline. This means that acidic gases such as SO_2 and HNO_3 can become adsorbed onto coarse particles. Particles in the fine mode are generally neutral to (slightly) acid. The greatest mass contributions to the fine particle mode are usually from sulphate and organic material, with a smaller contribution from nitrate (Heintzenberg, 1989). The acidic species are often wholly or partially neutralised by ammonia, scavenged from the gas phase (Clarke, 1987). Elemental carbon, primarily from incomplete combustion, makes a significant contribution to the fine mode, providing typically 5-10% of the $PM_{2.5}$ mass (Heintzenberg, 1989; Lowe et al., 1996).
Of the secondary aerosol in the Netherlands, 10% consist of sulphate, 25% of nitrate and 65% of ammonium on a molar basis. This means that an excess of ammonium is present, so that NH_4NO_3 and $(NH_4)_2SO_4$ will be the predominant salts in secondary aerosol. Acid aerosols concentrations are low in the Netherlands. For this reason secondary aerosol is more or less neutral in the Netherlands (Annema et al., 1996). Secundary aerosol looks white because it does not absorb light.
Carbonaceous particles consist of highly polymerized organic material with a low content of hydrogen and oxygen. This aerosol is usually divided into two fractions, black carbon (BC) and organic carbon (OC). Black carbon particles are defined on the basis of their strong absorption of solar radiation and their refractive behaviour to thermal and chemical attacks (Wolff and Klimish, 1982; Novakov, 1982). Observations in both remote and source areas indicate that the OC fraction of the carbonaceous aerosol is always larger than the BC fraction (Cachier, 1995).

2.4 Deposition velocities

Dry deposition of particles depends on both the aerodynamic properties of the particles and the interaction between boundary layer and the surface. This interaction depends on meteorological conditions (such as wind speed, friction velocity, relative humidity, temperature) and surface condition (roughness length, zero plane displacement height, canopy structure, chemical/biological/electrostatic properties, geometry, LAI, wetness). Several model, wind tunnel and field studies have shown deposition velocities to grass and other relatively smooth surfaces that vary from less than 0.1 $cm.s^{-1}$ to several $cm.s^{-1}$, depending on particle size (Chamberlain, 1967; Sehemel, 1980, Nicholson and Davies, 1987; Davidson and Wu, 1989). The lowest deposition velocities are reported for particles with diameters between 0.1 and 1 μm (Figure 1). the deposition onto vegetation is species dependent because many plants differ in structure and geometry of roughness elements, e.g. through pubescent hairs or other micro-structural elements which may intercept small particles.

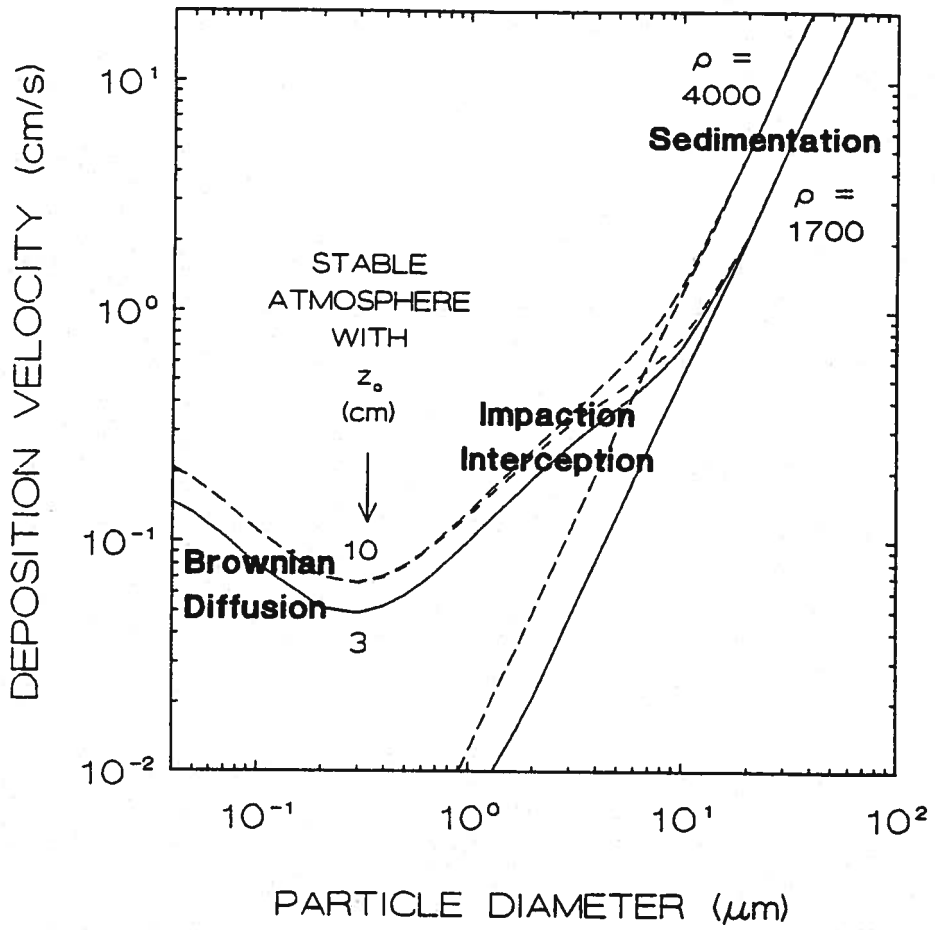

Figure 1 *Typical curves for the deposition velocity of particles as a function of size in a stable atmosphere for two different roughness lengths (z_0 in cm) and particle densities (ρ in $kg.m^{-3}$). The curves were calculated with the model of Sehmel and Hodgon (1980) assuming a wind speed of 5 $m.s^{-1}$.*

The most important parameter determining the dry deposition rate of particles is their size. Large particles (i.e ø > 10 μm) deposit under the influence of gravity and do not respond rapidly to turbulence-induced air motions. Contrary to this, fine particles are not influenced by gravitational settling and are transported mainly as a result of turbulent diffusion. The deposition of particles with diameters smaller than 1μm to vegetative surfaces can be considered a two-stage process: turbulent transport to the viscous sub-layer, followed by transport through this layer surrounding vegetation elements. A number of processes can be responsible for crossing the sub-layer. Brownian diffusion, resulting from the random movement due to the thermal energy of the molecules, is most efficient for very small particles with diameters less than 0.1 μm. There particles may also be deposited by phoretic processes or electrical interactions. These processes are generally found to be of minor importance compared to other processes (Chamberlain, 1960). This also holds for processes such as thermophoresis, i.e. transport along a temperature gradient, and diffusiophoresis, i.e. motion of a particle caused by non-uniformities in the suspending gas. For larger particles, near 1 μm, inertial impaction and inter-

ception become important deposition mechanisms. Interception occurs when particles moving with the mean air motion pass sufficiently close to an obstacle to collide with it, while impaction occurs when particles cannot follow rapid changes in direction of the mean air flow. Gravitational settling is only important for very large particles, i.e. larger than 10 μm (Erisman et al., 1994; Davidson and Wu, 1989)
Most up-to date parametrisations for the dry deposition velocity of fine and coarse particles are presented in Erisman and Draaijers (1995).

