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Atmospheric dilution, in rush hour pollution?
a study of diurnal variations of particulate matter concentrations

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Atmospheric Dilution, in Rush Hour Pollution?
A Study of Diurnal Variations of Particulate Matter Concentrations

E.T. Slikboer

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Abstract

In recent decades, much research has been done towards the health hazard of particulate matter (PM) and other forms of air pollution. As a counteract the European Union has set year- and day limits for PM, but not an hour limit. Therefore almost no research is done towards it. During the night there is a maximum reduction of turbulence in the atmosphere and in combination with the morning rush hour, which causes an increase in emission, this can result in elevated levels of PM concentrations. Especially in the winter the reduced levels of turbulence can interfere with the increase of pollutants due to traffic, because then the sun rises late and as such the pollutants are dispersed minimally within the stably stratified boundary layer. In this thesis we have examined the hourly average PM$_{10}$ concentrations for five different stations throughout the Netherlands. Main reason was to investigate if a combination of reduced turbulence in the atmosphere in the morning and increased emissions due to morning rush hour lead to elevated levels of PM concentration. The stations were examined for December, February, April and June and classifications were made in wind speed, because turbulence is dependent on wind speed and it dilutes ground concentrations.

In this research I have found that in the winter the concentrations of particulate matter (PM$_{10}$) is driven by the magnitude of the (10m) wind speed. When wind speed is low, i.e. less than 3 m/s, PM concentrations could become more than twice as large as concentrations when wind speed is high, more than 6 m/s. Furthermore we observed similar behavior between the stations with respect to the seasonal variations in the diurnal cycle of the PM concentration. In December concentrations generally rise during the day and reach a maximum at 18:00. Then for low wind speeds the PM concentration levels are 10 % higher than the day limit, which is set at 50 µg/m$^3$ by the European Union. After sunset the average concentration would decrease during the night reaching a minimum at 06:00. In February the evening peak had shifted towards noon for low wind speed showing a broad daytime maximum. For average wind speed two equal peaks are observed, in the morning and evening.

Seasonal changes are further visible in April, where only a significant morning peak was observed around 10:00 and during midday concentrations would decrease significantly. This had further evolved in June, because dependency on the magnitude of the wind speed had fully diminished and concentrations would not fluctuate much during the day. These seasonal variations in the diurnal cycle of PM concentrations can be explained by the fact that solar radiation is minimum in the winter and as such only ambient shear wind can generate turbulence. This means that when wind speed is low an accumulation of pollutants within a small volume can occur. In the summer convection is dominant – due to an increased amount of radiation – and pollutants are dispersed during the day regardless of the magnitude of the wind.

In this research I developed a simple model to simulate the seasonality in the diurnal cycle of particulate matter concentrations. The only input used is data of the CBS (Centraal Bureau voor de Statistiek) about the mobility of the Dutch population. The mobility in an indication of the diurnal cycle of the traffic density. This was needed to model the emissions of a city, as the assumption was made that the PM emission is linear dependent with the traffic density. Seasonal changes were included in the shape of the mixing layer. Here a Gaussian shape was used where the width was determined by time of sunrise and sunset. For all the examined months the model simulated the diurnal cycle of the PM concentration similar to our observations. As such the conclusions can be made that the dynamics of PM concentrations are governed by rather basic mechanisms, namely convection during the day and the stably stratification of the boundary layer during the night, which were included in our model by the shape of the mixing layer.
Lastly we examined the probability per hour to violate the day limit of 50 $\mu g/m^3$, which has an allowed number of exceedances of 35. With present legislation the average amount of violations of the day limit in a year, based of 24 hour averages, is 31 in Eindhoven. Because a large group of commuters are active when hourly average PM concentrations are highest, they are consequently exposed to higher concentrations than the day average. Commuters that travel in Eindhoven at 10:00 and 18:00 are exposed to concentrations which are higher than the day limit in the evening 53 times and in the morning 86 times in a year. So with present regulations of the European Union commuters’ exposure with respect to PM is consequently underestimated.
# Contents

1 Introduction ................................................................. 3
   1.1 The Stations ......................................................... 5
   1.2 Boundary Layer Meteorology ....................................... 6
      1.2.1 Diurnal Atmospheric Cycle ................................... 6
      1.2.2 Seasonal Differences ......................................... 9
   1.3 Meteorological Differences between Stations .................... 10
      1.3.1 Average Radiation ............................................. 11
      1.3.2 Daily Temperature Cycle ..................................... 12
      1.3.3 Average Wind Speed .......................................... 13

2 Hourly Particulate Matter Concentrations ................................ 15
   2.1 Comparison Particulate Matter Concentration for Different Stations .... 16
      2.1.1 Station 136: Heerlen ......................................... 16
      2.1.2 Station 237: Eindhoven ....................................... 17
      2.1.3 Station 433: Vlaardingen .................................... 19
      2.1.4 Station 520: Amsterdam ...................................... 20
      2.1.5 Station 639: Utrecht ......................................... 21
   2.2 Conclusions and Discussion ......................................... 22

3 Modeling the Hourly Particulate Matter Concentration .................... 24
   3.1 Model Setup .......................................................... 24
   3.2 Limit Analysis ....................................................... 29
3.3 Results and Conclusions ............................................. 30
  3.3.1 December and February ...................................... 31
  3.3.2 April and June ................................................ 32

4 Day Limit Violation Probabilities 34
  4.1 Probability per Station ......................................... 34
    4.1.1 Station 136: Heerlen ...................................... 36
    4.1.2 Station 237: Eindhoven .................................... 37
    4.1.3 Station 433: Vlaardingen ................................. 38
    4.1.4 Station 520: Amsterdam ................................... 39
    4.1.5 Station 639: Utrecht ...................................... 40
  4.2 Conclusions and Discussion ................................... 41

5 Summary 42

A Number of data points 46

B Scatterplots of all five stations 47
  B.1 Station 136: Heerlen ........................................... 47
  B.2 Station 237: Eindhoven ......................................... 48
  B.3 Station 433: Vlaardingen ..................................... 49
  B.4 Station 520: Amsterdam ....................................... 50
  B.5 Station 639: Utrecht .......................................... 51
Chapter 1

Introduction

Since the dawn of mankind anthropogenic air pollution has existed. Whether it was from burning logs for heating in prehistoric times or from burning coal for steam power, pollutants were released into the Earth’s atmosphere. It is only since a few decades that an interest has began to grow into the origin and consequences of air pollution. Air pollution is a compilation of different substances like sulphur oxides, nitrogen oxides, carbon monoxide, organic compounds and particulates. Recently in 2006 the World Health Organization investigated the consequences of air pollution and concluded that it can cause a decrease of life expectancy of 3 months in slightly polluted areas up to 3 years in severe polluted regions\(^1\). In 2012 the OECD predicted that in 2050 particulate matter and ground-level ozone have become the top cause of environmentally related deaths worldwide\(^2\). As a counteract to these health hazards the European Union has set limits for pollutants. Here, we study pollution concentrations in surface air through a physical perspective. By focussing on the relatively ‘inert’ particulate matter (PM\(_{10}\), i.e. particulate matter with diameter smaller than 10 \(\mu\)m) complicating factors such as photochemical reactions during dusk, which causes significant changes for e.g. NO\(_2\), are avoided. The European Union has set the year limit of PM at 40 \(\mu\)g/m\(^3\), meaning that the yearly average needs to equal or smaller than this limit. The 24 hour average needs to satisfy the day limit of 50 \(\mu\)g/m\(^3\), which has an allowed number of exceedances per year of 35. There is, in contrast with for example SO\(_2\) and NO\(_2\), no hour limit for PM.

