# Model simulation of the morning transition effect on boundary layer dynamics and chemistry during the PEGASOS campaign in the Netherlands

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# Preface

The master thesis that is lying in front of you is the result of eight-months' work and research at the department of Meteorology and Air Quality of Wageningen University and Research Center.

The whole process of this work and the report itself has experienced various ups and downs. Especially, the first months I experienced difficulties to find the right research topic for my case and formulate the research questions. Moreover, familiarize myself with the model, the data and in general the methods to process them resulting in me spending the first three to four months with those. It was important for me to carry a research that I was feeling comfortable with and I understood it. It was important for me also to find interesting the research topic that I had chosen since the whole project was intriguing and had a lot to offer. I think that through this research I gained a lot of knowledge for the physical processes of the boundary layer meteorology and chemistry. I realized the way research is conducted and the difficulties or disappointments you will face during the process.

First, I want to thank my supervisor Jordi Vilà-Guerau de Arellano for his help and good advices. Even though through the process there were arguments, this gave me a stronger motivation to continue.

Second, I want to thank Eduardo Barbaro for the numerous e-mails and meetings throughout these eight months, for his most useful help with the everyday difficulties and for listening, consulting and supporting when needed.

Third, I want to thank my mother for the moral and financial support reminding me to do my best, work hard and by this time next year I will have forgotten every disappointment or tiredness.

Last, but not least, I want to thank the person that I am sharing every day for almost two years now. His constant support, advices and listening made me not to lose my courage and or my belief in me.

Dimitra Kalosynaki
24-05-2014

# Summary

In this thesis we focus on the investigation of the morning transition phenomenon. The morning transition is defined as when the nocturnal stable boundary layer (SBL) transforms into a diurnal convective boundary layer (CBL). Even though the CBL has been exhaustively studied little focus has been given to the transition itself. The importance of studying the morning transition phenomenon is twofold since (i) sets the initial conditions for the CBL depth development and (ii) defines the timing that the chemicals start to mix with air masses and react with each other throughout the turbulent layer.

This study investigates and provides a quantification of the morning transition impacts on the boundary layer dynamics and on the chemistry based on surface and airborne measurement, processed with a MiXed Layer (MXL) model. The surface measurements are obtained from the Cabauw meteorological tower and the airborne data are obtained from the PEGASOS project during May 2012. During the selected day for this study, the 27$^{th}$ of May 2012, a Zeppelin platform was used to sample and measure variables during the beginning of the developing convective boundary layer in the Netherlands. The measurement strategy consists of performing measurements along the 213-m Cabauw tower, covering a range of heights above and below the height of the developing convective boundary layer.

Firstly, the case was reproduced and the model was validated. We concluded that the model is able to provide a reliable representation of the dynamics-chemistry interaction. Lastly, the most important factors affecting the morning transition phenomenon have been identified by performing a sensitivity analysis. Three mechanisms, shear, partitioning of surface fluxes and strength of inversion have been identified as the key parameters affecting the fashion of the morning transition. Our results showed that surface fluxes partitioning, leaded to the most significant changes on the CBL development by enhancing the CBL growth and consequently increasing the ozone concentrations.

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# Introduction

Morning transition is the process that a nocturnal stable boundary layer (SBL) transforms into a diurnal convective boundary layer (CBL). Even though the CBL have been the interest of many studies that have been working with meteorological and air quality phenomena, little focus have been given to the transformation itself (Angevine, Baltink, & Bosveld, 2002). Morning transition is a good case of dynamic variations that occur at the lowest part of the atmosphere and govern the chemical reactions (Sokol, Stachlewska, Ungureanu, & Stefan, 2014). Thus, investigating this phenomenon can provide valuable information for enhancing our understanding for the behavior of the lower part of atmosphere.

It has been suggested that the depth of the nocturnal SBL and the timing of the transition are critical characteristics that help to form prognostics relations for the CBL (Beare, 2008). Moreover, a layer called residual layer (RL) exists above the SBL, containing chemicals as remnants of the previous day. Often during the early morning, the RL exists for a while until it is being fully entrained into the CBL. As a result, chemical reactions among the RL's components are triggered, influencing the chemical composition of the CBL (Stull, 1988). Morning transition is an important intermediate phase, setting the initial conditions for the CBL depth and defining the timing that the chemicals start to mix with other air masses and react with each other (Doran, Berkowitz, Coulter, Shaw, & Spicer, 2003). A detailed investigation of the timing of the transition, leaded to divide the phenomenon into two phases, separated by three time events. First phase is framed within the time of sunrise and the time when the sensible heat flux becomes positive for the first time. The second phase follows ending with the time of the onset of the CBL, time when the turbulence is intensively active and the BL height has reached heights around 200-300m (Angevine et al., 2002).

Three key mechanisms that are identified to affect the transition are shear, buoyancy and strength of inversion. In the early stages of the transition, studies have shown there is a shallow convective layer topped by a shear driven SBL (Beare, 2008). If the wind increases, the SBL depth increases, resulting to a deeper CBL during the morning transition. Thus, morning transition is sensitive to shear (Beare, 2008).

Another important mechanism is buoyancy. The partitioning of the available radiation energy to sensible and latent heat flux, between the atmosphere and the surface, depends on the characteristics of the land surface. The surface is heating after sunrise, providing the atmosphere with energy thus the boundary layer is growing. Therefore, the land surface is involved with the boundary layer growth. As the day continues, buoyancy creates a uniformly mixed boundary layer (Maxwell, Chow, & Kollet, 2007).

Last, number of studies have shown that entrainment, related with weak or strong inversion, have an impact on the formation of the early morning BL (Lapworth, 2006). Morning transition is found to be influenced by surface heating, wind speed and inversion at the top of the boundary layer. Thus, the research question is:

***How the three key processes: shear, partitioning of surface fluxes and strength of inversion affect the dynamics and chemistry during the morning transition?***

Morning transition was investigated based on surface and airborne measurements to obtain a representative picture of the vertical structure of the atmosphere. Surface measurements provided from Cabauw, the meteorological tower of the Netherlands. Airborne data provided from the PEGASOS project (PEGASOS, 2011). During May 2012, PEGASOS campaign took place in the Netherlands, around the Cabauw tower. The project aims to gain better understanding between the atmospheric chemistry and the climate which will allow an assessment of the current policies and better planning of the future ones. A particular and unique feature of the project is the use of a Zeppelin. The aircraft performed measurements of meteorological variables, i.e. potential temperature and took samples of the chemistry, i.e. ozone. The morning transition is a process with rapid dynamics, vertical measurements covering the development of the CBL can provide a better insight of the complex relation between air quality and dynamics, during the morning transition (PEGASOS, 2011).

Observations were processed using a MiXed Layer (MXL) model coupled with a chemistry module (Heerwaarden, 2013). By doing so, we reproduce accurately the diurnal variability of the dynamics and chemistry in a convective boundary layer. At the same time, the chemistry module incorporates important reactions such as the system $O_3$-$NO_x$-VOCs (de Arellano et al., 2011). The model the variables are well-mixed and as a consequence, they are expected to be constant with height (Barbaro, de Arellano, Krol, & Holtslag, 2013).

The MSc thesis is structured as follows. Chapter 2 covers the theoretical background of the BL characteristics, the evolution of the morning transition as well as examining the three key mechanisms that have been mentioned before. Chapter 3 mentions the sources of the airborne and surface observations and explores the model. Chapter 4 analyzes the case study and strategy. The results cover the characterization of the case study and the sensitivity analysis in Chapter 5, followed by a discussion of the results and the model performance in Chapter 6.