2.5 Indicators

Monn et al. (1995) found in urban areas in Switzerland SO_2 and NO_2 well correlated with PM_{10} suggesting similar emission sources. Emission sources of SO_2 were predominately domestic heating and, to a minor extent, diesel exhaust emission. Emission sources of NO_2 are mainly automobile traffic and in winter domestic heating. In rural areas the correlations with SO_2 and NO_2 were much weaker. Among several meteorological parameters, temperature correlated best (negatively) with PM_{10}. A negative correlation was also found between PM_{10} and relative humidity showing that an increase of humidity leads to a decrease of PM_{10}. This was explained by washout-processes of particles from the atmosphere.
Clark (1996) found the seasonal variation in PM_{10} concentrations in urban areas in the United Kingdom driven by meteorology. Winter episodes occurred during very cold stable weather and during windless foggy weather combined with local bonfire. In the first situation all other air pollutants concentrations were also increased, most notably SO_2 and NO_2. During the latter situation especially CO levels were elevated. A typical summer episode is characterised by high ozone levels. PM_{10} concentrations were found increased, among others, by photochemically derived particles. The concentrations of ozone and PM_{10} became elevated together during summer episodes only when polluted air had been circulating over the UK during a period of several days of hot, sunny weather. The PM_{10} diurnal pattern was found to exhibit a morning peak around 8-10 am, decreasing during the day then giving a smaller peak around the evening rush hour. Diurnal variations were most closely related to those of NO_2. This was assumed caused by traffic being the primary source of PM_{10}. Resuspension of road dust was also assumed to contribute to the diurnal pattern.
On the basis of knowledge of air pollutants such as SO_2 and NO_2 it was expected by Annema et al. (1996) that PM_{10} concentrations would increase in a north-south direction over the Netherlands, with the concentration being markedly higher in winter than in summer. However, this turned out not to be the case. Almost no gradient is observed over the Netherlands and there are no marked differences in mean concentration between summer and winter. Also the concentration variations over the day are small. The morning and evening rush hours have only little effect on PM_{10} concentrations both in towns and rural areas. Average PM_{10} concentrations in towns have been found similar compared with nearby rural levels sug-

gesting only little impact of local emissions. The impact of local sources is found larger during calm weather (Annema et al., 1996).
The concentration of PM_{10} in the Netherlands is found heavily dependent upon wind speed, being considerably higher at low wind speed. This behaviour was found almost identical to that of CO. In busy streets almost all the CO is derived from vehicular traffic, and of the inner-city sources of PM_{10} (road traffic, small industries, space heating), the contribution from traffic was found dominant (Annema et al., 1996). Based on the ratio between the measured increases in CO concentration and PM_{10} concentration it was estimated that during episodes PM_{10} in urban air are about 20 $\mu g.m^{-3}$ higher than the levels in nearby rural areas (Annema et al., 1996).
Episodes of enhanced photochemical activity occur during summer when the weather is sunny and warm. Ozone is produced by the reaction of hydrocarbons with oxides of nitrogen in the presence of sunlight. Also the concentrations of primary (i.e. carbonaceous) and secondary aerosols are increased. Because the relative humidity is high at the same time, the average size of secondary aerosols in the fine mode increases due to deliquescence. The most noticeable consequence of this is visibility reduction, the characteristic summer haze (Van der Meulen, 1986). Fine particulate matter concentrations are generally higher during winter air pollution episodes than during summer episodes. In winter episodes, the aerosol is preliminary of local/regional origin, that is, mostly an urban/industrial problem, in which, in addition to space heating, vehicular traffic plays an increasingly important role. In urban/industrial areas in particular, the episode aerosol no longer has such a pronounced PM_{10} character as during the large-scale air pollution episodes in the 1970s and 1980s (Annema et al., 1996).

3. Natural emissions: sea salt

3.1 Process description

Sea salt aerosol is produced at the ocean surface by the bursting of air bubbles resulting from entrainment of air induced by wind stress. On bursting, these bubbles produce film and jet drops (Andreas et al., 1995). These bubbles are most concentrated in whitecaps associated with the breaking of waves which commences at approximately 3-4 $m.s^{-1}$. Depending on its size, each bubble can generate as much as 10 jet drops with a typical size of 1-2 μm radius (although extending to sizes greater than 10 μm), and up to several hundred film drops in the sub-micron range (Woolf et al., 1987). At wind speeds in excess of 7-11 $m.s^{-1}$, the tearing of wave crests results in the injection into the marine boundary layer of ultra-large (extending to sizes greater than 100 μm) spume sea salt particles (Smith et al., 1989; Wu, 1993). Sea salt number and concentrations are thus strongly dependent on wind speed. Monahan et al. (1983; 1986) developed a parameterisation for estimating sea salt emission in relation to particle size taken into account the impact of wind speed.

3.2 Contribution to atmospheric aerosol

Diederen and Guicherit (1981) have used a multiple linear regression method to estimate the contribution of the most important primary sources to the composition of aerosol samples taken in 1977 in Delft, Sloegebied, IJmuiden, Haamstede, Ugchelen and Schiermonnikoog. Results are presented in Table 1. Clear differences were found between the different sample locations. In comparison to urban areas, the total contribution of primary sources to aerosol was found smaller in rural areas. On the basis of aerosol analysis during two years in Delft it was concluded that the remaining part of the aerosol is produced due to chemical reactions in the atmosphere (secondary aerosol formation). Secondary aerosol thus contributes about 40% to the aerosol in urbanised areas and 80-90% to the aerosol in rural areas. It must be stated here that Diederen and Guicherit (1981) only used a small number of samples for their analysis. At each location two samples were taken during westerly winds and two samples during easterly winds. Due to prevailing western winds in the Netherlands, the contribution of sea salt to the aerosol as estimated by Diederen and Guicherit will be an underestimate of the yearly average contribution. No information is available on the wind speeds during aerosol samplings performed by Diederen and Guicherit (1981).

Table 1 *Estimated contribution of primary sources to aerosol at six different locations in the Netherlands in 1977 (weight %).*

Source category	Delft	Sloe	IJmuiden	Haamstede	Ugchelen	Schiermonnikoog
Paint	0.1	0.2	0.5	0.1	0.1	0.1
Traffic / transport	2.2	0.7	1.1	0.7	1.2	0.2
Soil dust	16.4	30.6	6.3	2.2	4.9	1.1
Steal industry	1.6	1.3	3.3	0.9	0.4	0.1
Cement industry	3.3	17.4	6.7	0.9	1.1	0.0
Sea salt	1.8	5.7	1.7	6.0	0.8	2.4
Coal combustion	0.0	0.0	0.0	0.7	0.0	0.6
Oil combustion	1.0	2.9	3.4	1.5	0.6	1.3
Total primary sources	26.4	58.8	53.0	13.0	9.1	5.8

Data from Diederen and Guicherit (1981) suggest a strong exponential gradient exists in the Netherlands for the contribution of sea salt to total suspended matter. Near the coast this contribution approximated 6% whereas more inland this contribution is only a few percent or even less. The mean annual contribution of sea salt to aerosol will strongly depend on the occurrence of strong westerly winds (gales) when very large amounts of sea salt aerosols are suspended into the atmosphere. This is illustrated in Figure 2 showing mean concentrations of Na^+ in precipitation measured at De Bilt during the period 1992-1994. Concentrations are found relatively large in the winter season. Na^+ in precipitation in the Netherlands is generally assumed to be completely due to scavenging of sea salt particles (e.g. Draaijers et al., 1996).