By order of the Dutch government the RIVM (Rijksinstituut voor Volksgezondheid en Milieu) measures and monitors the PM concentrations in the Netherlands. They also investigated the composition and origin of the particulate matter\(^3\) and keep records of the averages and number of exceedances in a year\(^4\). However because there is no hour limit, little research is done towards hourly particulate matter concentrations. This is mainly because for a single hourly PM concentration measurement the uncertainty is significant. That is because the particulate matter is measured using a dust monitor, which is influenced by (interfere) water vapor. Under specific atmospheric conditions however, hourly concentrations could show elevated levels while the 24 hour average may still be below the limit value for daily averages. The reason for this is the fact that during the night there is a great reduction of turbulence in the atmosphere. That is because there is no convection during the night and as such a stably stratified boundary layer forms. This way accumulation of pollutants in a small volume can occur. In the winter nights are longer, which means that there are many commuters
active when there is in the atmosphere a well developed stable boundary layer. The main subject of this thesis is to investigate if high emissions during rush hour, when atmospheric conditions cause pollutants to disperse minimally due to low levels of turbulence, cause PM concentration levels to rise. The probability of coinciding of the ‘sudden death’ (i.e. the low levels of turbulence due to the stably stratified boundary layer during the night) and the high emissions from rush hour is biggest in the winter, because during summer the sun rises earlier and due to convection a well developed mixing layer is formed at rush hour, this can be seen in figure 1.1. In this research hourly particulate matter concentrations of several stations in the Netherlands for the period from 2000 until 2012 will be examined to investigate the dependence with the atmospheric conditions in the boundary layer. This will be used to construct a simple conceptual model to comprehend and predict the concentrations levels at any hour. Also we will discuss the consequences of the hourly changes in concentration with respect to the daily routine of the people in the Netherlands.

Figure 1.1: Figures 1.1(a) and 1.1(b) show the reduction of turbulence present in respectively the summer and winter in combination with the amount of traffic emission.
1.1 The Stations

For this research five stations were initially chosen for analysis. These five stations – operated by the RIVM – were selected based on the characteristics of their location and on data availability. This means that the stations are located in an urban polluted area, because the assumptions is that the measurements of these stations are representative for the effects of local emissions. In a later stage an additional station was selected in order to analyse the dynamics of the ‘background concentration’, i.e. the concentration at a location representing the rural area. This point will be discussed in chapter 3. In figure 1.2 the locations of the different stations are visualized. Station 136 is located west of Heerlen and station 237 is located north west of Eindhoven. The third station 433 lies in Vlaardingen which is west of Rotterdam and north of the harbor of Rotterdam. Then stations 520 and 639 are located in the center of respectively Amsterdam and Utrecht. The last station is located in a small town with mostly agriculture, namely station 929 in Valthermond.

Table 1.1 shows the characteristics of the examined stations. As stated the first five stations are located in an urbanized area, and thus are specified as street/urban station, except station 520 in Amsterdam. It is specified as a city/urban location and this is because it is located in

![Figure 1.2: The locations of the different stations used in this research. Location A represents station 136 in Heerlen, location B station 237 in Eindhoven, location C station 433 in Vlaardingen, location D station 520 in Amsterdam, location E station 639 in Utrecht en lastly location F shows station 929 in Valthermond.](image-url)
a park. This means that the distance towards traffic emission sources is larger. Station 929 in Valthermond is a regional/rural station in a location which is characterized by agriculture. The type of a station, i.e. *regional, city or street*, is based on the amount of traffic that passes by the station in 24 hours. A *city station* is located in an urban area which is placed so that the number of passing vehicles within a radius of 35 m from the station is less than 2750 per 24 hours. For a *street station* this number is at least 10,000 per 24 hours. A *regional station* is defined by a location outside a built-up area and placed in order that the measurements are not being influenced by local sources[5].

<table>
<thead>
<tr>
<th>Station</th>
<th>City</th>
<th>Street</th>
<th>Geographical Coordinates</th>
<th>Type</th>
<th>Station Type</th>
<th>Characterisation of Zone</th>
</tr>
</thead>
<tbody>
<tr>
<td>136</td>
<td>Heerlen</td>
<td>Looierstraat</td>
<td>50°53’17”N 05°58’14”O</td>
<td>Street/U</td>
<td>Traffic</td>
<td>Residential</td>
</tr>
<tr>
<td>237</td>
<td>Eindhoven</td>
<td>Noordbrabantlaan</td>
<td>51°26’43”N 05°25’44”O</td>
<td>Street/U</td>
<td>Traffic</td>
<td>Residential</td>
</tr>
<tr>
<td>433</td>
<td>Vlaardingen</td>
<td>Floreslaan</td>
<td>51°54’41”N 04°19’37”O</td>
<td>Street/U</td>
<td>Traffic</td>
<td>Residential/Industrial</td>
</tr>
<tr>
<td>520</td>
<td>Amsterdam</td>
<td>Florapark</td>
<td>52°23’31”N 04°55’04”O</td>
<td>city/U</td>
<td>Background</td>
<td>Residential/Commercial</td>
</tr>
<tr>
<td>639</td>
<td>Utrecht</td>
<td>Erzeijstraat</td>
<td>52°04’08”N 05°07’16”O</td>
<td>Street/U</td>
<td>Traffic</td>
<td>Residential</td>
</tr>
<tr>
<td>929</td>
<td>Valthermond</td>
<td>Noorderdiep</td>
<td>52°52’36”N 06°55’52”O</td>
<td>regional/R</td>
<td>Background</td>
<td>Agricultural/Natural</td>
</tr>
</tbody>
</table>

### 1.2 Boundary Layer Meteorology

Anthropogenic air pollutants enter the atmosphere mainly within the *atmospheric boundary layer* (ABL). This layer, adjacent to the surface, is directly influenced by the presence of the earth and by the diurnal cycle in surface heating/cooling. The depth of this layer may vary between 1 - 10 km during daytime and between 50 - 300 m during nighttime, depending on the particular weather conditions. The dynamics within this layer strongly influence the variations in the observed PM ground concentrations. Other factors that influence ground concentrations are e.g. the emission source, natural removal processes and the height of the emittance of pollutants in the atmosphere. In the ABL turbulence facilitates mass transport, which causes pollutants that originate from ground emissions like traffic and factories to disperse within the atmosphere. Convection and wind shear are the two major drivers which generate turbulence during the day and as such the shape the ABL.

#### 1.2.1 Diurnal Atmospheric Cycle

When the sun rises short wave radiation starts to heat the earth’s surface, see figure 1.3(a). Warm air has a lower density than the ambient air and hence thermal convection will start. This is because when the air parcel’s density becomes lower than the surroundings the pressure
difference this creates will force the lighter air to move upwards, this is called buoyancy. This process insures heat transfer (free convection) in the atmosphere and causes mass transport during day. The solar radiation is absorbed partially by the earth’s surface as ground heat and the remainder will return in the atmosphere in the form of a sensible heat flux and latent heat flux, see figure 1.3(a). The turbulence created by convection will mix the air, thus creating a mixing layer where pollutants are more or less homogeneously distributed. The intensity of radiation from the sun increases till noon, see figure 1.3(b) and the height of the mixing layer grows accordingly. After maximum radiation intensity at noon the growth of the mixing layer height stagnates. Within the mixing layer convection continues to create turbulence. The mixing layer is separated from the free atmosphere (FA) by the entrainment zone (EZ). Because of this strongly stable layer the pollutants are trapped within the mixing layer.