# 2. Theoretical background

This chapter outlines the theoretical background of this research. It will discuss the previous knowledge on the morning transition from a stable to a convective boundary layer and the effects on chemistry. Moreover, processes that are involved during the morning transition will be discussed.

## 2.1 Atmospheric boundary layer

The **atmospheric convective boundary layer** (ACBL) is defined as "*the part of the troposphere that is directly influenced by the presence of the earth's surface, and responds to the surface forcings with a timescale of about an hour or less*" (Stull, 1988). One of the main characteristics of the boundary layer is the diurnal variation of temperature near the surface. The diurnal variation of temperature is caused mainly indirectly through the absorption of solar radiation from the ground. So, whether the ground is getting warmer or cooler, responding to available radiation, results to changes in the boundary layer due to transport mechanisms. The main transport mechanism within the boundary layer is the turbulence, characterized by irregular motions called eddies (Stull, 1988).

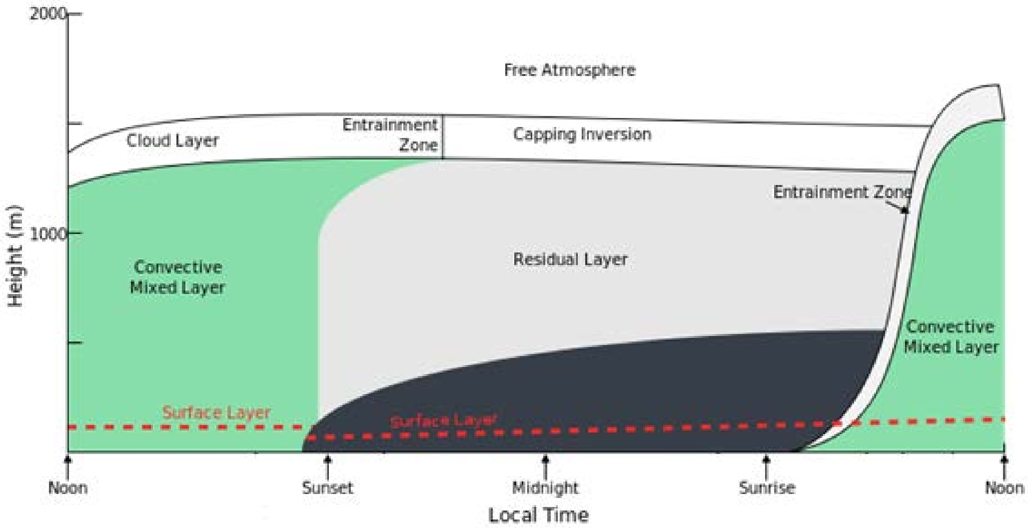


*Figure 1: Boundary layer representation (inspired by Stull, 1988).*

Figure 1 shows the structure of the boundary layer during one whole day. The boundary layer can be separated into three sublayers with different characteristics. The first sublayer is called **surface layer** (5-10% of the total volume) and it is adjacent to the ground(Stull, 1988). This part of the boundary layer is strongly affected by soil and vegetation and moreover is the part where chemical species are emitted or deposited (Heerwaarden, 2013). The next sublayer is called **mixed layer** (35-80% of the total volume) and here exist larger thermals. Last, at the top of the boundary layer there is the **entrainment zone** (10-60% of the total volume) which includes clouds(Stull, 1988).

Due to its importance, often the whole ACBL is called mixed layer (ML). It is a part that characterized by intense vertical mixing. Therefore, variables such as potential temperature and specific humidity stay constant with height (Heerwaarden, 2013). The vertical mixing within the ML is caused due to shear (mechanical turbulence) or buoyancy (convective turbulence). As a result of the influence of the convective turbulence the layer is called **convective boundary layer** (CBL) (Stull, 1988).

Shear favors horizontal motions while buoyancy favors vertical motions. Mechanical turbulence happens when the wind contacts the surface and the fiction between them causes the wind to be sheared and turbulence to be created. So, when shear dominates the wind is connected with variables such as friction velocity ($u^*$) and roughness length ($z_{om}$) which are consequently related to the level of turbulence in the lowest part of the atmosphere (Stull, 1988).

The quota between shear and buoyancy has an important impact on the depth of the mixed layer, the characteristics of the CBL and the turbulence (Pino, De Arellano, & Duynkerke, 2003). Therefore, one of this study's interest is to identify the impact of shear and buoyancy on diurnal variation of dynamics and chemistry, as well as assessing the magnitude of its process's impact.

At the top of the mixed layer, there is a stable layer called **entrainment zone** (EZ) which restraints the rising thermals so, restrains the turbulence and allows entrainment from the free troposphere into the ML. Often, EZ presents to be strong enough to cause temperature inversion thus, the temperature to increase with height. A strong inversion at the top of the boundary layer leads also to trapping of pollutants within the ML (Stull, 1988). Therefore, the presence of strong or weak inversion at the entrainment zone has an impact on the diurnal variation of dynamics and chemistry and this study includes it in its interests for research.

Figure 2 shows the vertical evolution of potential temperature and scalar. The fact that the variables within the CBL are considered constant with height leads to describe it as a bulk layer. Thus, the vertical evolution of a variable can be given as a single mixed-layer value (Heerwaarden, 2013). Mathematical analysis of the prognostic equations will be given in chapter 3:

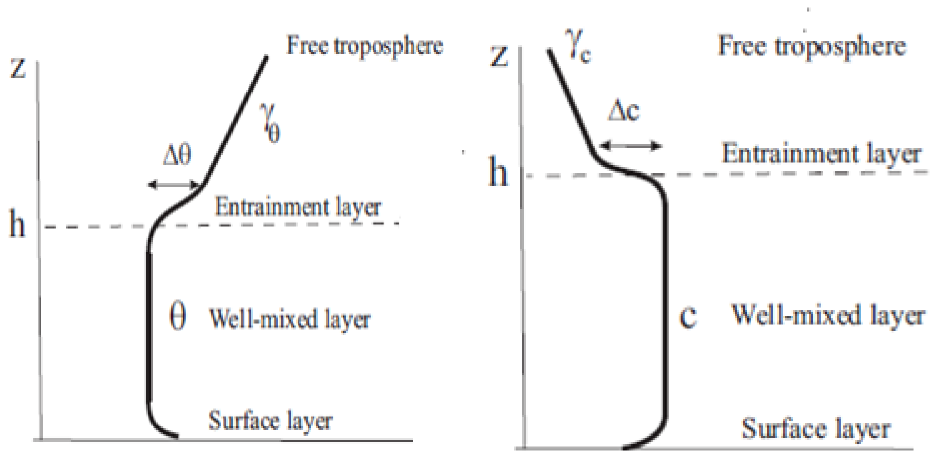


*Figure 2: Conceptual representation of vertical evolution of (left) potential temperature and (right) chemical species within the mixed layer (Heerwaarden, 2013).*

## 2.2 Evolution of the boundary layer: the morning transition

The ABL's structure changes depending on the time of the day (Sokol et al., 2014). During the day there are mostly unstable conditions while during the night there are stratified conditions. The transition periods between these two phases are complex phenomena and they are an ongoing area for research (Beare, 2008). This research's main interest is to study is the process of morning transition from a nocturnal stable (SBL) to a convective boundary layer (CBL) and afterwards to study the dynamics and chemistry during the convective period.

Soon after sunrise, the solar radiation coming from the sun results into changing the sensible heat flux from negative to positive and altering the conditions (i.e. stability) within the boundary layer. The early morning boundary layer is a shallow layer and it increases slowly in the beginning since the nocturnal SBL is a strong layer and its erosion takes time. Later in the morning, the increase of the boundary layer is fastening (Stull, 1988).