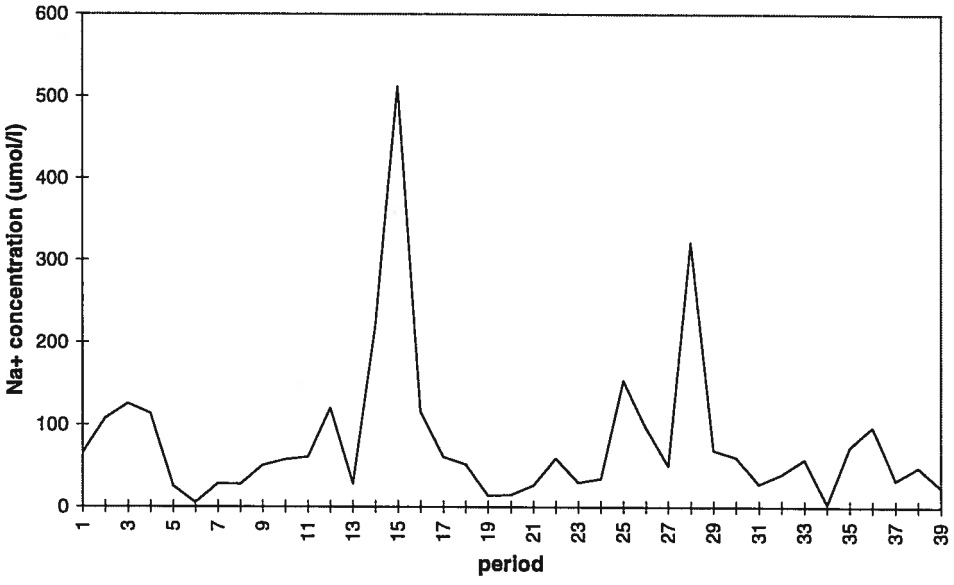

Figure 2 *Mean Na^+ concentration at De Bilt for 39 four-weekly periods during 1992-1994 (data from the National Precipitation Monitoring Network).*

From measured PM_{10} and the percentage secondary aerosol at several monitoring stations in the LML, Janssen and Van der Wal (1996) conclude that near the Dutch coast sea salt contributes more than 10% to the PM_{10} concentrations. In the eastern part of the Netherlands this percentage was assumed smaller than 5%. Janssen and Van der Wal (1996) note, however, that PM_{10} concentrations are estimated using a β-dust monitor whereas secondary aerosol concentrations are analysed from filters of low-volume samplers. Differences in sampling efficiency between the two methods may influence the calculated ratio between primary and secondary aerosol (now assumed 0.6 for 'background' stations) to a large extent and, in turn, the calculated contribution of sea salt particles to PM_{10} concentrations.

An indicator for estimating the contribution

Na^+ and part of the Mg^{2+}, Ca^{2+}, K^+, Cl^- and SO_4^{2-} present in precipitation samples is the result of the scavenging of sea salt particles. The part of Mg^{2+}, Ca^{2+}, K^+, Cl^- and SO_4^{2-} of sea salt origin can be estimated by using the fixed ratio found in sea water between the concentration of Mg^{2+}, Ca^{2+}, K^+, Cl^- and SO_4^{2-} on the one hand and Na^+ on the other. On an equivalent basis these ratios equal 0.227 for Mg^{2+}/Na^+, 0.044 for Ca^{2+}/Na^+, 0.021 for K^+/Na^+, 1.164 for Cl^-/Na^+ and 0.12 for SO_4^{2-}/Na^+ (Weast, 1975). The ratio of the sea salt part of the concentrations of Na^+, Mg^{2+}, Ca^{2+}, K^+, Cl^- and SO_4^{2-} with the total ionic load in precipitation samples can be regarded as an indicator for the contribution of sea salt particles to the total suspended matter in the atmosphere.

Part of the SO_4^{2-}, NO_3^- and NH_4^+ present in the precipitation samples will be the result of in-cloud and below-cloud scavenging of gases and is thus not the result of scavenging of aerosols. It has been estimated that in-cloud scavenging of aerosols containing SO_4^{2-} and NO_3^- is responsible for most of the SO_4^{2-} and NO_3^- in precipitation (Garland, 1978; Fowler, 1984). The other sources are scavenging of SO_2 and HNO_3. For NH_4^+ in precipitation, usually in-cloud scavenging of NH_4^+ containing particles is the major removal route but under Dutch conditions below-cloud scavenging of NH_3 may also be important (Asman, 1985). For the calculation of above-mentioned ratio it has been assumed that 70% of all the SO_4^{2-} and NO_3^- and 50% of all the NH_4^+ present in precipitation samples in the Netherlands is the result scavenging of aerosols. The indicator (I) thus equals:

$$I = \frac{\{[Na^+] + [Mg^{2+}]_{ss} + [Ca^{2+}]_{ss} + [K^+]_{ss} + [Cl^-]_{ss} + [0.7 * SO_4^{2-}]_{ss}\} * 100\%}{\{[Na^+] + [Mg^{2+}] + [Ca^{2+}] + [K^+] + [Cl^-] + [0.7 * SO_4^{2-}] + [0.7 * NO_3^-] + [0.5 * NH_4^+]\}} \qquad (1)$$

in which *ss* denotes the sea salt contribution. Other ions like F^- and other inorganic micro-components also contribute to the total ionic load of the precipitation sample but their concentrations are usually found very small in comparison to the other ions. Concentrations in equation (1) need to be expressed in equivalents (moles per charge).

Figure 3 shows indicator values for different monitoring stations in the Netherlands. According to the indicator the contribution of sea salt to the total suspended

matter in the Dutch atmosphere amounts 35-75%. Using the ratio of the sea salt part of the concentrations of Na^+, Mg^{2+}, Ca^{2+}, K^+, Cl^- and SO_4^{2-} with the total ionic load *in precipitation* samples as a measure for the contribution of sea salt to the total suspended matter *in the atmosphere* it is assumed that the scavenging efficiency of all aerosols is the same. This assumption is not valid. The scavenging of aerosols has been found to depend on the particle size (Kane et al., 1994; Jaffrezo and Colin, 1988; Buat-Menard and Duce, 1986) and to a lesser extent on their solubility (Slinn et al., 1978; Jaffrezo and Colin, 1988). Sea salt aerosols usually have relative large mass median diameters through which their scavenging will be relatively more efficient in comparison to e.g. acidifying aerosols. This means that the contribution of sea salt to the total suspended matter in the atmosphere will be overestimated by using equation (1). From data of Kane et al. (1994) it can be estimated that the scavenging efficiency of coarse aerosols is usually 2 to 3 times larger in comparison to that of fine aerosols. For this reason in Figure 2 also indicator values are presented which are calculated according to equation (1) but now by dividing $[Na^+]$, $[Mg^{2+}]$, $[Ca^{2+}]$, $[K^+]$, $[Cl^-]$ $[Mg^{2+}]_{ss}$, $[Ca^{2+}]_{ss}$, $[K^+]_{ss}$ and $[Cl^-]_{ss}$ by 2, respectively 3. According to this procedure, the contribution of sea salt to the total suspended matter in the Dutch atmosphere approximates 25-60% and 20-50% with the largest contributions at monitoring sites situated near the coast. Similar percentages are reported by Lowe et al. (1996) for a site at the Northwest coast of Scotland. In case of maritime air masses 60% of the accumulation mode aerosol composition was found to consist of NaCl. For continental air masses this was 15%. For the larger size ranges these percentages would have been larger as it may be expected that the bulk of the sea salt particle mass will occur in the coarse mode (Davidson and Wu, 1985). Pio et al. (1996) reported for a site at the west coast of Portugal sea spray to be by far the major contributor to the coarse fraction of the atmospheric aerosol with an average of 88% of the suspended coarse aerosol mass. In the fine aerosol mass sea spray contributed 14% of the mass loading.