The shortwave solar radiation stops at sunset. The earth’s surface will cool down by long wave radiative cooling. That is because the earth’s surface is warmer than the air above. This flux is approximately constant during the night, indicated with a gray area in figure 1.3(b). As such, there is no convection and thus the amount of shear turbulence in the mixing layer diminishes. A stable boundary layer (SBL) forms, where turbulence only can be generated by ambient wind shear. Figure 1.4 shows the evolution of the different layers within the ABL during day and night. The mixing layers evolves into a residual layer where the pollutants from the previous day reside. The entrainment zone becomes capping inversion (CI) after sunset. This is a non-turbulent separation layer. During the day the height of the mixing layer will be 0.5 - 2 km and during the night the height of the stable boundary layer is 50 - 300 m.
The typical shape of the horizontal wind profile changes during the diurnal cycle of the ABL. During the day convection causes wind speeds to be uniform across the mixed layer, with exception winds near the surface. There the wind speed is low due to friction. When convection diminishes at sunset, turbulent vortices disappear and horizontal winds are able to accelerate within the stable boundary layer. As such the magnitude of ambient wind is greater at night than during day. Thus turbulence can be generated during the night, but only through ambient wind instead of free convection. However buoyancy acts to counteract vertical mixing. This is because a stratification occurs due to the fact that cold air is heavier and stays near the earth surface.

**Figure 1.4:** The idealized diurnal cycle of the shape of the ABL in the summer\(^6\).

**Figure 1.5:** Figure 1.5(a) shows the wind profile during day in the ABL, consisting of the mixing layer, entrainment zone and free atmosphere. Figure 1.5(b) shows the wind profile during night, then the ABL consists of the stable boundary layer, residual layer, capping inversion and free atmosphere. The dashed line labeled G marks the geostrophic wind speed.
1.2.2 Seasonal Differences

As stated earlier, convection is the dominant process during the day. In the summer due to the angle of earth towards the sun, the intensity of the solar radiation is higher in the Netherlands. In general this means that the height of the mixing layer is much larger in the summer than in the winter, because convection is more dominant. In the winter radiation intensity is low and during the day turbulence is weak, a large stable boundary layer is present. Figure 1.4 gives a representation of the ABL during the summer and figure 1.6 shows the daily evolution in the winter. In the winter the mixing height which forms during the day is smaller than in the summer. Also the stable boundary layer is bigger, because sun rises later and sets earlier.

![Diagram of ABL during the summer](image)

*Figure 1.6: The idealized diurnal cycle of the shape of the ABL in the winter[^6].*

Furthermore in the summer days are much longer compared with the nights. The mixing layer evolves at 06:00 during sunrise and it is only after sunset at 21:30 that convection diminishes and the stable boundary layer will form. The atmosphere is mixed for a relatively long period. In the winter relatively weak convection will create turbulence between 08:30 and 16:30. This means that during a significant portion of the day pollutants will not be mixed by convection and only ambient wind can cause dispersion. In general, convection will disperse pollutants in the mixing layer, which is greatest in the summer because than the radiation intensity is maximal. Thus we expect, if ground emissions are similar throughout the year, PM concentrations to be highest in the winter, especially when wind speed is low because that is the only mechanism in the winter which can cause turbulence and dilute the concentration.
1.3 Meteorological Differences between Stations

In order to understand the dynamics of PM-concentrations at the various locations, first the typical meteorological conditions for each of the locations are examined. Reason for this is that the observed fluctuations in PM concentrations are strongly influenced by externally forced meteorological conditions in general. Unfortunately, meteorological quantities like wind speed or temperature are not reported by the RIVM stations directly. As such we have to rely on proxy-weather data which are available through the KNMI weather station network. The RIVM stations were combined with the closest KNMI stations in order to match the hourly PM concentrations with the meteorological quantities. The following KNMI stations were selected:

- Maastricht (15 km from RIVM station Heerlen)
- Eindhoven (5.0 km from RIVM station Eindhoven)
- Rotterdam (10.3 km from RIVM station Vlaardingen)
- Schiphol (12.1 km from RIVM station Amsterdam)
- De Bilt (5.6 km from RIVM station Utrecht)

In our analysis of both meteorological conditions and PM concentrations the months December, February, April and June were selected to investigate. Reason for this is that we can compare winter and summer and also investigate the transition between the seasons.
1.3.1 Average Radiation

Figure 1.7 shows the monthly average of the diurnal radiation cycle for the five different stations. In general differences between the various locations appear to be small. That is what was expected, because the stations are located close to each other on a global scale. As stated radiation causes convection which generates turbulence. At noon the amount of radiation differs significantly between the summer (550 W/m²) and the winter (110 W/m²). As such in the summer ground emissions of PM are dispersed upwards quicker than in the winter.

Figure 1.7: Figures 1.7(a), 1.7(b), 1.7(c) and 1.7(d) show the monthly average of the diurnal radiation cycle of all five stations.
1.3.2 Daily Temperature Cycle

Figure 1.8 shows the daily temperature cycle for the five different stations. In contrast with the radiation the temperature differs between the stations. In de winter Heerlen and Eindhoven are the coldest stations while in the summer they are the hottest. This was expected, because they are the furthest inland and thus the other cities are influenced more by the large thermal inertia of the North Sea. The temperature contrast between day and night reaches a minimum in December (approximately 3 degrees) and a maximum in June (approximately 9 degrees).

Figure 1.8: Figures 1.8(a), 1.8(b), 1.8(c) and 1.8(d) show the daily temperature cycle of all five stations.
1.3.3 Average Wind Speed

Figure 1.9 shows the diurnal cycle of the hourly averaged wind speed for the five different stations. In general, at the coastal stations higher wind speeds are observed compared to the inland regions. This can be explained by the fact that there is less friction above sea, allowing the wind to accelerate more. Other reasons could be the specific locations of the stations within a city, which are not examined in detail further. At night the wind speed is higher in the winter than in April or June and during the day the wind speeds are highest in February.

Wind speed is an important aspect concerning ground concentrations, because the amount of non-convective turbulence is dependent on the (vertical gradient in) horizontal wind speed. So milder winds causes less turbulence which will lead to higher ground concentrations, as discussed earlier in section 1.2. At night the wind speed is lower in the summer because of the larger temperature difference during the day. Because of the relatively warm day a stratification occurs during the night where cold air is trapped between the surface and a layer which is warm. This way winds at the surface are not able to accelerate as much as they do in the winter[6].
Based on figures 1.7, 1.8 and 1.9 a preliminary conclusion can be made about the expected PM concentration at the different stations. In Amsterdam wind speeds are always higher which suggest that the surrounding atmosphere is ‘refreshed’ quicker than for example in Utrecht. There the wind will not disperse the particulates as much as in the other station leading to a higher ground concentration. Besides the station in Amsterdam is located in a park and specified as a city/urban station and not as a street/urban station like the other stations so in combination with the higher winds this leads to the assumption that the observed PM concentrations will be lower in Amsterdam compared to the other cities.
Chapter 2

Hourly Particulate Matter Concentrations

Since the meteorological conditions of the stations are examined, the dynamics of the diurnal variations in PM concentration will be investigated next. Normally the hourly measurements\(^1\) have a large uncertainty. This is caused by the radioactive source, which is used in the measurement equipment, and by the fact that water vapor causes noise in dust monitors. Comparison studies show that the uncertainty of the 24 hour average satisfies the legislative 25%\(^2\). In order to have a well defined \emph{average day} enough hours need to be included in the calculation. Our data set contains 13 years of hourly PM concentrations (year 2000 till 2012). As such for each hourly interval the maximum number of data points, in a month with 31 days e.g. December, would amount to \(13 \times 31 = 403\). However, in practice this maximum is largely reduced due to the following aspects:

- Not every station has been operational since 2000;
- Not every station has been operational every hour;
- Weekends are excluded from calculation. This is done in order to use data measured under similar conditions and traffic emissions differ significantly between weekdays and weekends.

Finally, roughly 40 \% to 60 \% of the data is used depending on the station, which still gives a representative average. Furthermore we classified the concentration data according to the magnitude of the ambient wind. As discussed in section 1.2 wind speed is one of the major drivers behind turbulent diffusion of particulate matter.