Morning transition is an important intermediate phase, setting the initial conditions for the CBL depth and defining the timing that the pollutants start to mix and distribute. Investigating with more details the timing of the transition, leaded to two important phases separated by three time events. First, time event is considered the sunrise. The second time event is the time when the surface sensible heat flux becomes positive for the first time. These two time events form the first phase of morning transition. Last time event is the onset, where the SBL has been eroded, turbulence is intensively active and the BL height has reached higher heights, around 200-300m. This is the second phase of the phenomenon (Angevine et al., 2002). The definition of the three time events leads to a characterization of three different heights during the morning transition (Bange, Spiess, & van den Kroonenberg, 2007).

This research's attempt is to focus on morning transition and enhance our understanding of the lower atmosphere's behavior at that time. It is a phenomenon that determines the initial conditions for the later growth phase. Therefore, gaining knowledge of how parameters that mentioned before, such as friction velocity, surface fluxes and inversion, affect the CBL will help first the prognostics for the CBL's depth and second the timing of the transition will help for studying and modelling the chemistry of the atmosphere (Angevine et al., 2002). Defining the time events of the morning transition is an important aspect of the air quality studies and applications since, it often coincides with the beginning of anthropogenic and biogenic surface emissions (White et al., 2003).

### 2.3 Shear – Impact of Low-Level Jet

Within this lowest part of the atmosphere the horizontal mean wind is affected by the surface friction. In this sublayer, the wind speed is affected by friction drag near the surface and it becomes zero close to the ground. On the other hand the pressure gradient enhances the wind to increase with height (Stull, 1988). As a consequence, the important variable is the friction velocity, associated with the mechanical turbulence which generated or controlled from wind shear near the ground:

$$u_*^2 = \sqrt{\overline{u'w'}^2 + \overline{v'w'}^2} \qquad (i)$$

The height that the wind speed becomes zero is defined as roughness length $z_{om}$ . This variable is determined only by the nature of the surface. Surfaces with higher vegetation and in general more complex terrain, have larger values of roughness length (Stull, 1988).

The effect of shear to BL growth occurs from the equation below:

$$\frac{\partial h}{\partial t} = -\frac{1}{\Delta\theta_{vh}}\left[\left(\overline{w'\theta_v'}\right)_e + 5u_*^3\left(\frac{\theta_v}{gh}\right)\right] + w_s \quad (ii)$$

If there is wind shear at the entrainment zone, equation $(x)$ has a more complex form by adding the jump of the wind components (Heerwaarden, 2013).

During night time, turbulence is mainly produced by the wind shear. Shear affects the depth of the SBL during the night. During morning transition, there is a shallow convective layer topped by a shear driven BL. If the wind increases the SBL depth increases, resulting to a deeper CBL during the morning transition. Thus, morning transition is sensitive to shear. Afterwards, the transition is completed, shear loses the dominant role, and buoyancy governs the evolution of CBL (Beare, 2008).

As it mentioned before, shear is linked with wind. Wind and shear are also linked with the presence of Low-Level Jet (LLJ). Low-level jet is a characteristic of the nocturnal stable boundary layer. It is a thin air layer with wind speed between 10-20ms$^{-1}$ and length up to thousand kilometers and width up to hundred kilometers (Stull, 1988). The wind speed has larger values than the geostrophic wind speed of the free atmosphere above the BL. In some cases such as of inclined surfaces, it is possible the LLJ to appear during daytime (Wang et al., 2007).

As the day proceeds, around sunset, the friction drag decreases resulting to an acceleration of the air. The wind decouples from the surface as the nocturnal stable layer is forming and the wind above the stable layer quickens along the pressure gradient (Wang et al., 2007).

The LLJ starts to appear after sunset but and as the time passes it becomes stronger (Wang et al., 2007). It can be an additional source  of turbulence beside shear near the surface in the stable boundary layer (Stull, 1988). Additionally, it is influenced by roughness elements as it flows over a surface. Thus, surfaces with larger roughness values enhance the LLJ (Wang et al., 2007).

### 2.4 Surface fluxes partitioning – Impact of land use

The partitioning of the available energy to sensible and latent heat flux, between the atmosphere and the surface, depends on the characteristics of the land surface. Soil moisture variability is among the factors that affect the CBL evolution during the morning transition. Previous studies have proved that the level of soil moisture in the surface defines the transmission of heat therefore, the surface fluxes and buoyancy (Maxwell et al., 2007).

Buoyancy is among the driving forces for turbulence in a boundary layer. When buoyancy is the dominating process over shear, the boundary layer is in free convection state (Stull, 1988).

The surface is heating after sunrise, providing the atmosphere with energy thus the boundary layer is growing. The depth of the boundary layer is defined by the potential temperature gradient. Therefore, the land surface is involved with the boundary layer growth. For example, in case of a wet soil (higher soil moisture), there will be lower soil temperatures resulting to less available heat and a lower boundary layer height. Latent heat flux is part of the coupling system. During the morning transition this process is quite noticeable and as the convective boundary layer is formed and the day continues, buoyancy creates a uniformly mixed boundary layer (Maxwell et al., 2007).

Thus, among this research's interests is to investigate the processes that connects the land surface and the atmosphere by altering the partitioning of sensible and latent heat flux keeping the summation of the radiation the same during the morning transition.

### 2.5 Strength of inversion – Impact of upper air conditions

The CBL is closely linked apart from land surface, with the free troposphere (van Heerwaarden, de Arellano, Moene, & Holtslag, 2009). According to Angevine, (2002), apart from the surface fluxes and shear, entrainment is important during the morning transition. Generally, studies have shown the significant impact of the nocturnal radiative processes on the dynamics and chemistry. Among the factors that cause the deepening of the mixed layer is the radiation reduction which reduces the temperature's vertical gradient (Edwards J.M., 2013).

As it mentioned again earlier, during the day sun provides energy and warms the surface through shortwave radiation. Subsequently, the presence of radiation leads to boundary layer growth (Edwards J.M., 2013). However, during the night, there is no solar radiation. So, within the nocturnal boundary layer only longwave radiation originated from the surface can be found which cools the air near the surface (Lapworth, 2006). Cooling is stronger during clear-sky conditions and week winds (Shaw, Doran, & Coulter, 2005). Therefore, during clear nights the cooling effect, affects the temperature profile and reduces the strength of the inversion. As a result, this reduction affects the developing mixed layer until the onset of the CBL, which is the time that the mixed layer has completely replaced the nocturnal stable boundary layer (Shaw et al., 2005). Consequently, the mixed layer is growing more (Edwards J.M., 2013).

This research's focusing on the entrainment zone, attempts to study the impact of strong or weak inversion on the boundary layer growth and its role during the morning transition.

# 3. Observations and Methods

This chapter describes the available data used in this research and explains the characteristics of the measurements sites. The main goal of the PEGASOS campaign is discussed and its relevance with the studied phenomena.

## 3.1. Observations

### 3.1.1 Measurement Site

***CABAUW, Netherlands***

The 213-m meteorological tower CABAUW of Netherlands is located in the central part of the country (51.97$^o$ N, 4.93$^o$ E) (Angevine et al., 2002). The tower is located between big urban areas, Utrecht and Rotterdam, and 1 km northwest of the River Lek. The measuring site is flat and covered with grass extended in all directions(Casso-Torralba et al., 2008). The meteorological tower of Cabauw is operating constantly since 1973. Data provided from the 213-m high tower and from radiosondes that were released from De Bilt. De Bilt is located 25 km northeast of the tower and provided two radiosondes at 06 and 0826h (Tolk et al., 2009).