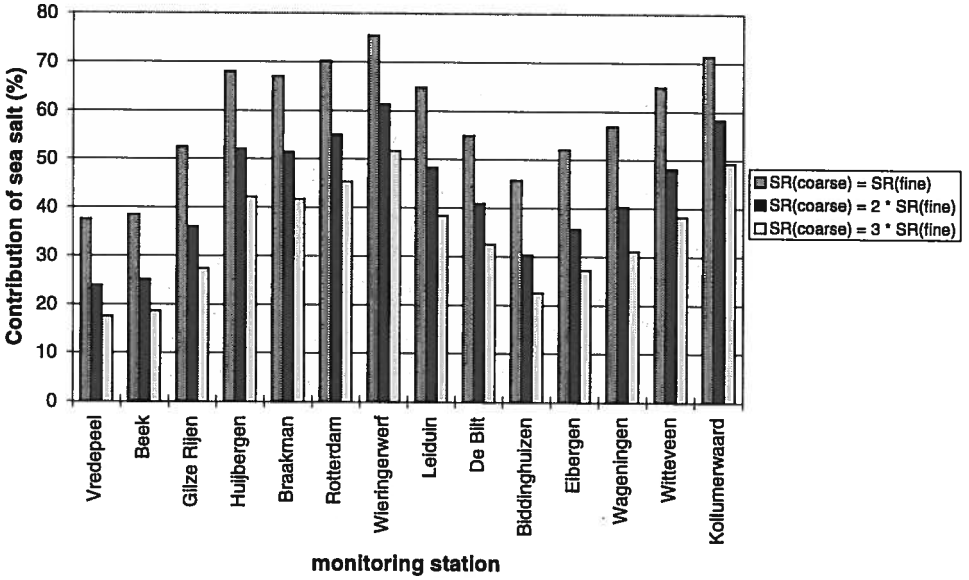

Figure 3 *Indicator values as estimated according to equation (1) on the basis of annual mean precipitation concentrations measured at different monitoring stations in the Netherlands in 1993 (in %). Indicator values assuming equal scavenging ratios for coarse particles (SR(coarse)) and fine particles (SR(fine)) are presented as well as indicator values assuming SR(coarse) being 2 or 3 times larger than SR(fine).*

Precipitation concentrations will reflect atmospheric concentrations of the entire atmospheric column from cloud top to surface level and thus will reflect the large scale 'background' situation. A strong correlation with surface level air concentrations will only be present in well-mixed conditions at sufficient distance from sources. Close to sources surface level air concentrations usually will be considerably higher. As a consequence, near the coast the contribution of sea salt to the total suspended matter *in the surface level* air will be underestimated by using the method described above.

The indicator gives information on the contribution of sea salt particles to the total suspended matter in the atmosphere. Sea salt particles usually have relatively large diameters, i.e. 1-20 μm. In a review of sizes of particulate trace elements in the atmosphere, Milford and Davidson (1985) end up with a median mass diameter of 3.8 μm for Na^+ containing particles with a geometric standard deviation of 4.0. The indicator thus will overestimate the contribution of sea salt particles to the $PM_{2.5}$ and especially $PM_{0.1}$ fraction of atmospheric aerosol. For PM_{10} the indicator probably gives quit reliable results.

Finally it must be mentioned that the indicator gives the contribution of sea salt to the soluble fraction of particulate matter present in the atmosphere. Part of the particulate matter (e.g. silicates, oxides and C-containing material) will be insoluble which is not reflected in the indicator.

3.3 Composition of sea salt aerosols

Sea salt aerosols usually consist of combinations of NaCl, mixed-cation (Na, Mg, K, Ca)sulphates, $NaNO_3$, Na_2SO_4 (with minor Mg, K and Ca), $(NH_4)_2SO_4$, and terrestrial minerals (mostly silicates and anhydrite) (Posfai et al., 1995). Sea water contains sea salt to about 3.5% by weight, of which 85% is NaCl and it can safely be assumed that the sea-salt content of film and jet drops is similar. Particles of sea salt origin can be distinguished on the basis of their sea-water-like Mg/Na, K/Na and Ca/Na ratios. The magnitude of the Cl loss from these particles and the amount of sulphate and nitrate that formed on them define distinct compositional groups. Such groups represent different stages during sea-salt conversion, and reflect the effects of reactions between NaCl and atmospheric S- and N-bearing compounds. Particles formed under clean oceanic conditions have rather uniform compositions. Their excess sulphate and nitrate probably will be formed through the oxidation of SO_2 by O_3 in the sea salt aerosol water and by direct reactions between NO_x (NO_2 and N_2O_5) and NaCl. In unperturbed marine environments, the source of this sulphate is dimithyl sulphide (DMS) from the ocean (Tarrasón et al., 1995). A large fraction of DMS is converted to sulphuric acid, which partly settles onto the sea-salt aerosol. In more polluted areas HNO_3 may be formed during the day as a result of photochemical reactions which, in turn, can volatilize Cl from sea salt and convert NaCl into $NaNO_3$. Long-range transport of continental air masses may also result in many crustal and individual Na_2SO_4 particles. A probable source of this sulphate is anthropogenic SO_2 (Posfai et al., 1995).
Lowe et al. (1996) found for a site at the Northwest coast of Scotland a significant correlation between sea salt mass and organic carbon mass, suggesting that the bulk of the organic material, rather than being sourced from anthropogenic processes as usually assumed, may have originated from the ocean. The upper region of the ocean is usually supersaturated with organic materials, providing a flux of gaseous organic species to the atmosphere where they may be oxidised to less volatile species which condense on to the aerosol phase (Warneck, 1988). The organic carbon encountered by Lowe et al. (1996) had a spectral shape almost identical to that of sea salt, strongly suggesting, the organic material entered the atmosphere as part of the sea spray droplet production process.
The composition of sea salt aerosols will change in relation to the distance to the coast due to the impact of continental air masses. For example, HNO_3 and H_2SO_4 present in continental air masses may volatilize Cl from sea salt and convert NaCl into $NaNO_3$ and Na_2SO_4, respectively, simultaneously releasing HCl(g) to the atmosphere (Mamane and Gottlieb, 1992). NO_2 and N_2O_5 have been found to react with sea salt particles forming $NaNO_3$ and releasing NOCl(g) (Karlsson and Ljungström, 1995; Finlaysson-Pitts et al., 1989). If Cl loss is suspected, one can compare the Na/Cl ratio, which is a good indicator of the Cl depletion, since Na is a conservative element. The Na/Cl ratio for the bulk deposition fluxes measured in the Netherlands is very close to the sea-water ratio (0.88) (Figure 4), with no clear gradient over the Netherlands thus leading to the conclusion that Cl loss to the gas phase is insignificant.