\(^{1}\)These data of the LML (\textit{Landelijk Meetnet Luchtkwaliteit}) are made available by the RIVM
2.1 Comparison Particulate Matter Concentration for Different Stations

Here, the analysis is discussed for the station in Heerlen. The other stations are analysed similarly. The hourly average PM concentrations are calculated for the four different months within a specific wind speed classification. For December, February and April the wind speed $w$ in m/s is classified in three groups, low wind speed $0 < w < 3$, average wind speed $3 < w < 6$ and high wind speed $6 < w < 9$. In April there are too few data points for the last group to have a representative average and thus we did not include it in the graph. In June the classification is from $0 < w < 2.5$ and $2.5 < w < 5$. In every graph the result for all wind speeds is represented by the black dotted line. The average number of data points used to calculate the hourly average is indicated in the legend by $n$. In the appendix A the number of data points of each individual hour interval can be found for Heerlen in December, which is used to determine $n$ for figure 2.1(a). The uncertainty of the hourly averages is not plotted in the graphs. Because the number of data points each classification is significant the uncertainty is small. We have calculated that the standard deviation of the hourly averages are smaller than $3 \mu g/m^3$.

2.1.1 Station 136: Heerlen

Figure 2.1 shows the hourly average of PM concentration for the different classifications. Also the day and year limit are plotted each month as a reference. In December a continuous increase in the concentration level is observed during daytime. This increase may be caused by an accumulation effect of the pollution emittance during the day. In combination with the fact that little convection is expected during winter, a similar feature appears to be present in February. We expected pollution build-up to occur with specific atmospheric condition, i.e. no convection and low wind speeds, because then pollutants would accumulate within a small volume. Ultimately the average for low wind speed grows as high as the day limit at 18:00 in December.

For the residents of Heerlen this means that every windless day in December the exposure to PM is high and that this can cause long term health problems. The PM concentrations are highly dependent on the amount of wind speed in December and February. This dependence has decreased in April and has almost completely disappeared in June. Reason for this is that radiation intensity has increased and thus convection dilutes ground concentration whether wind speed is high or low. Then the concentration levels are significantly decreased. An interesting evolution is visible in the transition from winter to summer. The cumulative rise in December leading to a peak around 18:00 changes to a different pattern in April were the peak lies in the morning around 09:00. This was expected because in the morning emissions pollute the atmosphere when the mixing layer is still not well developed, so a brief build up of particulates is observed. During the day convection will dilute ground concentration.
Figure 2.1: Figures 2.1(a), 2.1(b), 2.1(c) and 2.1(d) show the hourly average PM$_{10}$ concentration for different wind speeds in Heerlen. The number of data points used is indicated by $n$.

2.1.2 Station 237: Eindhoven

The hourly average concentrations of PM for the second station, Eindhoven, can be seen in figure 2.2. As was seen in Heerlen an average day in December is defined by the cumulative rise of the concentration. Then for Eindhoven when winds speeds are low ($0 < w < 3$) the hourly average becomes 10 % higher than the daily limit at the beginning of the evening. Concentrations in Eindhoven are higher than in Heerlen, this can have multiple reason e.g. the location of the RIVM station in relation with traffic in a city or a difference in source intensity. In February the day limit is reached from 10:00 till 22:00. More resemblances with Heerlen are observed in April, where concentrations are maximal in the morning, and in June, there sensitivity to the magnitude of the ambient wind has decreased. This can be explained
by the fact that there is more radiation during the summer and thus convection is dominant creating shear driven turbulence. Then there are high levels of turbulence in the atmosphere and a large mixing layer is formed, horizontal wind speeds are not necessary for dispersing pollutants. Though the diurnal cycle of PM concentration in Eindhoven is similar towards Heerlen, the levels are consequently higher.

Figure 2.2: Figures 2.2(a), 2.2(b), 2.2(c) and 2.2(d) show the hourly average PM\textsubscript{10} concentration for different wind speeds in Eindhoven.
2.1.3 Station 433: Vlaardingen

The results of the third station can be seen in figure 2.3. In many ways Vlaardingen is similar to Heerlen. The transition from winter to summer is equal and the concentration levels maximize at the same time near the day limit. The morning peak in April is not as high as it is in Eindhoven.

![Graphs showing PM10 concentrations](image)

Figure 2.3: Figures 2.3(a), 2.3(b), 2.3(c) and 2.3(d) show the hourly average PM$_{10}$ concentration for different wind speeds in Vlaardingen.
2.1.4 Station 520: Amsterdam

The hourly average PM concentrations in Amsterdam differs slightly from the other stations, see figure 2.4. The cumulative growth of concentration we observed for the other stations in December is hardly visible and the peak at 18:00 is approximately 25% lower near the year limit. A third difference can be seen in April, were the morning peak lasts until 13:00 in stead of 10:00. Naturally there are some similarities with the other stations, like the diurnal variations in February and the diminishing wind dependence in June. The differences with the other stations can be caused by the specific location of the station within the city. The station is specified as a city/urban station, in contrast with the other stations which are street/urban stations. This means that the other stations are located closer to traffic emission sources.

Figure 2.4: Figures 2.4(a), 2.4(b), 2.4(c) and 2.4(d) show the hourly average PM$_{10}$ concentration for different wind speeds in Amsterdam.
2.1.5 Station 639: Utrecht

For the last station, Utrecht, many resemblances with the first three stations are observed, see figure 2.5. The accumulation in December and morning peak in April are all visible. Typically the maximum levels of PM concentrations are around the day limit, for the low wind classification. In general, very systematic behavior is observed.

![Graphs showing hourly average PM10 concentration for different wind speeds in Utrecht.](image)

*Figure 2.5: Figures 2.5(a), 2.5(b), 2.5(c) and 2.5(d) show the hourly average PM10 concentration for different wind speeds in Utrecht.*
2.2 Conclusions and Discussion

In general, the results from the various stations appear to be consistent. This is illustrated by figure 2.6 which shows a combination of the results for all stations in the class with low wind speed, i.e. \(0 < w < 3\). In the winter hourly PM concentrations rise during the day and are very dependent on wind speed. When the wind speed is low the ground concentrations are not diluted effectively and thus the concentrations are high, averaged around the day limit of 50 µg/m³ at 18:00. When wind speed is high, \(6 < w < 9\), the concentration is always low, because then turbulence mixes air pollution rapidly.

In the transition from winter to the spring we observed a peak shift from the evening towards the morning. As discussed in section 1.2, in winter wind speed alter the amount of turbulence and convection does not play a major role, but that changes during the summer. During a day in the summer convection is dominant – because there is much more radiation – and thus ground emissions are mixed regardless of the strength of the horizontal wind. In April the hourly concentrations peak in the morning and that was expected because in the morning emissions pollute the atmosphere when the mixing layer is still not well developed, so a brief build up of particulates is observed. During the day convection will dilute ground concentration. In our opinion the consistency of the results suggests that the diurnal variations of PM concentration is governed by rather simple basic mechanisms. As such in the next chapter a conceptual model will be designed to examine this.

![Figure 2.6: The above figures show the hourly average PM_{10} concentration for low wind speeds for all stations.](image-url)
Lastly the consistent results have some consequences, especially for commuters. The CBS (Centraal Bureau voor de Statistiek) has registered that half of all workers in the Netherlands do not work in their own municipality and thus have to travel in the morning to go to work and in the evening to get back home[7]. Thus commuters are consequently exposed to elevated concentrations when they are travelling. During a day with low wind speed the exposure is high during the evening rush hour in the winter months and high during the morning rush hour in the so called transition months between winter and summer. This is illustrated in figure 2.6 with a black arrow. For commuters it is not meaningful to monitor day averages, because they are active when hourly concentrations are consequently higher, which means that they are exposed to more particulate matter.
Chapter 3

Modeling the Hourly Particulate Matter Concentration

3.1 Model Setup

As concluded in the previous chapter, the diurnal variations in PM concentrations appear to be governed by basic mechanisms, judging from the general patterns observed for all stations. Now a model will be designed, based on the seasonal meteorological differences between the examined months, as discussed in section 1.2. Figure 3.1 shows the set up for this model.