Measurements for meteorological variables including temperature, humidity and wind have taken at various heights 2, 10, 20, 40, 80, 140 and 200 m (Pino et al., 2012). Surface fluxes, sensible and latent heat flux, were also available (Tolk et al., 2009). Additionally, in-situ measurements for $NO_2$, NO and $O_3$ near the tower were also provided. Typical value for roughness length at Cabauw is 0.1m which is a larger than a typical value for grassland (Beljaars & Bosveld, 1997).

Additionally, Royal Netherlands Meteorological Institute (KNMI) provided measurements for temporal evolution of BL height. The data were retrieved from 1290 MHz wind profile measurements. The height of the CBL can be occurred from a wind profiler measurement using the modified signal-to-noise-ratio detection algorithm (Bianco & Wilczak, 2002).

***PEGASOS campaign***

PEGASOS campaign is a large scale project consisting of twenty six partners from twelve EU countries (http://eu-pegasos.blogspot.nl/, 2013). It aims to gain better understanding between the atmospheric chemistry and the climate which will allow an assessment of the current policies and better planning of the future ones. A particular and unique feature of the project is the use of a Zeppelin. The campaign uses a Zeppelin, equipped, with state-of-art technology. The advantage of using a zeppelin aircraft is that it provides with measurements above and below the convective boundary layer (CBL) within a few minutes. The morning transition is a process with rapid dynamics, vertical measurements covering the development of the CBL can provide a better insight of the phenomenon. The zeppelin reached up to ≈700 m height, resulting in measurements that can be used as a good indication for the processes that take place, during the morning transition, below and above the under development CBL.

For the 27$^{th}$ of May 2012, the Zeppelin provides an extensive set of data that can be used as model input and validation (http://eu-pegasos.blogspot.nl/, 2013).


*Figure 3 : The zeppelin taking measurements next to Cabauw tower at 27th of May, 2012(http://eu-pegasos.blogspot.nl/, 2013).*

## 3.2. Model

In this section we describe the numerical modelling framework used to reproduce the boundary layer dynamics and chemistry, including a description of the model.

### 3.2.1 Mixed layer approach

In this report we use numerical modelling of the CBL dynamics. Hence, a MiXed Layer (MXL) model coupled with a chemistry module (Heerwaarden, 2013). By doing so, we reproduce accurately the diurnal variability of the dynamics and chemistry in a convective boundary layer. At the same time, the chemistry module incorporates important reactions such as the system $O_3$-$NO_x$-VOCs (de Arellano et al., 2011).

The model the variables are well-mixed and as a consequence, they are expected to be constant with height (Barbaro et al., 2013). Therefore, the mixed layer can be compared with a homogeneous reactive box where various chemical reactions take place (Janssen et al., 2012). Above the mixed layer the variables are determined by the conditions of the free atmosphere (FT) which are defined as stably stratified. Thus, the variables are determined by both the conditions in the free troposphere which are represented by the jump of the variable at the "entrainment zone" and the lapse rate of the specific chemical reaction.

Moreover, the model solves the temporal evolution of dynamic variables such as potential temperature (θ), specific humidity (q) and chemical species like ozone ($O_3$). The time integration is from sunrise to sunset, providing diurnal variability(Heerwaarden, 2013). Based on Garratt (1992), the mixed layer temporal evolution of the potential temperature is be defined as(Garratt, 1992) :

$$\frac{\partial < \theta >}{\partial t} = \frac{(\overline{w'\theta'})_s - \left(\overline{w'\theta'}\right)_e}{h} + adv \quad (iii)$$

The first term in the left hand side of the equation describes the temporal evolution of potential temperature. The first term in the right hand side the equation contains the entrainment process which is represented by $\left(\overline{w'S'}\right)_e$ and the surface flux represented by$(\overline{w'S'})_s$. The second term on the right is the advection term associated with mean wind in horizontal direction. Advection affects the turbulence within the boundary layer and can be advection of momentum, heat, moisture or pollutants (Stull, 1988). The equation indicates that any variable within the CBL depends on the vertical flux difference between the surface and the entrainment zone as well as the horizontal advection.

According to the MXL model, the entrainment flux comes as a product of the entrainment velocity $w_e$ (upward direction means positive value) and the potential temperature jump $\Delta\theta_h$ at the inversion:

$$(\overline{w'\theta'})_e = -\Delta\theta_h(\overline{w'\theta'})_e \quad (iv)$$

The subsidence velocity is also:

$$w_s = -w_{ls}h \quad (iii)$$

Equation $(iii)$ includes the large scale vertical velocity ($w_{ls}$) as function of horizontal wind. Moreover, the prognostic equation for the potential temperature jump is:

$$\frac{\partial\Delta\theta}{\partial t} = \frac{\partial\theta_{FT}}{\partial t} - \frac{\partial < \theta >}{\partial t} = \gamma_\theta\left(\frac{\partial h}{\partial t} - w_s\right) - \frac{\partial < \theta >}{\partial t} \quad (v)$$

The forth equation describes the conditions above and below the inversion as well as the free tropospheric lapse rate. The mixed layer jump indicates the separation between the unstable mixed layer and the stable free troposphere (van Stratum et al., 2012). Indicating the temporal evolution of CBL height, the main equations of the MXL model have been covered(Barbaro et al., 2013):

$$\frac{\partial h}{\partial t} = w_e + w_s \quad (vi)$$

Last, an important closure has been assumed:

$$(\overline{w'\theta'})_e = -\beta(\overline{w'\theta'})_s \quad (vii)$$

Equation $(vii)$ describes the relation between the surface and the entrainment flux. The coefficient β in our study is equal to 0.2(C., 2013). The given equations show that the evolution of the variables depends on the surface and the entrainment air from the free troposphere.

Additionally, for the chemical species the variables depend on emissions or deposition fluxes (van Stratum et al., 2012). Combining the chemistry scheme we reproduce the diurnal variability of chemical species such as ozone. The governing equation is:

$$\frac{\partial < C >}{\partial t} = \frac{(\overline{w'C'})_s - \left(\overline{w'C'}\right)_e}{h} + R \quad (viii)$$

Equation $(viii)$ shows that the chemicals varies on time depending on first, emission/deposition processes at the surface $(\overline{w'C'})_s$, second dynamic effects $(\overline{w'C'})_e$ and last chemical reactions/transformations (R) (C., 2013).

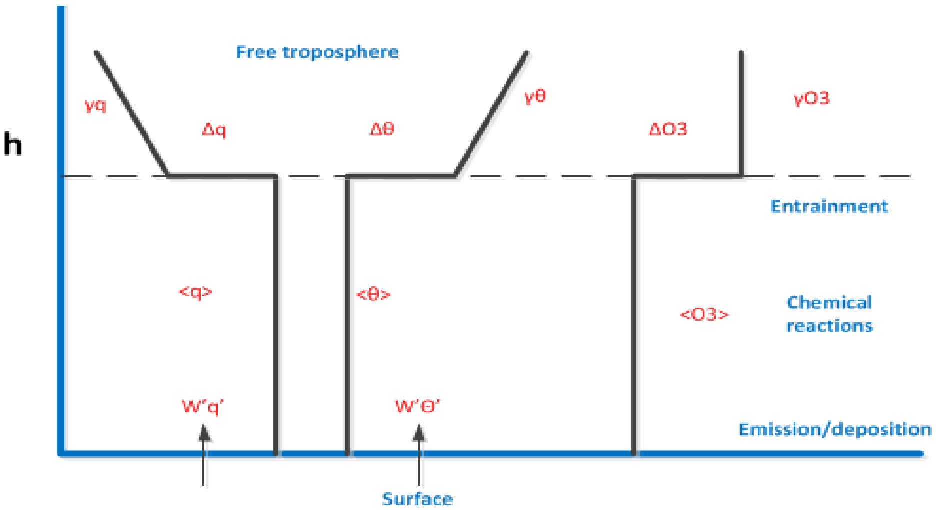


*Figure 4: Conceptual representation of vertical profiles of main components.*

Figure 4 shows that according to the model all the variables are constant with height and covered at the top by an inversion. Moreover, on the right chemical reactions, relevant to the case, are mentioned i.e. emission/deposition and chemical reactions.