There are indications that there is a relationship between the composition of sea salt aerosols and their radius. In clean marine air, particles with diameters smaller than 0.5 μm are found to be composed predominantly of non-sea salt sulphate. There is considerable evidence that this nss-sulphate is formed by gas-to-particle conversion of the oxidation products of organosulphur gases (principally DMS) emitted by the ocean. The principle gas-to-particle conversion mechanisms are particle formation by homogeneous nucleation of low-volatility gas-phase reaction products, condensation of these products on existing particles, and SO_2-to-sulphate conversion in cloud droplets. In clean marine air, particles with diameters larger than 0.5 μm are composed primarily of sea salt (Fitzgerald, 1991).

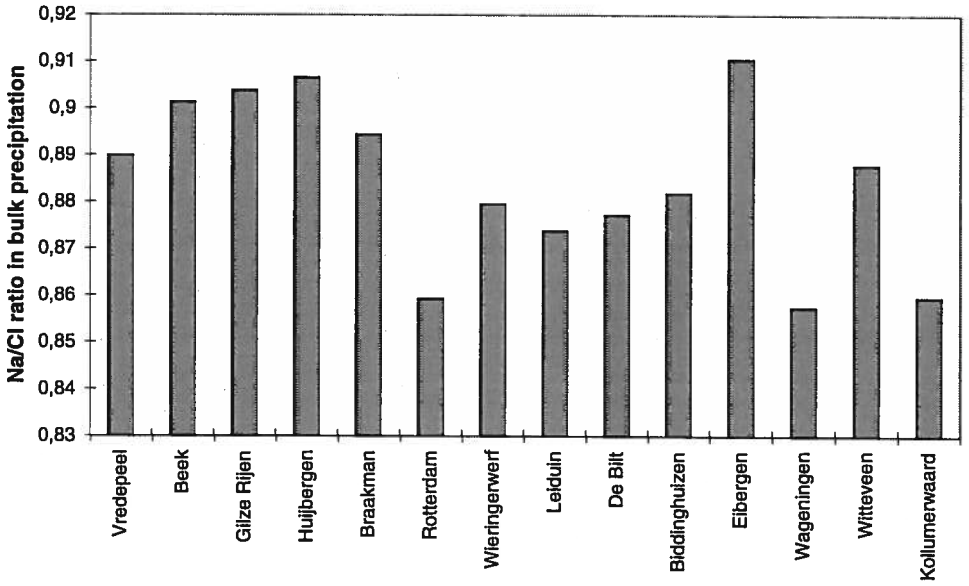

Figure 4 *Annual mean ratio between Na^+ and Cl^- concentration in precipitation measured at different monitoring stations in the Netherlands in 1993.*

4. Natural emissions: soil dust

4.1 Process description

Soils are formed by the weathering of crustal material of the earth. Rocks, stones, and pebbles slowly disintegrate through the action of water, chemically by the leaching of soluble elements, and mechanically by the freeze-thaw cycle of water entering into pores and cracks. In this way, igneous rocks are transformed into clay-minerals, carbonates, and quartz grains (sand). Soil dust emission can take place as a result of *i)* wind erosion, *ii)* travel on unpaved roads, *iii)* agricultural tillage practices, *iv)* activity at construction sites, *v)* surface mining and *vi)* mineral tailings piles.

4.2 Parameters important for soil dust emissions

Soil dust production is connected to aeolian transport of soil grains. From a physical point of view, the particles motion initiated by wind is controlled by the forces acting on them. For a particle at rest, these forces are the weight, the interparticle cohesion forces and the wind shear stress on the surface. The first ones are size dependent; the last one depends on the transfer of the wind energy to the erodible surface which is controlled by the presence of roughness elements on the surface. All together determine the minimum threshold friction velocity (defined as the square root of the ratio of surface stress to air density), u_{*t}, required to initiate particle motion (Marticorena and Bergametti (1996).
Once the particle is in motion, its path depends on the budget between its weight acting downward and the opposite aerodynamic drag. The vertical extent of these trajectories defines three major types of grain motion, generally classified in relation with the particle size (Bagnold, 1941). Particles smaller than 60 μm are small enough to be transported upward by turbulent eddies, sometimes very far from the sources (Swap et al., 1992). This movement is called 'suspension'. The soil grains in the range of 60-2000 μm are able to be lifted from the surface at a height of some tenths of centimetres but as the drag is not sufficient to exceed the weight, they are carried downwind back to the surface. Such trajectories define a motion called 'saltation'. Particles too large or too heavy to be lifted from the surface (>2000 μm) roll and creep along the surface in a motion called 'creeping'.
The fine suspended particles constitute the dust flux, which is referred as the vertical flux. Since dust production experiments can hardly be performed in wind tunnels, the physical processes of dust emission are not yet well identified and described. For this reason, the vertical flux is generally estimated from the horizontal flux and so involves equations having roughly the same form. In such expressions the term characterizing the source specifity is the threshold friction velocity. It has been experimentally shown that both the eroded mass flux and the size distribution of the flux depend on the size distribution of the initial particle bed (Sörensen,

1985; Li and Marz, 1994). Nickling (1988) has indicated that the threshold friction velocity of a natural sediment can not be represented by a single value. Erosion thresholds observed for both natural and wind tunnel experiments on rough surfaces have been found significantly higher than those observed on smooth surfaces (Musick and Gillette, 1990; Gillette et al., 1982).
Experiments of Gillette (1988) have indicated that precipitation in excess of 5 cm destroys aggregate structures in soil and thus lowers the threshold friction velocity. Drying may cause crusts to form on all but sandy textures, thereby increasing the threshold friction velocity. Occurrence of high winds immediately after a soaking rainfall led to destruction of soil aggregates and had the effect of lowering the threshold friction velocity before the soil becomes crusted. Empirical effects of vegetation, live and dead, have been specified by increasing the threshold friction velocity for amounts of vegetation in excess of threshold amounts as a function of plant type (Gillette and Passi, 1988). Prolonged lack of moisture results not only in dry soils but also in lack of vegetation that would have been protective of the surface. For agricultural soils, lack of soil moisture coupled with cultivation leads to weakening of soil aggregates, loss of cover, and lowering of threshold velocities for wind erosion (Gillette, 1988).
Dust emissions associated with travel on unpaved roads have been found to depend on road surface moisture and silt content, as well as on vehicle speed (Cowherd et al., 1974). The amount of dust produced per acre of agricultural tilling depends upon many factors, including soil silt content, soil moisture, and tilling implement speed (Cowherd et al., 1974). Total annual emissions per acre will also depend upon the number of times the land is tilled each year. Dust emissions at construction sites are generated by such activities as land clearing, blasting, ground excavation, cut and fill operations, and travel on access roads and on the site. Emissions will vary according to the level of activity and weather conditions. Dust emissions due to surface mining are associated with the removal of overburden, drilling, and blasting, ore extraction and loading, and traffic on access roads. In the mineral industries, two operations, overburden removal and milling of ores, produce huge volumes of wastes. The milling of ores results in solid waste which leaves the mill as a slurry. The slurry is channelled to a tailings pond, where solids are allowed to settle out of suspension. After the separation of water and solids, a dried pile of mil tailings remains. These piles are subject to wind erosion.