Figure 3.1: This figure shows the set up used to make the model. The concentration observed by a station is dependent on the amount of ground emissions, the height of the mixing layer $h$, and the background concentrations. These are all time dependent.

The PM concentration observed at a RIVM station, $C_{obs}$ in $\mu g/m^3$, is the quantity of interest and we make the assumption that its change is only determined by the difference with the concentration of its surroundings. The surroundings consists of the background environment concentration $C_{env}$ plus the so called city concentration $C_{city}$. It is expected that when the pollution source will be eliminated that the observed concentration will relax to its background value. There, we assume that this relaxation process can be characterized by a typical time scale $\tau_{env}$. As definition $\tau_{env} = \frac{1}{k_{env}}$, with $k_{env}$ the effective rate constant for the removal of particulates due to advection and precipitation. For the typical time scale of the system response after a sudden increase in street emission $\tau_{street}$ is used. Similar as definition $\tau_{street} = \frac{1}{k_{street}}$, here $k_{street}$ is the effective rate constant for observing an additional particulate by the RIVM station. The RIVM stations we observed this thesis are all within the borders of a city and very close to traffic. As such the response towards a suddenly increase in concentration...
due to pollution in the city will be faster than the relaxation time towards the environmental levels so the time constant $\tau_{\text{street}}$ must be smaller than time constant $\tau_{\text{env}}$. To make an indication of the order of $\tau_{\text{street}}$ a simple calculation shows that in a city with a section of 10 km where wind speeds are 2 m/s, it would take 5000 seconds ($\approx 1.5$ hours) to ‘clean’ the polluted atmosphere in the city. However in this calculation the assumption is made that footprint of the traffic emission is as large as the size of a city, but cities are usually part of an urban agglomeration and as such the polluted area can be significantly increased. Hence it would take more time to ‘refresh’ the city and thus we expect the time constants to be larger. Similar we observed, e.g. figure 2.6(a), that in the night when emissions are low it takes the entire night for the PM concentrations to evolve towards the environmental background, which also indicates larger time scales. However it is difficult to physically interpret the timescales specifically. The following differential equation results

$$\frac{dC_{\text{obs}}(t)}{dt} = \frac{C_{\text{city}}(t) - C_{\text{obs}}(t)}{\tau_{\text{street}}} - \frac{C_{\text{obs}}(t) - C_{\text{env}}(t)}{\tau_{\text{env}}} \quad (3.1)$$

Equation (3.1) shows that $C_{\text{obs}}$, $C_{\text{city}}$ and $C_{\text{env}}$ are all time dependent. For $C_{\text{city}}$ and $C_{\text{env}}$ a daily periodic time dependency will be prescribed. In order to investigate the diurnal variations of the environmental background concentration a station is examined, representing a rural area. The RIVM station used for this is 929 in Valthermond, for the location see figure 1.2. In June the hourly average concentration in Valthermond can be seen in figure 3.2(a). These diurnal variations are used for every month to model the environmental background concentration, see figure 3.2(b).

![Figure 3.2](image-url)

**Figure 3.2:** Figure 3.2(a) shows the hourly PM concentration in Valthermond in June and figure 3.2(b) shows an idealization of $C_{\text{env}}$ used in the model based on figure 3.2(a).
In a city different emitters contribute to concentration increase as compared to the background concentration, for instance traffic, industry or construction. In order to keep this model basic we assume that the hourly city concentration is highly dependent on the traffic density. Thus $C_{\text{city}} = C_{\text{env}} + C_{\text{street}}$. The assumption is made that particulate matter that originates from non-traffic sources, e.g. sea salt or industrial areas in Germany or Belgium, is included within the environmental background. Hence the street concentration is only determined by the amount of traffic. The traffic density is modelled using data from the CBS (Centraal Bureau voor de Statistiek), which has investigated the mobility of the Dutch population\[8\]. This result can be seen in figure 3.3(a) and it is used to model the scaled emission of the city. The shape of the scaled ground emission of the city, $E_{\text{city}}(t)$ see figure 3.3(b), is inspired by the daily periodic variations of the mobility of the Dutch people. That is because we assume a linear dependency between the city emission and the traffic density. As such $C_{\text{street}}$ in $\mu g/m^3$ is calculated by dividing $E_{\text{city}}$ by the height of the mixing layer $h$ in m and multiplying it with a calibration factor $N$ in $\mu g/m^3$, thus $C_{\text{city}} = C_{\text{env}} + \frac{E_{\text{city}} \cdot N}{h(t)}$. Here the assumption is made that all particulates are trapped within the boundary layer and no removal due to e.g. precipitation is present. $N$ is the parameter to calibrate the model. This will only be done once.

![Figure 3.3](image)

**Figure 3.3:** Figure 3.3(a) shows the mobility of the Dutch population and figure 3.3(b) shows the scaled emission of the city used in the model, which is inspired on figure 3.3(a).

As stated earlier the seasonal changes in the amount of turbulence during the day and night determine the monthly differences in PM concentrations. This will be simulated in the model with use of seasonal time dependency of the height of the mixing layer $h$. During the winter, December and February, daily temperature differences are small, the sun rises late and sets early. This way the mixing boundary layer is relatively thin and because there is only little convection during the day the height of the mixing layer only grows to 500 meter. At night the height is 200 meter. In April the sun is up much longer and thus the mixing layer grows higher, namely 800 meter during the day. Finally in the summer in June when convection is the most dominant the maximum mixing height is 1100 meter.
We used a Gaussian function where the width is determined by the time of sunrise and sunset. The shape was based on figure 1.7, which shows the average radiation for each station. Based on time of sunrise and sunset the following Gaussian distribution \( \mathcal{N}(\mu, \sigma^2) \) was chosen, with 
\[
\mu = \frac{t_{\text{sunrise}} + t_{\text{sunset}}}{2} \quad \text{and} \quad \sigma = (\mu - t_{\text{sunrise}}) \cdot \frac{4}{10}
\]

- December: \( t_{\text{sunrise}} = 8.5 \) and \( t_{\text{sunset}} = 16.5 \) so \( \mathcal{N}(\mu = 12.5, \sigma^2 = 2.56) \);
- February: \( t_{\text{sunrise}} = 8 \) and \( t_{\text{sunset}} = 17.75 \) so \( \mathcal{N}(12.88, 3.81) \);
- April: \( t_{\text{sunrise}} = 7 \) and \( t_{\text{sunset}} = 20.5 \) so \( \mathcal{N}(13.75, 7.29) \);
- June: \( t_{\text{sunrise}} = 5.33 \) and \( t_{\text{sunset}} = 22 \) so \( \mathcal{N}(13.66, 11.10) \).

Figure 3.4 shows the assumed height of the mixing layer used in the model for the four different months. The shape of the mixing height is debateable, since here it is not based on atmospheric observational data directly. A more realistic shape would be one which rises as a Gaussian function like we used, then stagnates to a plateau and finally it will collapse quickly and evolve into a residual layer after the sun sets, as discussed in section 1.2 and is visualized in figure 1.4. We used the Gaussian shape due to its simplicity, as such this aspect has to be investigated further, and because it is not the goal of this model to investigate the evolution of the mixing height during the day, we will not examine it further. The optimal value was found to be 400 meter in June at night and thus larger than in the other months. This contradicts the fact that wind speeds are lower in June, see figure 1.9(d), what would normally result in a lower mixing height because at night the mixing height is proportional to the ambient wind. But for the results in June it was optimal to set the night mixing height at 400 meter.