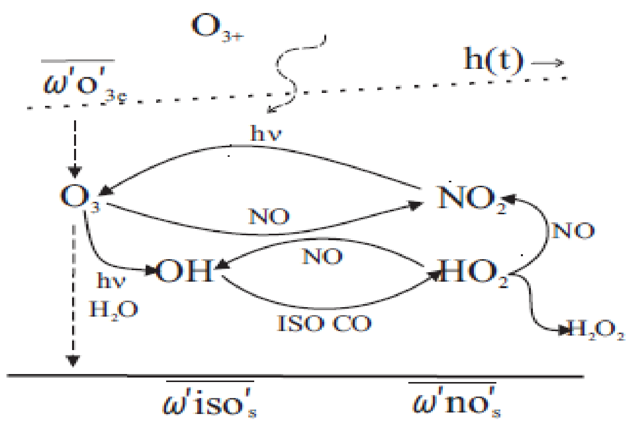


*Figure 5: Conceptual representation of the main reactions that affect the ozone formations and its precursors (C., 2013).*

The chemistry module of the model is based on a system of 27 chemical reactions (see Table 1 APPENDIX I) Two very important reactions are:

$$NO_2 + hv \rightarrow NO + O_3 \quad (R.5)$$

$$NO + O_3 \rightarrow NO_2 + O_2 \quad (R.11)$$

The above reactions represent the photo stationary state and the formation of ozone. The NO emissions originated from the surface and the anthropogenic activities (i.e.

fuel combustion) and affect the ozone and hydroxyl radical (OH) formation (Janssen et al., 2012). There are, also, other important reactions involving isoprene (ISO) and hydroperoxyl radicals ($HO_2$) (C., 2013). Isoprene is one of the most emitted Volatile Organic Compounds (VOC). VOCs have a key role in the photochemistry of the boundary layer. As it shown in figure 5, isoprene can affect the ozone concentration due to isoprene oxidation which depends on nitrogen concentrations (Curci et al., 2009).

The distribution of ozone is related, among various factors, to the surface type, rural or urban area and to the flow processes. These processes can be complex and variant and include low-level jet (LLJ), convective mixing and local circulation (wind shear). During the morning transition, changes at the chemical composition are moving fast. The nocturnal stable layer increases at volume and transports ozone and nitrogen compounds vertically (Lee et al., 2003). The effect of the nocturnal conditions on the diurnal processes it represented by the model by a basic chemical mechanism (Heerwaarden, 2013).

# 4. Case study and Research design

In this chapter a description of the case study and the research design of this report will be discussed. Also, the synoptic conditions of the selected day will be explained.

### 4.1 Synoptic

We select a case based on the intensive observational period of the Zeppelin. The data are provided for 27th of May 2012 at Cabauw, Netherlands (51.97$^o$ N, 4.93$^o$ E). Sunrise and sunset took place at 05:28 and 21:42 respectively. In addition, The MXL model requires input data, the initial and boundary conditions for meteorological variables, surface layer and chemicals.

In the previous chapter the various sources of data were discussed in detailed. They include the meteorological tower of Cabauw, RIVM chemical data from the Cabauw area, the Zeppelin platform from the PEGASOS project and the KNMI. Additionally, two radiosondes were released that day from De Bilt. The tower provided extensive 10-minutes measurements and the zeppelin platform provided with extremely detailed measurements for every 100 milliseconds from 0509 to 0900UTC covering physical dynamics and chemistry. The zeppelin departed at 04:30UTC next to Cabauw, conducting several vertical profiles during sunrise(http://eu-pegasos.blogspot.nl/, 2013). Afterwards, it moved towards Rotterdam for refueling and returned to continue with the height profiles near the tower. Its journey for that day stopped at 11:00UTC when it landed in Rotterdam. Having multiple and various sources of measurements for the selected day, created a unique dataset and leaded for the 27th of May to be considered ideal for the purposes of this study(2011).

Another reason that this day was selected is because of the fair weather conditions. Figure 6 represents a synoptic situation of the observational period.

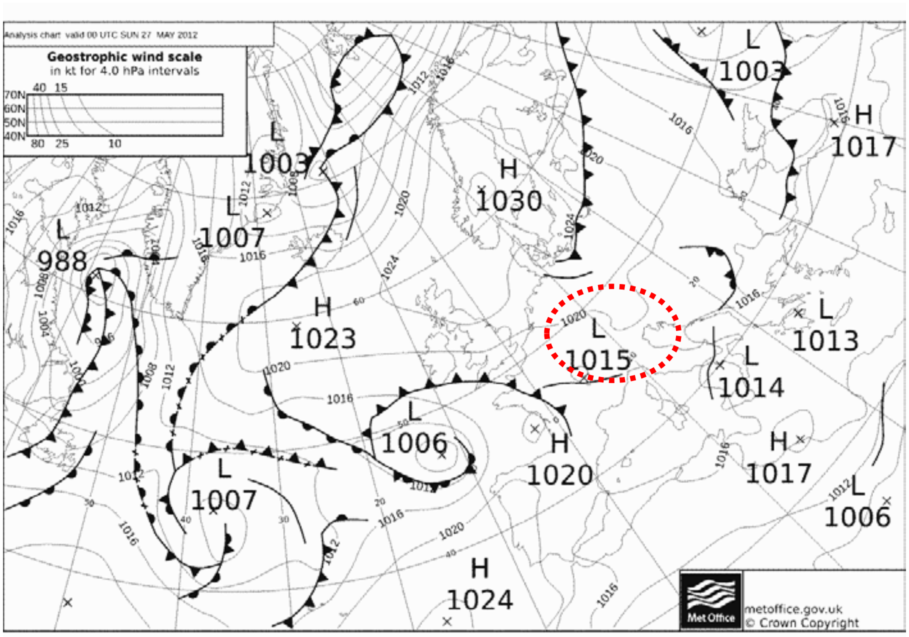


*Figure 6 : Synoptic conditions at 27th of May in Europe ("Wetterzentrale,").*

High pressure is linked with fair weather. In this case the weather was sunny and the sky was cloud-free. The isobars around the area are not close to each other therefore the winds are not strong during the day. Wind speed started with almost $6ms^{-1}$ in the morning and after 14:00 it became stronger reaching $9ms^{-1}$. The wind direction was dominantly eastern. In the morning it was south-east and after 1400UTC it turned to north-east.