4.3 Current models for estimating soil dust emissions

A physically based model for estimating soil dust emissions resulting from wind erosion is developed by Gillette and Passi (1988). This model has been applied for dust emission estimation in the USA (Gillette et al., 1988) and part of Asia (Chang et al., 1996). The general form of the model is given in an equation for expected dust emission due to wind erosion, E_{we}, for a given soil, for a time duration δT, having area A:

$$E_{we} = k\, \delta T\, A \int_{u*t}^{\infty} G(u*)\, p(u*)\, du*$$

where $G(u_*)$ the vertical dust flux as function of friction velocity and $p(u_*)$ the probability density function of friction velocity. The constant k is calibrated to be $1.4\pm0.1 \times 10^{-15}$ (Gillette and Passi, 1988). The most sensitive parameter expressing the effect of soils is the threshold friction velocity u_{*t}. Gillette (1988) has determined u_{*t} for different land uses and vegetation covers as a function of soil texture and precipitation during the preceding month. Threshold friction velocities were assigned to nine different soil-type families. The function of total mass flux (G) is theoretically derived by Owen (1987) and confirmed by experiments performed in wind eroding fields by Gillette (1981) and Gillette and Passi (1988). The form of the function G is asymptotically proportional to the fourth power of the friction velocity. The probability density function used for wind speed is the Weibull distribution. For wind speeds greater than threshold for wind erosion, the atmospheric stratification is close to neutral. The expected wind erosion is therefore calculated by converting average wind speed to friction velocity using the wind drag coefficient c_d. Values for c_d have been determined from field studies by Gillette (1981) and Gillette and Passi (1988).

The approach of Gillette (1988) is valuable when a large number of reliable measurements of threshold friction velocities is available. However, it can not be used as an interpretative tool of the mechanisms of dust production and their evolution. From this point of view, an explicit parametrization of the threshold friction velocity in relation with the surface characteristics of the sources is needed. Marticorena and Bergametti (1995) developed a parametrization of u_{*t} in relation to the size of the erodible particles and the effect of the roughness elements, which controls the wind shear stress on the surface. These two parameters are included in a formulation of the threshold wind friction velocity by adapting a size-dependent parametrization proposed by Iversen and White (1982) and by applying to the rough erodible surfaces a drag partition scheme derived from Arya (1975). This parametrization of the threshold friction velocity has been included in a horizontal flux equation proposed by White (1979). This allows to attribute a specific production rate to each soil size range for each type of surface. The dust flux F is then considered as a fraction of the total horizontal flux G, the value of the ratio F/G being imposed by the soil clay content. In the model of Marticorena and Bergametti (1995) the computed mass fluxes thus depend on the soil size distribution, the roughness lengths, and the wind friction velocity. The different steps in their calculation scheme have been independently validated by comparison with relevant experimental data.

Some factors influencing wind erosion threshold such as moisture, salts, or organic matter content, are not explicitly parameterized by Marticorena and Bergametti (1995). Although their effects are poorly documented, they mainly affect the soil cohesion (Nickling and Ecclestone, 1981; Gillette et al., 1982; Breuniger et al., 1989), and thus the size distribution of the in-place aggregates. Moreover, in their approach the sources are considered as continuous, no parametrization of the soil evolution being performed. Thus, the kinetics of the soil depletion is ignored in the model. Consequently, specific emissions from supply-limited sources cannot be correctly simulated. The major weakness of the dust emission schemes of Marti-

corena and Bergametti (1995) and Gillette et al. (1992) concerns the estimation of the vertical flux, which is considered a ratio of the horizontal flux depending on the soil clay content. It does not account for the physical processes really involved but up to know these processes are not well identified and quantified.
Another approach has been followed by Tegen and Fung (1994) when estimating global soil dust emissions due to wind erosion. They assume surface wind speed, soil water content, and vegetation cover as the crucial factors for the determination of the source strength of mineral aerosols. Disturbance of the soil surface by land use is also considered important but neglected in their model. Wind erosion is assumed only to occur on dry soils when the soil matric potential is higher than 10^4 $J.kg^{-1}$, when the soil starts to be hygroscopic (Scheffer and Schachtschabel, 1992). The soil matric potential is dependent on soil moisture and texture and is calculated using the model employed by Bouwman et al. (1993). There, water content at field capacity (SSC) and monthly mean water content (SWC) in the upper 30 cm are determined from monthly climatological rain data (Shea, 1986), soil types, and soil texture (Zobler, 1986). Scheffer and Schachtschabel (1992) give typical curves of the dependency of soil matric potential on soil moisture for sand, silt and clay. From these curves Tegen and Fung (1994) determined the saturation ratio SWC/SSC necessary to obtain a matric potential of 10^4 $J.kg^{-1}$ to be 20% of the SSC for sand, 25% for silt, and 50% for clay. With these values the areas of dry soil from the saturation ratio for each month were determined. Uplift of dust can only occur where the vegetation is low and sparse. Tegen and Fung (1994) use the vegetation cover data set of Matthews (1983) to exclude regions with tall vegetation (e.g. forests) as possible dust sources and assumed wind erosion to be possible in desert, grassland, and shrub land regions. Snow-covered regions were excluded as possible dust sources using monthly mean spatial snow coverage maps (Rossow et al., 1991). The amount of dust emission from a surface due to wind erosion, E_{we}, was calculated according to (Gillette, 1978):

$$E_{we} = C\,(u - u_{*t})\,u^2$$

where u is the surface wind speed, u_{*t} the threshold velocity and C a constant to be determined a posteriori. The threshold surface wind velocity at 10m height was chosen to be 6.5 $m.s^{-1}$, corresponding to Kalma et al. (1988). Tegen and Fung (1994) used ECMWF wind products (10m surface winds) with a resolution of 1.125°x1.125° and 6 hourly time steps.
Soil dust emissions as a result of travel on unpaved roads, agricultural tillage, construction site activity, surface mining and mineral tailings piles have been estimated for the United States by Evans and Cooper (1980). For estimating unpaved road dust emissions they used transportation statistics (a.o. amount of vehicle miles on unpaved roads) together with emission factors of Cowherd et al. (1974). The following approximation was used to estimate soil dust emissions as a result of traffic on unpaved roads (E_{ur}):

$$E_{ur} = [0.65S_r(S/40)(d/365)][\Sigma\, u_{ij}M_{ij}]/200$$

in which S_r is the road surface silt content, S the vehicle speed, d the number of dry days, u_{ij} the unpaved road use factor i,j (vehicle-miles/miles), M_{ij} the unpaved mileage of road type i,j (mileage), i the road type index and j the rural, urban index. A mean vehicle speed of 40 mph was assumed. The amount of dust produced by agricultural tilling, E_{at}, in the United States was estimated from:

$$E_{at} = [1.1\ S_a(S_i/5.5)/(PE/50)^2](HT)/2000$$

where S_a is the agricultural soil silt content, S_i the implement speed, PE the Thornwaite's PE index, H the acreage of harvested cropland and T the number of tillings per year. Soil moisture is reflected in the Thornwaite's precipitation-evaporation (PE) index. Dust emissions as a result of activities at construction sites, E_{cs}, have been measured by Evans and Cooper (1980) using the following approximation:

$$E_{cs} = [D][(E)(M)]$$

where D is the duration of the activity, E the aerial extent of the activity and M the construction expenditure. Representative values were attached to different activities. Dust emissions associated with surface mining, E_{sm}, were estimated from:

$$E_{sm} = [2(0.5(d/d + c/c)]T_s/2000$$

where d is the number of dry days per year, c a climatic factor and T_s the amount of minerals surface mined. A rough emission factor of 2 lb/ton of production for all surface mining was used. Dust emissions as a result of wind erosion of mineral tailing piles (E_{tp}) were approximated from:

$$E_{tp} = (0.133\ C)(A)$$

where C is a climatic factor and A the area of the tailings piles.