Now we can rewrite equation (3.1) to find the differential equation used in the model:

\[
\frac{dC_{\text{obs}}(t)}{dt} = \frac{N \cdot E(t)}{h(t)} + C_{\text{env}} - C_{\text{obs}}(t) \frac{C_{\text{obs}}(t) - C_{\text{env}}(t)}{\tau_{\text{street}}} - \frac{C_{\text{obs}}(t) - C_{\text{env}}(t)}{\tau_{\text{env}}}
\]  

(3.2)
Figure 3.4: Figure 3.4(a), 3.4(b), 3.4(c) and 3.4(d) show height of the mixing layer $h$ in m during the day.
3.2 Limit Analysis

The first limit of equation (3.2) which is examined is the absence of emission sources, so $N = 0$. As such equation (3.2) can be written as

$$\frac{dC_{obs}(t)}{dt} = \frac{C_{env} - C_{obs}(t)}{\tau_{street}} - \frac{C_{obs}(t) - C_{env}(t)}{\tau_{env}}$$

$$= -(C_{obs}(t) - C_{env}(t)) \left( \frac{1}{\tau_{street}} + \frac{1}{\tau_{env}} \right)$$

$$= \frac{C_{obs}(t) - C_{env}(t)}{\tau}$$

(3.3)

This results in a steady state with $C_{obs} = C_{env}$.

Equation (3.3) suggests an exponentially decrease of $C_{obs}$ towards $C_{env}$ with time constant $\tau$, which is exactly what would happen if no sources would be present. Next the stationary solution is considered. Then $\frac{dC_{obs}}{dt} = 0$ and equation (3.2) becomes

$$0 = \frac{N \cdot E(t)}{h(t)} + C_{env} - C_{obs}(t) = \frac{\tau_{street}}{\tau_{env}} (C_{obs} - C_{env})$$

$$\Leftrightarrow N \cdot E(t) = (1 + \frac{\tau_{street}}{\tau_{env}})(C_{obs} - C_{env})$$

$$\Leftrightarrow C_{obs} = C_{env} + \frac{N \cdot E(t)}{(1 + \tau_{street}/\tau_{env})}$$

For $\tau_{env} \gg \tau_{street}$,

$$C_{obs} \approx C_{env} + \frac{N \cdot E(t)}{h(t)}$$

(3.4)

Physically this tells us that if we allow the system to find its equilibrium, i.e. $t \gg \tau_{env} \gg \tau_{street}$, the observed concentration at the station will be the background environmental concentration plus the emissions from the city. Again when the pollution source is eliminated, the steady state is the same as in equation 3.3.
3.3 Results and Conclusions

With equation (3.2) the diurnal cycle of PM concentration will be simulated, unfortunately with this basic conceptual model classifications in the magnitude of the ambient wind can not be made. As such the model will simulate the general classification with all wind speeds, which were visualized in the graphs of the previous sections as black dashed lines. In order to model the different classification the model needs to be extended.

Thus far the environmental concentration, city emissions and daily evolution of the mixing height are prescribed, so lastly time constants $\tau_{\text{street}}$, $\tau_{\text{env}}$ and calibration factor $N$ need to be set. As stated earlier the time constant $\tau_{\text{street}}$ must be smaller than time constant $\tau_{\text{env}}$, because the RIVM stations we observed are all within the borders of a city and very close to traffic. The response towards a suddenly increase in concentration due to pollution in the city will thus be faster than the relaxation time towards the environmental levels. This can also be seen in figure 2.6(a), here the observation is made that during the night when no emissions are present it takes all night for the observed concentration to evolve towards the environmental level.

Firstly the time constants are set, resulting in $\tau_{\text{street}} = 2$ hours and $\tau_{\text{env}} = 7$ hours. This is done by calibrating the model’s result on the observations in December. Thus by trial and error the optimum is found such that the model’s result give the most similar representation of the observed diurnal variations in PM concentration in December. Because no classification can be made on wind speed, the model’s results are compared with the observations of all wind classes. The timescales are higher than 1.5 hours, which was calculated in section 3.1 as an indication. In this simple calculation however, an effect as resuspension of matter within the city is not included. Furthermore in chapter 2 results of the diurnal cycle of PM concentrations indicate larger timescales, see figure 2.6, because as stated earlier at night when emission levels are low the observed concentration evolves towards the background environmental level the entire night. This contradicts the physical interpretation of the timescale, since the time scales are too large to be the characteristic time for an observant response towards a sudden increase of street concentration or the evolution towards the environmental background, as we defined earlier. An explanation can be that the observed concentration at a street/city station does not only represent the concentration of that street/city, but for the entire urban agglomeration. Then larger time scales are necessary, because the area (i.e. footprint) over which traffic-based particulate matter influence ground concentrations is larger. Since this is only a hypotheses, the intrinsic use of the time scales is only to simulate the diurnal variations and thus at this point no physical interpretation can be given to the values.

During the calibration of the time scales, $N$ is set at $N = 12500\mu g/m^3$, again by trial and error. These parameters were calibrated in order that the model’s result give the most similar representation of the diurnal variations in December, however we will also use these same values for simulating the other months. This is because all seasonal variations are represented within the changing structure of the mixing layer and we have assumed that the mobility of the Dutch population is similar throughout the year.
3.3.1 December and February

For December the result of the model can be seen in figure 3.5. In figure 3.5(a) the hourly average – for all wind speeds – is plotted for the five stations. Next in figure 3.5(b) the result of the model is shown, which has some striking similarities. First the concentration is highest at 18:00 and second the decrease of concentration at night is the same indicating that the model reproduces the wintertime build-up of particulate matter in a shallow boundary layer during the day, followed by a reduction at night when emissions are lower. However there are also some differences. A larger peak at 10:00 is observed in the model than is measured by the station and at noon when the average hourly concentration stagnates the model shows a decrease in concentration. Both peaks are to sharp, compared to the result from figure 3.5(a) and this is largely because of the gaussian shape of the mixing layer we have chosen. If a more specific time evolution of the mixing height was used, like visualized in figure 1.4, the model's result would probably be better, so more research need to be done towards this.

Figure 3.5: Figure 3.5(a) shows the average hourly concentrations of all stations examined in a previous section. For December the result of the model is shown in Figure 3.5(b).

The results for February are shown in figure 3.6. The three parameters we have set for December $\tau_{env}$, $\tau_{city}$ and $N$ are valid for all seasons, so the only input we used in this model is the mobility data of the CBS. Also with respect to December the background concentration and the emission of the city are not changed. The only parameter that has changed is the shape of the mixing height, according to figure 3.4. The sun is up a half hour earlier and sets an hour later. Further the maximum height is as high as in December. These changes are not very significant and so the result of the model only changes slightly. Now the morning and evening peak are of the same height, which is similar to the results shown in figure 3.6(a). However the evening peak is around 17:00 in the model and not at 19:00. Again the peaks are to sharp, because of our chosen mixing layer shape.
3.3.2 April and June

For April figure 3.7 shows that changes in the mixing height structure have caused a transition in the profile of the average day, which is similar with the RIVM measured data. Again no changes are made to the time constants, scalar \(N\), environmental concentration or traffic density. So \(\tau_{env} = 2\) hours, \(\tau_{city} = 7\) hours and \(N = 12500\), despite the absence of a physical interpretation the parameters’ values result in many resemblances between the model and the observed variations. Now, due to summertime, the mixing layer is highest at approximately 14:00 instead of 13:00 in December and February. Also the sun is up earlier and sets later and there is more convection during the day. This way emissions in the afternoon are diluted more and thus only the morning peak is visible. This is a good result, however the morning peak is not as significant as it is in figure 3.7(a), nor is the decrease in concentration as significant as in the afternoon. Naturally there are some differences between the stations, e.g. in Vlaardingen the decrease of PM at noon is larger, which can not be simulated with the model, but the overall diurnal variations are similar.
Figure 3.7: Figure 3.7(a) shows the average hourly concentrations of all stations examined in a previous section. For April the result of the model is shown in Figure 3.7(b).

Lastly figure 3.8 shows the result of the model compared to the average hourly concentrations of all stations in June. Similar as observed the concentrations are always low in June and no significant structure – despite the minor morning peak that is visible in the model – is visualized. This is caused by the higher mixing layer diluting the emissions of the city more than in April.