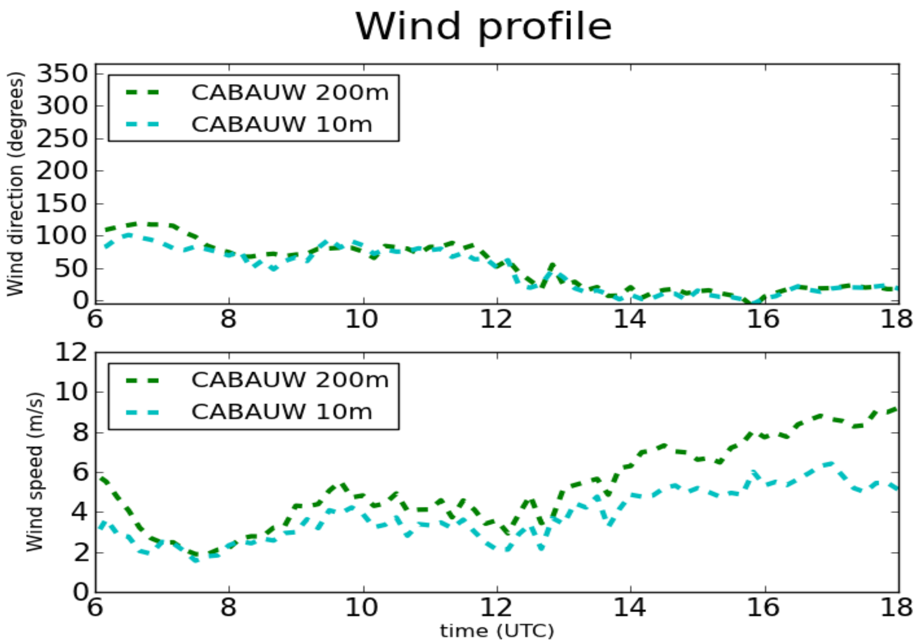


*Figure 7: Wind direction and speed at 27th of May in Cabauw.*

The initial height profiles at early in the morning showed $NO_X$ concentrations originated from the east affecting the measurements (Ankie Piters, personal communication). However, after 06UTC there was no other indication for advection of chemicals from the east.

Figure 8 represents with a conceptual way the location of the Zeppelin, the Cabauw meteorological tower and boundary layer height for three critical time events for the morning transition.

This study will add to the existing research of morning transition by investigating the impact of the involved processes such as entrainment or surface fluxes, on the diurnal evolution of various variables related to atmospheric dynamics and air quality. Equally important is the fact that it will provide a validation of the PEGASOS campaign for the Netherlands case and provide an initial complete idea of the variables that were measured and the measurements themselves.

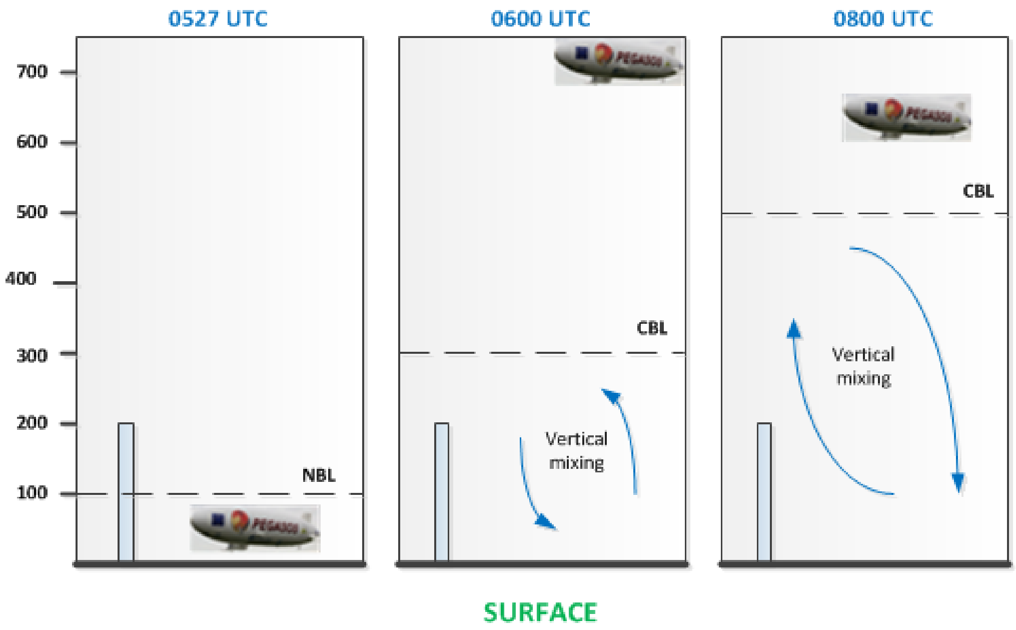


*Figure 8: Conceptual representation of the experiment for three critical timings. The location of the Zeppelin, the BL height and the depth of the vertical mixing are indicated.*

### 4.2 Case study

The aims of this research are twofold: first is understand and analyze a complete characterization of the case by analyzing the diurnal evolution of main physical and chemical variables. Second is to perform a sensitivity analysis to investigate the impact of specific processes that are crucial for the evolution of the variables during the morning transition. These processes are the shear production, the buoyancy and the entrainment. Additionally, the magnitude of the impact of these processes will be evaluated. To fulfill these goals of this research the steps that were taken were:

1) Reproduce with the mixed-layer model for the 27$^{th}$ of May 2012
2) Analysis of the case
    - Validation of the Cabauw, PEGASOS, RIVM and KNMI data
    - Results for the diurnal temporal and vertical evolution of physical and chemical variables.

It is expected from this part of the process to identify the importance of physics as well as chemistry.

3) Sensitivity analysis
    - Low-Level Jet: Examine the impact of shear production on physical components such as friction velocity, boundary layer height and wind speed and on chemical species such as ozone, nitrogen dioxide and nitric oxide through changes on roughness length.
    - Precipitation/soil moisture: Examine the impact of buoyancy on the surface fluxes, sensible and latent heat flux, boundary layer height and chemical species such as ozone, nitrogen dioxide and nitric oxide through changes on sensible heat flux.

- Strength inversion: Examine the impact of entrainment on the boundary layer height and chemical species such as ozone, nitrogen dioxide and nitric oxide through changes on the potential temperature jump.

It is expected to identify how these processes affect the variables (see Table 3), i.e. either suppressing or enhancing and moreover how much each of them affect. During the analysis, each process will be associated with physical conditions such cloudless or not nights etc. in order to provide complete case scenarios for the sensitivity analysis.

By using the MXL model, we will simulate the single day that was selected, the 27th of May 2012. The measurements will provide the initial and boundary conditions. For example, the radiosonde provided the values for the initial potential temperature, lapse rate and potential temperature jump. This set of options will set up the control case for all the further investigations or changes. Therefore, each run will have a similar initial and boundary conditions except for the changes mentioned above in the steps (see table 1). For the control case and for each other run, several graphs are plotted. For the control case, both temporal and vertical evolution of the variables was plotted. For the sensitivity analysis, only the temporal evolution was studied.

*Table 1: Overview of the numerical experiments and processes that used.*

| Name of the run | Control case | Shear production | Buoyancy | Entrainment |
|---|---|---|---|---|
| model | ✓ | | | |
| model 08UTC | ✓ | | | |
| model 09UTC | ✓ | | | |
| CBW 200m | ✓ | | | |
| CBW 10m | ✓ | | | |
| CBW 08UTC | ✓ | | | |
| CBW 09UTC | ✓ | | | |
| ZEP 0830-09UTC | ✓ | | | |
| ZEP 08-0830UTC | ✓ | | | |
| ZEP 400-600m | ✓ | | | |
| ZEP 60-100m | ✓ | | | |
| KNMI | ✓ | | | |
| RIVM | ✓ | | | |
| control case | | ✓ | | |
| forest | | ✓ | | |
| bush | | ✓ | | |
| bare soil | | ✓ | | |
| 2SH | | | ✓ | |
| SH/2 | | | ✓ | |
| dth/3 | | | | ✓ |
| 3dth | | | | ✓ |

Table 2 below represents the variables that were studied for each part of the process. The dynamics investigated by their association with the surface fluxes (sensible and latent heat flux), temperature, humidity and boundary layer height.