4.4 Agricultural sources contributing to soil dust emissions

Agricultural sources contributing to soil dust emissions include *i)* wind erosion from croplands, *ii)* agricultural tillage practices such as ploughing, liming, planting, harvesting, and *iii)* factory farming resulting in resuspension of indoor-dust consisting of food and manure. Other dust emission sources associated with agriculture include burning of agricultural waste and biological mobilization of plant material such as pollen, weathered leaf cuticle and leaf hairs. It is expected that the amount of dust emission due to the latter two sources is relatively small in comparison to agricultural sources associated with wind erosion from croplands, agricultural tillage and factory farming.

The amount of dust produced by wind erosion of cropland depends upon soil type and moisture content, wind velocity, vegetative cover, and both field and surface geometry (Woodruff and Siddoway, 1965; Cowherd et al., 1974). Evans and Cooper (1980) estimated the wind erosion of agricultural fields in the United States from:

$$E = [0.025\ g\ (I_a, K', C/100, L', V')](H)$$

where g is a function defined by Woodruff and Siddoway, I_a a soil erodibility index, K' a surface roughness factor, C a climatic factor, L' a unsheltered field width factor, V' a vegetation cover factor and H the acreage of harvested cropland. The erodibility index was determined for eleven different soil types based on soil texture. Soil dust emissions from agricultural fields can also be estimated using models such as described in section 2.2.

The amount of dust produced per acre of agricultural tilling depends upon many factors, including soil silt content, soil moisture, and tilling implement speed (Cowherd et al., 1974). Total annual emissions per acre also depend upon the number of times the land is tilled each year (usually three times). Evans and Cooper (1980) have estimated the amount of dust produced by agricultural tilling in the United States from:

$$E = [1.1\ S_a(S_i/5.5)/(PE/50)^2](HT)/2000$$

where Sa is the agricultural soil silt content, Si the implement speed, PE the Thornwaite's PE index, H the acreage of harvested cropland and T the number of tillings per year. Soil moisture is reflected in the Thornwaite's precipitation-evaporation (PE) index.

Dust emissions as a result of factory farming depend on the ventilation rate in animal stocks and the type and the number of animals. Rough estimates for dust emissions due to chicken and pig farming in Europe were made by Berdowski et al. (1997) using measurement results of PM-10 concentrations (ref.) and ventilation rates (ref.) in different factories. Dust emission estimates for factory farming of other animals are not available.

4.5 Composition of soil dust aerosols

The chemical composition of soil dust will reflect the contributions of elements present in the earth's crust, provided other contributions from e.g. anthropogenic or volcanic sources are negligible. However, this premise does not hold for all constituents of the soil dust aerosol. Some trace components are considerably enriched compared with their crustal abundances. For this purpose, a so-called enrichment factor (EF(x)) has been defined (Rahn, 1976):

$$EF(x) = (x)/(Ref)_{aerosols} / (x)/Ref)_{source}$$

where x is the element under consideration and Ref a reference element. An appropriate choice of reference elements is required, as well as a tabulation of elemental compositions of source materials. Elements that are useful as reference elements for crustal material include silicon, aluminium, iron, and titanium (Chow et al., 1994; Warneck, 1986). All these elements are abundant in rocks. A determination of the contribution of the crustal component to atmospheric aerosol is difficult as a result of imprecise knowledge of the composition of material resulting

from the wind erosion of soils. Usually a surrogate composition is used. Possibilities include the average composition of crustal rocks, bulk soil, or the aerosol-size fraction of the soil. In most cases, globally averaged rock or soil is used. The elemental composition of the soil-derived fraction of aerosol, however, has been found to deviate appreciably from that of average crustal rock or average soil. The relative abundances of the major elements have been found to differ individually by factors of about three (Warneck, 1986). The EF(x) values should therefore only be regarded as order of magnitude estimates of crustal sources. Values near unity suggest crustal sources as the predominant source of element x (Milford and Davidson, 1985).According to Rahn (1976) EF(x) values larger than 5 may be considered to be evidence of other than crustal sources.

The enrichment factor express the fraction of elemental mass for particles that are suspended in air to the fraction of elemental mass for all soil mass particles. The fraction of elements in suspended particles, however, will not be the same as the elemental fraction in the whole soil because the greatest part of the mass for suspended particles will be for sizes < 10 μm. It is known that the composition of soil varies as a function of size. Sand particles range in size from 50 μm to 2 mm. The dominant composition of sand is quartz, which is in general chemically inactive. Alkaline mass, for example, may be assumed completely associated with the silt and clay fractions of the soil (this is, particles < 50 μm). It follows that a simple estimation of the enrichment factor would be the inverse of the fraction of silt and clay mass to the whole soil mass. since, by definition, silt plus clay fractions are equal to 1 - sand fraction, the enrichment factor computed for the soil texture is simply $(1\text{-}s)^{-1}$, where s is the sand fraction of the soil mass. For example, for a soil having 10% of its mass in silt and clay, s would equal 0.9 and an enrichment factor of 10 would be multiplied by the whole soil alkaline fraction to give the fraction of alkaline material for the silt and clay (Gillette et al., 1992).

5. Anthropogenic emissions: tyre wear and other traffic dust

5.1 Process description

In the literature of air pollution sciences no sufficient description of the process of tyre wear is present. Cadle (1978) describes an experiment where a tyre was placed in an experimental chamber. From the results of the experiments he concludes that most wear losses are the result of mechanical processes. These mechanical processes usually produce rather large particles (diameter >7 μm). A minor part of the emissions are attributed to processes with a thermomechanical basis. Driving force of these processes is the heat that is produced by friction forces. As a result of these processes a small quantity of gases is released and small particles (diameter
<0.4 μm) are formed. These small particles only account for a minor part of the total mass emission but probably account for the greater part of numbers of particles emitted. Rauterberg-Wulff (1995) describes the typical shape of particles originating from mechanical tyre wear processes as cylindrical (cigar-like) with lengths between 2 and 35 μm and thicknesses between 1 and 7 μm.

5.2 Contribution to atmospheric aerosol

About total mass emission factors as a result of tyre wear the scarce literature is rather consistent. This is not surprising since it is easy to weigh mass loss and next to it travel distances of vehicles are known, so emission factors easily can be calculated. Emission factors for total mass loss lie between 20 and 30 mg.tyre kilometre^{-1} for passenger cars (Cadle 1978, Thomas 1992, Bayerisches Landesamt 1988). In the Dutch Emission Inventory 23 mg.tyre kilometre^{-1} is used for passenger cars, while 45 mg.tyre kilometre^{-1} is used for trucks and buses. This means that total mass emission of tyre wear is in the same order of magnitude as is tailpipe emissions. From the literature however no definitive conclusions can been drawn for which part of the total tyre wear loss can be assigned as PM_{10}. The US Environmental Protection Agency (US-EPA, 1995) uses a figure of 2‰ PM_{10} as a fraction of total tyre wear loss. In the Dutch Emission Inventory a figure of 5% PM_{10} of tyre wear is actually used. This means in the Dutch situation that PM_{10} tailpipe emission factors for cars with catalyst are practically equal to tyre wear PM_{10} emission factors. For trucks and buses tyre wear emission factors are only about 1% of tailpipe emission factors.
Rauterberg-Wulf (1995) attributed on the basis of on-site measurements in a tunnel about 25% of soot (with particle diameter range: 2 μm<ø<16 μm) to tyre wear; in a busy street in Berlin (about 40.000 vehicles.day^{-1}) this was about 15%. From the low concentration of soot in the intermediate particle size range (2 μm<ø<16 μm) measured in a near located background-station it was concluded that traffic was indeed the source of this particles. So the influence of tyre wear only is measurable

very near (meters) to busy roads. It might be concluded that the atmospheric residence time of the largest part of tyre wear probably is low compared to smaller particles. The article is concluded with the note that investigations are started in search for a specific chemical tyre dust tracer.
Presently there is no literature available to conclude whether the EPA (2‰ PM_{10}) or the Dutch estimate (5% PM_{10}) is more near the reality.