Figure 3.8: Figure 3.8(a) shows the average hourly concentrations of all stations examined in a previous section. For June the result of the model is shown in Figure 3.8(b).
Chapter 4

Day Limit Violation Probabilities

In chapter 2 the diurnal cycle of PM concentration was examined for different months and with specific atmospheric conditions – e.g. low wind speeds – the hourly average may well exceed the day limit. However this observation does not tell anything about the total number of limit exceedances. This information is given by the RIVM, which publishes tables every year that give the number of exceedances of the day limit per station per year. By European legislation a station is allowed to violate the day limit – so the 24 hour average is higher than 50 \( \mu g/m^3 \) 35 times annually. In this section violations of the day limit will be considered by investigating the probability to exceed the limit. Based on the data, which is the same set as is used in the previous sections, the probability of violating the day limit is simply calculated by dividing the number of exceedances by the total number of data points.

4.1 Probability per Station

To calculate the probability per hour (per classification) scatter plots are used. An example can be seen in figure 4.1, here a scatter plot is made for the hourly PM measurements in Heerlen at 18:00 (i.e. 17:00 till 18:00) in December. In total the station measured the PM concentration 187 times and with 47 of those measurements the day limit was exceeded. As such the probability is 25 \%, meaning that generally in Heerlen at 18.00 in December once every four days the day limit is exceeded, regardless the magnitude of the wind. Similar as in previous sections, classifications according to the wind speed regime are made. From the scatter plot, figure 4.1, it follows that for low wind speed, i.e. \( 0 < w < 3 \), 35 of in total 73 measurements violate the day limit. This means that 48 \% of all days with low wind speed the day limit is exceeded in Heerlen at 18:00 in December. Such probabilities are calculated for every hour with use of scatter plots. In appendix B the scatter plots can be seen of every stations at 18:00 each examined month. Lastly figure 4.1 shows the relation between the ground concentration of particulate matter and the wind speed. This appears to be a non-linear relationship, because the concentration decreases rapidly when wind speed is increased.
Figure 4.1: A scatterplot is shown of the concentration versus the wind speed at 18:00 in Heerlen. The number of data points included is $n = 187$. 

\[ \text{PM10 Concentration (\mu g/m}^3) \]

\[ \text{Windspeed (m/s)} \]
4.1.1 Station 136: Heerlen

Figure 4.2 shows the hourly probability of violating the day limit in Heerlen each month per wind speed classification. In figure 4.2(a) the examples discussed in the introduction of this section can be found. In general all figures show a large resemblance with the diurnal cycle of average PM concentration observed in section 2.1.1. In December the PM concentration continuously increased until it reached its maximum at 18:00 and from figure 4.2(a) the same observation is made. Then there is a 48 % chance of violating the day limit. The second resemblance is observed in April where probabilities in the beginning of the evening have dropped to 20 % and in the morning probabilities peak and are almost 40 %. Thirdly in June the probabilities are always low, despite low or high wind speeds, similar like the observation that the average concentrations is low in June, because convection is dominant.

Figure 4.2: Figures 4.2(a), 4.2(b), 4.2(c) and 4.2(d) show the probability to violate the day limit PM\textsubscript{10} concentration at different hours.
4.1.2 Station 237: Eindhoven

For Eindhoven the diurnal variations in probabilities are similar with Heerlen, except that they are systematically higher, see figure 4.3. The cumulative rise in December for low wind speeds \((0 < w < 3)\) reaches its maximum at 14:00, but stays around 45 % until 19:00. In February probabilities are 50 % from 10:00 until 16:00 for low wind speeds. The general classification (black dashed line) with all wind speeds results in a peak in the morning for February and decreases at noon towards probabilities similar at night. That can be explained by the fact that the magnitude of the horizontal wind is greatest at noon, so more turbulence is generated. In April the midday probabilities are lower than nightly hours, but during the morning rush hour they increase again. In June probabilities are low and not dependent on wind speed.

\[\text{Figure 4.3: Figures 4.3(a), 4.3(b), 4.3(c) and 4.3(d) show the probability to violate the day limit PM}_{10} \text{ concentration at different hours.}\]
4.1.3 Station 433: Vlaardingen

For the third station, i.e. Vlaardingen, probabilities are not as high as in Eindhoven, but during a day in February when wind speed is low a significant portion of the day there is a 40% chance to violate the day limit, see figure 4.4. For average wind speed the probability is highest in the morning around 09:00. In April the same variations are observed as in Eindhoven, generally that the probability is minimal after noon. This is reversed in December, when the probabilities are lowest at the end of the night. Again this shows many resemblances with the diurnal variation of the PM concentration, see figure 2.3.

Figure 4.4: Figures 4.4(a), 4.4(b), 4.4(c) and 4.4(d) show the probability to violate the day limit PM$_{10}$ concentration at different hours.
4.1.4 Station 520: Amsterdam

The diurnal cycle of PM concentrations in Amsterdam, discussed in section 2.1.4, showed differences with the other stations. The cumulative rise in December was not observed and the morning peak in April lasted longer. In figure 4.6 these differences can be observed again. In December from 11:00 until 23:00 the probability fluctuate around 30% and in April the morning peak lasts until 13:00, while at the other stations it lasted till 10:00. Despite the lower probabilities in December, significant chances to violate the limit are seen in February and April for low wind speed classification. The general classification shows great resemblances with the first three stations.

![Figure 4.5](image1)

**(a) December**

![Figure 4.5](image2)

**(b) February**

![Figure 4.5](image3)

**(c) April**

![Figure 4.5](image4)

**(d) June**

*Figure 4.5: Figures 4.5(a), 4.5(b), 4.5(c) and 4.5(d) show the probability to violate the day limit PM$_{10}$ concentration at different hours.*
4.1.5 Station 639: Utrecht

Lastly the hourly probabilities of violation in Utrecht are shown in figure 4.6. The general classification rises continuously in December, has maxima in the morning and evening in February, has only a morning maxima in April and is always low in June. The probabilities for low wind speeds differ from the other stations, for instance in February it is peaked at 14:00.

Figure 4.6: Figures 4.6(a), 4.6(b), 4.6(c) and 4.6(d) show the probability to violate the day limit PM$_{10}$ concentration at different hours.
4.2 Conclusions and Discussion

In general consistent behavior between the stations with respect towards the hourly probabilities are observed. In December probabilities are highest at 18:00 and decrease rapidly during the night. When wind speed is low chances typically become as large as 50%. In February the maximum has already shifted a bit towards the morning and probabilities are still very sensible for the magnitude of the wind speed. In April increasing radiation causes more convection and in combination with the longer lasting days generally a significant morning peak is observed during the morning rush hour. The wind dependence has decreased and it is further diminished in June. Then at every hour probabilities are low.

As stated in section 2.2, commuters are consequently exposed to elevated levels and their exposure is underestimated by only monitoring the day limit. This can now be further elaborated because we can calculate the expected total exceedances in a year for commuters based on the probabilities. As a result of our choice to examine only December, February, April and June, the assumption has to be made that January is similar to December and November towards February. This assumption is based on the hypothesis that the monthly differences are cause by the seasonal changes like the amount of radiation which causes convection, as we discussed in section 1.2. As such we further assume that March, September and October are similar like April and May, July and August are similar to June.