These variables affect the development of the boundary layer and the mixing of chemicals. Thus, an adequate representation between them shows how they are related.
For chemistry, apart from the dynamics, chemical reactions are important. Ozone attracts the special focus of this study, even though the chemicals that are involved into its cycle of production/destruction will be represented. So, the graphs include the temporal evolution of NO, $NO_2$ and isoprene. The chemical scheme of the model is also indicates these chemicals as important for determination of ozone mixing ratio.

*Table 2: Variables that were studied/plotted during the reproduction of the case and the sensitivity analysis.*

| Name of variable | Control case | Shear production | Surface fluxes partitioning | Strength of inversion |
|---|---|---|---|---|
| **Sensible heat flux** | ✓ | | ✓ | |
| **Latent heat flux** | ✓ | | ✓ | |
| **Potential temperature** | ✓ | | | |
| **Specific humidity** | ✓ | | | |
| **Wind speed** | ✓ | ✓ | | |
| **Wind direction** | ✓ | | | |
| **Friction velocity** | | ✓ | | |
| **BL height** | ✓ | ✓ | ✓ | ✓ |
| **$O_3$** | ✓ | ✓ | ✓ | ✓ |
| **NO** | ✓ | ✓ | ✓ | ✓ |
| **$NO_2$** | ✓ | ✓ | ✓ | ✓ |
| **ISO** | ✓ | | | |

The choice of the variables that are represented above in table 3, was based on first the nature of the PEGASOS campaign, since it emphasizes on photochemistry such as ozone. Second, based on the possibilities on the CLASS version of the MXL model can provide. Last, based on which processes and variables were involved in each step. For example, in case of buoyancy, we decided to investigate the surface fluxes but not the physical dynamics. The boundary layer height and ozone concentration remained important for the research in every step of the project.

# 5. Results

Here we present the results from the validation of the MXL model against the Cabauw and Zeppelin datasets. We evaluate the ability of the MXL model to reproduce the case study that under investigation. Our objective is to demonstrate the feedbacks between dynamics and chemistry. The results show that the MXL model was able to reproduce the surface measurements adequately. Second, we present the results from the sensitivity analysis. This part is divided into four sections showing the impact of three key processes – shear, partitioning of surface fluxes and strength of inversion – to the morning transition - between the stable to the convective boundary layer. The results cover the influence of these processes on the dynamics and the chemistry. Finally, in the last part, the three processes will be individually to evaluate which of these has the largest effect on the morning transition.

## 5.1 Validation of the case study

### 5.1.1 Surface fluxes and temporal profiles

Here we verify the capability of the MXL model to reproduce first, the Cabauw measurements of potential temperature, specific humidity, boundary layer height and surface heat fluxes. Moreover, to reproduce the Zeppelin measurements of the potential temperature, specific humidity and ozone.
The solar radiation at the ground, is separated into sensible (SH) and latent (LE) heat flux according to equation$(ix)$:

$$Q_* = LE + SH \quad (ix)$$

$Q_*$ is the net radiation. To be precise, there is also the ground flux (G) which is minus the summation of sensible and latent heat flux. In our case we do not consider a value for the ground flux. Furthermore, the Bowen ration is defined as:

$$\beta = \frac{SH}{LE} \quad (x)$$

Table 3 below indicates the initial and boundary conditions estimated from the Cabauw tower and used as input in the MXL model. The diurnal cycle of the prescribed surface heat fluxes (SH and LE) is prescribed at 11hours ($T_{diurnal}$) to match with the observations. By so doing, we ensure that the well-mixed period of the CBL is well reproduced by the MXL model (Janssen et al., 2012).

*Table 3: Initial conditions for surface fluxes as prescribed to the MXL model.*

| Variable | Value |
|---|---|
| **Surface heat flux** $\overline{w'\theta'}_s$ $[K\,ms^{-1}]$ | 0.045 |
| **Latent heat flux** $\overline{w'q'}_s$ $[gkg^{-1}ms^{-1}]$ | 0.12 |
| **Bower Ratio** | 0.375 |

For the reproduction of the case, the initial conditions for the surface fluxes used in our MXL simulations were obtained from the Cabauw dataset. The maximum sensible heat flux (at 12 UTC) is approximately 54Wm$^{-2}$ and the maximum latent heat flux amounts approximately 362 Wm$^{-2}$ at the same time. Around Cabauw area the latent heat flux is larger than the sensible heat flux (LE>>SH) due to water availability and type of vegetation (Tolk et al., 2009).
The CBL conditions are influenced by the available levels of energy coming from the surface (Porporato, 2009). Moreover, the daytime value for the Bower Ratio is 0.375 characteristic of wet conditions. Bowen ratio values between 0.3-0.4 may also indicate occurrence of precipitation before the measuring time (de Arellano et al., 2004).
The CBL evolution during daytime is strongly influenced by the nocturnal SBL from the previous night. As mentioned previously in the theory chapter, due to turbulent mixing the SBL is completely eroded during the morning transition, becoming a CBL. The potential temperature and specific humidity fluxes follow a sinusoidal temporal evolution as part of a convective boundary layer (Casso-Torralba et al., 2008).
Boundary layer data from KNMI and the radiosonde at 06UTC that was released from De Bilt provided the initial conditions for the potential temperature, specific humidity and boundary-layer height. These conditions are shown in Table 4.

*Table 4: Initial conditions for heat and specific humidity values as prescribed in the MXL model.*

| Variable | Value |
|---|---|
| **Initial BL height $h_0$ [m]** | 300 |
| **Initial potential temperature <θ> [K]** | 293.5 |
| **Initial potential temperature jump Δθ [K]** | 1.45 |
| **Initial potential temperature lapse rate $\gamma_\theta$ [Km$^{-1}$]** | 0.00236 |
| **Initial advection of heat $\theta_{adv}$ [Ks$^{-1}$]** | 0.000123 |
| **Initial specific humidity <q> [gkg$^{-1}$]** | 6.5 |
| **Initial specific humidity jump Δq [gkg$^{-1}$]** | -0.94 |
| **Initial specific humidity lapse rate $\gamma_q$ [gkg$^{-1}$m$^{-1}$]** | -0.00058 |

Figure 9 presents the temporal evolution of the CBL height (h), surface fluxes (SH and LE), potential temperature (θ) and specific humidity (q). The plots show a satisfactory agreement between the MXL model output and the measurements and that the MXL model is able to represent the (thermo)dynamics of the BL during the convective period.
First, the boundary layer height shows to be in general agreement with the MXL model output. However, the data have fluctuations specially after 12 UTC. Previous studies have shown that the fluctuations at the boudary layer height might are related with a meausrement error (Steeneveld, de Wiel, & Holtslag, 2007). The MXL model also captures well the time where the boundary layer becomes well mixed, i.e. when the temperature observations at all heights become homogeneous. Specially the Cabauw data from height 200m are in best agreement with the model. Specific

humidity is a variable that can indicate the different feedbacks from the surface and the entrainment fluxes (van Heerwaarden, de Arellano, Gounou, Guichard, & Couvreux, 2010). Figure 9 shows that the model output agrees with the measurements. The small differences observed after 09 UTC are due to entrainment of drier air from aloft or due to horizontal advection.