5.3 Composition of tyre wear particles

Because tyre wear particles mainly consist of small parts of the original tyres, the composition of the original tyres could be taken as a measure of composition. On the basis of chemical composition Rab (1992) concludes that tyre dust contains 60% organic carbon (OC) and 30% elemental carbon. This theoretical figure is very near to the figure measured by Israel (1994). He finds 53% OC and 34% EC. The theoretical figure is probably more representative because Israel took only one type of tyre.

5.4 Other sources of traffic dust

According to Rauterberg-Wulf (1995) a large part (50-75%) of the traffic contribution of particulate matter in the 2-12 μm range contains no organic carbon. This may probably be attributed to inorganic road dust that is brought in circulation by passing vehicles. In Norway NILU (Hagen, 1995a,b, 1996) has carried out series of investigations on PM_{10} and $PM_{2.5}$ in Oslo. They found a typical daily pattern with large concentrations during rush hours. The $PM_{2.5}$ showed a less pronounced pattern then PM_{10} (which is much steeper in traffic rush hours) indicating that traffic was probably the main source of the larger particles.
The influence of the cleaning of streets on PM_{10} concentrations by means of sweeping was investigated by Haugsbak (1995) in Trondheim in one street with two monitoring stations. One station stood in a part of the streets that was swept by means of a truck with rotating bristles while the other station stood near a part that was not swept. Systematically higher PM_{10}-concentrations were measured near the road part that was swept. In the report it was not made clear whether the station in the swept part of the street was influenced by the sweeping process itself.

6. Anthropogenic emissions: wood stoves and fire places

6.1 Process description

Particles emitted by residential wood combustion are formed by a combination of processes beginning with combustion, continuing as the flue gas cools in the flue pipe and ending when the flue gas is cooled and diluted with ambient air. Particles initially formed in combustion acquire mass and undergo a change in size distribution by coagulation and by organic vapor condensation processes. As the flue gas mixes with ambient air, both processes are essentially stopped and particulate composition becomes quasi-stable (Rau, 1989). The composition variability of residential wood smoke particulate therefore is determined by burn conditions, sample procedures, organic vapor adsorption on the filter sampling medium and the carbon analysis analytical method.

6.2 Contribution to atmospheric aerosol

Emission factors of residential wood combustion are between 5 and 15 g per kg of wood burned (Stern et al, 1992). CO-emission factors are between 50 and 125 g per kg of wood burned with an average around 80 $g.kg^{-1}$. Using an average PM-emission factor of 10 $g.kg^{-1}$ and 1 Million tons of wood burned the emission of PM in the Netherlands can be calculated to equal about 10 $kilotons.year^{-1}$. Emission factors actually used in the Dutch Emission Inventory are 9 g/kg for wood burning in stoves and 2.5 $g.kg^{-1}$ for wood burning in fire places. The latter emission factor seems to be rather low. This emission figure leads to the conclusion that residential wood burning potentially make up about 25% of the total anthropogenic PM emissions in the Netherlands. The contribution of residential wood burning to the atmospheric aerosol concentrations will be relatively large in urban areas in wintertime. Up to now no measurement studies are available to confirm this. From studies in the US, however, it is known that residential wood burning is a major source of inhalable particle carbon (up to 80%) in winter urban air (Hawthorne et al., 1992). Hawthorne found a very good correlation (r^2=0.9) of the concentration of guiacols (methoxylated phenols) and residential wood smoke carbon. He also found that PAH and other compounds are no good tracers for woodstove emissions because other sources (i.e. road traffic) contribute also significantly to these emissions.

6.3 Size and composition of residential wood smoke particles

The aerodynamic particle size of PM emitted by residential fire places depends on the burning temperature but essentially all particles emitted are found smaller then 10 μm. Particles emitted by hot burning are for the largest part (>50%) smaller

then 0.3 μm diameter (Rau, 1989). According to Nussbaumer (1989), more than 80% of the total particle mass of (industrial) woodburning is smaller then 0.5 μm. Under conditions of cool burning a bimodal particle size distribution appears next to the maximum of particles around 0.1 μm a second maximum near 1 μm diameter.

The mass of total carbon (TC) makes around 60% of the total particle mass. The organic carbon (OC) fraction is about 50% of the total particle mass while elemental carbon (EC) is only about 10% of the total particle mass (Rau, 1989). Tan (1992) showed that a simple catalytic combustor placed directly on a conventional woodstove can reduce the PAH-content of wood smoke particles by 80-95%.

7. Conclusions

From this literature study the following conclusions can be drawn:

1. Particulate matter (PM) concentrations in urban areas in the Netherlands are generally highest during winter air pollution episodes. The PM is preliminary of local/regional origin with space heating and vehicular traffic being the major sources.
2. PM concentrations in urban areas in the Netherlands strongly depend on wind speed and show temporal variations comparable to those of CO and to a lesser extent NO_2 concentrations.
3. Initial calculations indicate that sea salt particles may contribute approximately 20-50% to the total PM_{10} concentration in the Netherlands. The largest contribution is found near the coast. The contribution of sea salt particles to the $PM_{2.5}$ and especially to the $PM_{0.1}$ fraction of atmospheric aerosol is smaller. An physically-based emission model has been developed to estimate marine aerosol generation but has not been applied yet.
4. Sea salt particles in a clean marine atmosphere usually consist of a combination of NaCl and mixed-cation(Na, Mg, K, Ca)sulphates. The source of the sulphate is dimethyl sulphide emitted from the ocean. In the Netherlands, the composition of marine particles does not change significantly in relation to the distance to the coast.
5. Soil dust emission can take place as a result of wind erosion, travel on unpaved roads, agricultural tillage practices, activity at construction sites, surface mining and mineral tailing piles. For each type of soil dust emission physically-based or empirical models have been developed to quantify the emission fluxes. So far, no estimates are available on soil dust emissions in the Netherlands or Europe.
6. The chemical composition of atmospheric soil dust reflects the contribution of the elements present in the earth's crust. The composition of soil-derived material also depends on soil texture.
7. For passenger cars with catalyst, PM_{10} emissions due to tyre wear are about equal to PM_{10} emissions from vehicle tailpipes. For trucks and busses, tyre wear emissions are only about 1% of tailpipe emissions. Inorganic road dust that is brought in circulation by passing vehicles is another PM source associated with traffic.
8. The chemical composition of tyre wear particles reflects the composition of the original tyres (60% organic carbon, 30% elemental carbon).
9. Theoretical calculations indicate that residential wood burning can make up about 25% of the total anthropogenic PM emissions in the Netherlands. Up to now no measurement studies are available to confirm this.
10. About 50% of the total mass of residential wood smoke particles consist of organic carbon and about 10% of elemental carbon.

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9. Authentication

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