Then a calculation can be made for a commuter for example near Eindhoven how often the day limit of 50 µg/m^3 is violated at 10:00 and 18:00. With use of figure 4.3 the amount of exceedances at 18:00 in January and December is 0.28 \times (31 + 31) = 17.4 and in February and November 0.27 \times (28 + 30) = 15.7 – probabilities of the general classification are taken for the calculation. For March, April, September and October this amount is 0.08 \times (30 + 31 + 31 + 30) = 9.8 and for May, June, July and August it will be 0.08 \times (31 + 30 + 31 + 31) = 9.8. As such the expected number of violations in a year at 18:00 in Eindhoven is 53. At 10:00 the amount of exceedances in January and December is 0.20 \times (31 + 31) = 12.4 and in February and November 0.34 \times (28 + 30) = 19.7. For March, April, September and October this amount is 0.34 \times (30 + 31 + 31 + 30) = 41.4 and for May, June, July and August it will be 0.10 \times (31 + 30 + 31 + 31) = 12.3. As such the expected number of violations in a year at 10:00 in Eindhoven is 86. By legislation of the European Union the day limit is allowed to be violated 35 times in a year, based on a 24 hour average. With this 24 hour average the amount of violation in a year in Eindhoven is 31. These amounts show that a commuter, that travels near Eindhoven between 09:00 and 10:00 in the morning and 17:00 and 18:00 in the evening, is exposed to more particulate matter than is expected when 24 hour averages are used instead of hourly averages.
Chapter 5

Summary

The World Health Organization states that air pollution causes a decrease in life expectancy of 3 years in severe polluted areas\cite{1} and the OECD predicted that Particulate Matter (PM) will become the top cause of environmentally related deaths worldwide by 2050\cite{2}. In contrast with for example SO$_2$ and NO$_2$ and despite of these warnings the European Union only has regulations for year and 24 hour averages of PM$_{10}$, which have to be lower than respectively 40 µg/m$^3$ and 50 µg/m$^3$. So no research is done towards hourly averages of PM. In this thesis we have examined the diurnal dynamics of hourly PM concentrations for five different station throughout the Netherlands for the months December, February, April and June. Classifications were made according to wind speed. Reason for this is the fact that horizontal wind shear and convection are the two major drivers of atmospheric turbulence.

First the diurnal variations of PM concentration was examined and a strong sensitivity for the magnitude of horizontal (10m) wind speed was observed in the winter, but not in the summer. This can be explained by convection becoming dominant in June, because then there is much more radiation. This dilutes the ground emissions despite of the magnitude of the wind speed. In December and February hourly average PM concentrations when wind speed is low – i.e. 3 m/s or less – could become more than twice as large as concentrations when wind speed is high, i.e. 6 m/s or more. Furthermore similar behavior was observed between the stations with respect to the seasonal variations of PM concentration. To begin in December concentrations would show a continuous rise during the day and reach its peak at 18:00. Then after sunset the average concentration would decrease during de night reaching its minimum at 06:00. In February the evening peak had shifted towards noon for low wind speed creating broad daytime maximum. For average wind speed two equal peaks are observed, in the morning and evening. Nocturnal concentrations are lowest at 06:00. These observations can be explained by the fact that the intensity of solar radiation is minimal during the winter and thus convection can generate only little turbulence. Hence when wind speeds are low, an accumulation of particulates within a small volume can appear.

In April seasonal changes were observed, since the PM concentration was maximal only in the morning at 10:00 and during midday concentrations would become lower than the night average. This is because in the morning the mixing layer is not well developed – due to the time of sun rise and radiation intensity – and thus an accumulation of particulates can
occur. However during the day convection will become more dominant and dilute ground concentrations. This had further evolved in June where wind dependence had fully diminished and concentrations would not fluctuate much during the day. In general, the stations showed similar seasonal variations and as such we believe the diurnal variations of particulate matter concentrations within an urban area are governed by rather basic mechanisms. During the winter months the sun rises late, sets early and the radiation intensity is low, causing a large stable boundary layer during the night and only a small mixing boundary during the day when emission are highest. In the summer the sun is up early and sets late in the evening. This way the height of the mixing boundary layer is much larger and ground emissions are diluted more.

These changes in the mixing boundary layer were used in a model to simulate the seasonal differences in hourly average PM concentrations. Only input data of the CBS (Centraal Bureau voor de Statistiek) was used to model the emission of the city, by assuming that the hourly city concentration is linear dependent with the traffic density. To model the environmental background concentration a station in Valthermond was examined, because it is located in a rural area. The time constants were set as $\tau_{\text{street}} = 2$ hour and $\tau_{\text{env}} = 7$ hour and lastly parameter $N$ was calibrated resulting in $N = 12500$ $\mu g/m^3$. This means a higher value for the time scales was found than a physical interpretation would suggest, i.e. 1.5 hours. This can indicate that the observed concentration at a street/city station does not only represent the concentration of that street/city, but for the entire urban agglomeration. Then larger time scales are necessary, because the area (i.e. footprint) over which traffic-based particulate matter influence ground concentrations is larger. Since this is only a hypotheses, the intrinsic use of the time scales is only to simulate the diurnal variations and at this point no physical interpretation can be given to the values. The three parameters are valid for all seasons. The only parameter which changes is the shape of the mixing layer. We used a Gaussian shape which is dependent with the time of sun rise and sun set. This is an approximation of the more realistic shape which rises as a Gaussian shape, then stagnates after noon and diminishes after sunset.

In December the model accurately simulated the maximum at 18:00 as well as the decrease of concentration during the night. Only the accumulate rise during the morning was not represented well, because the model showed a morning peak and a decrease around noon. In February the model is similar due to the fact that the time of sunrise and sunset is nearly the same as in December and the height of the mixing layer is small. The model showed two equal peak in the morning and evening. In April a morning peak is accurately simulated by the model, but concentration levels are not as large as observed earlier. Secondly the decrease in concentration at noon is not as significant as observations we made earlier. The result of the model in June does represent the hourly concentration well, because then concentrations are near the 24 hour average throughout the entire day. In general, with exception of the peaks which were simulated too pungent due to the simple Gaussian shaped mixing layer, the model correctly predicts the diurnal variations of PM concentration, while only data of the CBS is used as input.

Lastly the probability per hour to violate the day limit of 50 $\mu g/m^3$ is examined, which has an allowed number of exceedances of 35. Because a large group of commuters are active when hourly average PM concentrations are highest they are consequently exposed to higher...
concentrations than the day average. In section 4.2 a simple calculation is made to show that
commuters that travel near Eindhoven at 10:00 and 18:00 are exposed to concentrations which
are higher than the day limit in the evening 53 times and in the morning 86 times in a year.
We made the assumption – because only four months were examined– that for example March,
September and October are similar to April, because solar radiation intensity is approximately
equal those month, but more research needs to be done towards this. Furthermore the health
hazards especially for commuters, should also become an topic of interest because with present
regulations of the European Union their exposure is consequently underestimated.
Bibliography


Appendix A

Number of data points

In this appendix the table is shown, which is used in section 2.1 to calculate the average included number of data points.

Table A.1: The table shows the number of data points used to calculate the hourly average per classification

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185±2 69±5 75±7 32±4
Appendix B

Scatterplots of all five stations

In this appendix the scatter plots can be found, which are used to calculate the probability to violate the day limit at each hour interval, for the different wind classifications, stations and months.

B.1 Station 136: Heerlen

Figure B.1: Figures B.1(a), B.1(b), B.1(c) and B.1(d) show a scatter plot of the concentration versus the wind speed at 18:00 in Heerlen.
B.2 Station 237: Eindhoven

Figure B.2: Figures B.2(a), B.2(b), B.2(c) and B.2(d) show a scatter plot of the concentration versus the wind speed at 18:00 in Eindhoven.
B.3 Station 433: Vlaardingen

Figure B.3: Figures B.3(a), B.3(b), B.3(c) and B.3(d) show a scatter plot of the concentration versus the wind speed at 18:00 in Vlaardingen.
B.4 Station 520: Amsterdam

Figure B.4: Figures B.4(a), B.4(b), B.4(c) and B.4(d) show a scatter plot of the concentration versus the wind speed at 18:00 in Amsterdam.
B.5 Station 639: Utrecht

Figure B.5: Figures B.5(a), B.5(b), B.5(c) and B.5(d) show a scatter plot of the concentration versus the wind speed at 18:00 in Utrecht.