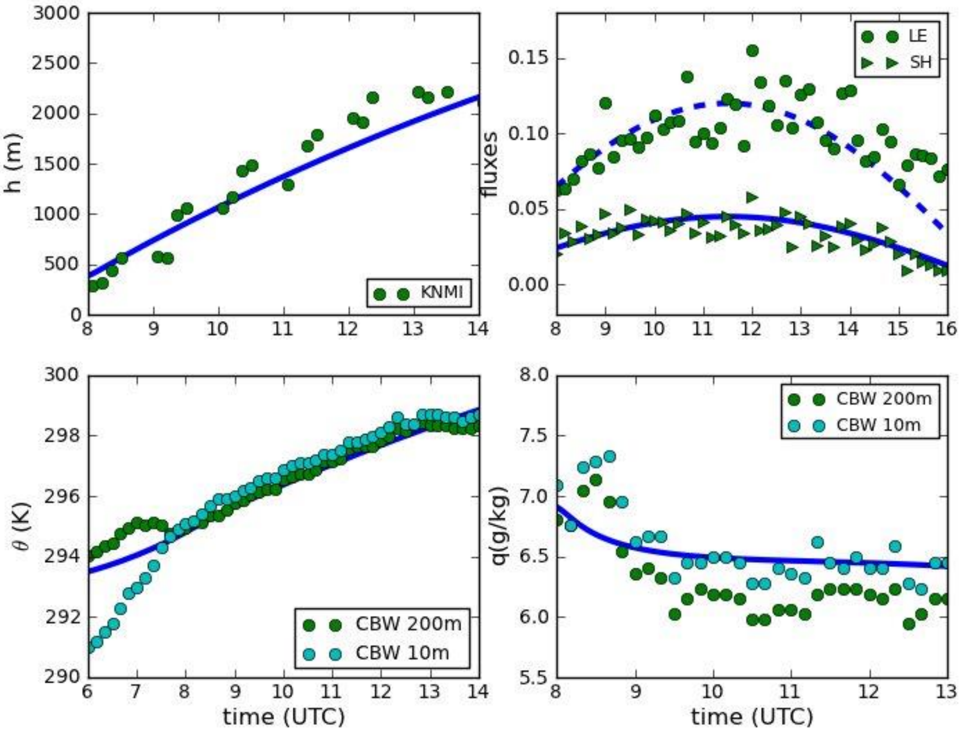


*Figure 9: Diurnal evolution of (top left) boundary layer height, (top right) surface heat fluxes, (bottom left) potential temperature and (bottom right) specific humidity. Solid and dashed lines represent the model output and the symbols the observations.*

Regarding the PEGASOS results, we splited the data into two parts, first when the Zeppelin was near the surface (around 20-90 m) and second when the Zeppelin was at 400-600 m to compare them with the Cabauw data from the surface and from 200m height. Figure 10 shows the temporal evolution of the potential temperature combining the tower data from Cabauw, the airborne data from the Zeppelin and the MXL model output during the morning transition (06-09 UTC). We observe that the Zeppelin data follow the general tendency of the tower data biased by about 1K compared to the observations and the MXL results. Despite that, the Zeppelin is able to capture the temperature differences before the boundary layer became well mixed (until around 08 UTC). We especulate here that this is may be due to a measuring error or innacurate calibration of the temperature sensors.

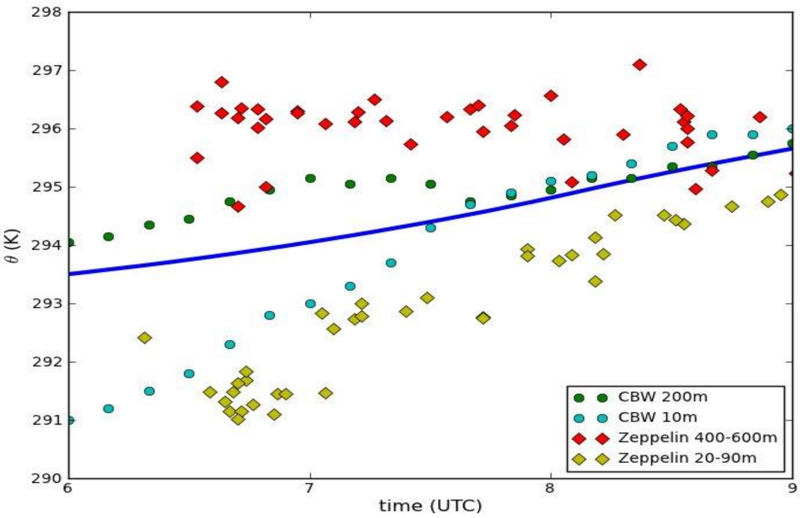


*Figure 10: Diurnal evolution of potential temperature. The solid line represents the MXL model, the circular symbols the tower data and rhombus symbols are the airborne data.*

### 5.1.2 Chemical species

Completing the temporal evolution of variables, four chemicals were reproduced, NO-$NO_2$-$O_3$-VOCs, due to their importance in air chemistry and to their data availability. The dynamics are indicated through the surface emissions/depositions and the entrainment of air from the free troposphere and the chemistry is indicated by the net of reactions for production or loss between them.

Nitrous oxide (NO), nitric dioxide ($NO_2$) and ozone ($O_3$) are connected through a photochemical reaction. Isoprene (ISO) concentrations are linked with NO and $NO_2$ transformation of $O_3$, as triggering factors between them (Heerwaarden, 2013).

The initial concentrations for the chemicals were retrieved from the RIVM surface dataset and the emission fluxes were prescribed based on literature (Table 5).

*Table 5: Initial mixing rations in BL and FA and surface emission fluxes of the chemical species as prescribed to the MXL model.*

| Variable | Value |
|---|---|
| **<NO> [ppb]** | 0.705 |
| **ΔNO [ppb]** | 0.1 |
| **γNO [ppbm$^{-1}$]** | 0 |
| $(\overline{w'NO'})$ **[ppb m s$^{-1}$]** | 8.14 $10^{-3}$ |
| | |
| **<$NO_2$> [ppb]** | 6.19 |
| **Δ $NO_2$ [ppb]** | 0.1 |
| **γ $NO_2$ [ppbm$^{-1}$]** | 0 |
| $(\overline{w'NO_2'})$ **[ppb m s$^{-1}$]** | 1.06 $10^{-4}$ |
| | |
| **<$O_3$> [ppb]** | 26.77 |
| **Δ $O_3$ [ppb]** | 23 |

| | |
|---|---|
| **γ $O_3$ [ppbm$^{-1}$]** | 0.02 |
| $\left(\overline{w'O_3'}\right)$ **[ppb m s$^{-1}$]** | -0.06 |

| | |
|---|---|
| **<ISO> [ppb]** | 0.03 |
| **ΔISO [ppb]** | 0.02 |
| **γISO [ppbm$^{-1}$]** | 0 |
| $\left(\overline{w'ISO'}\right)$ **[ppb m s$^{-1}$]** | 0.0038 |

Based on literature, measurements for the ozone deposition fluxes over a grassland showed a mean value of -0.063 ppb m s$^{-1}$ (Bassin, Calanca, Weidinger, Gerosa, & Fuhrer, 2004). Additionally, Williams and Fehsenfeld investigated NO and $NO_2$ emission fluxes over various types of surfaces and indicated that grassland has high emissions comparing i.e. with a coastal environment or a forest. The NO emissions flux is 10.0 ng N m$^{-2}$ s$^{-1}$ (8.14 10$^{-3}$ ppb m s$^{-1}$) and the $NO_2$ emissions flux is 0.20 ng N m$^{-2}$ s$^{-1}$ (1.06 10$^{-4}$ ppb m s$^{-1}$) (Williams & Fehsenfeld, 1991). The concentrations of isoprene are small and the value for the isoprene emissions flux was fitted to the observations.

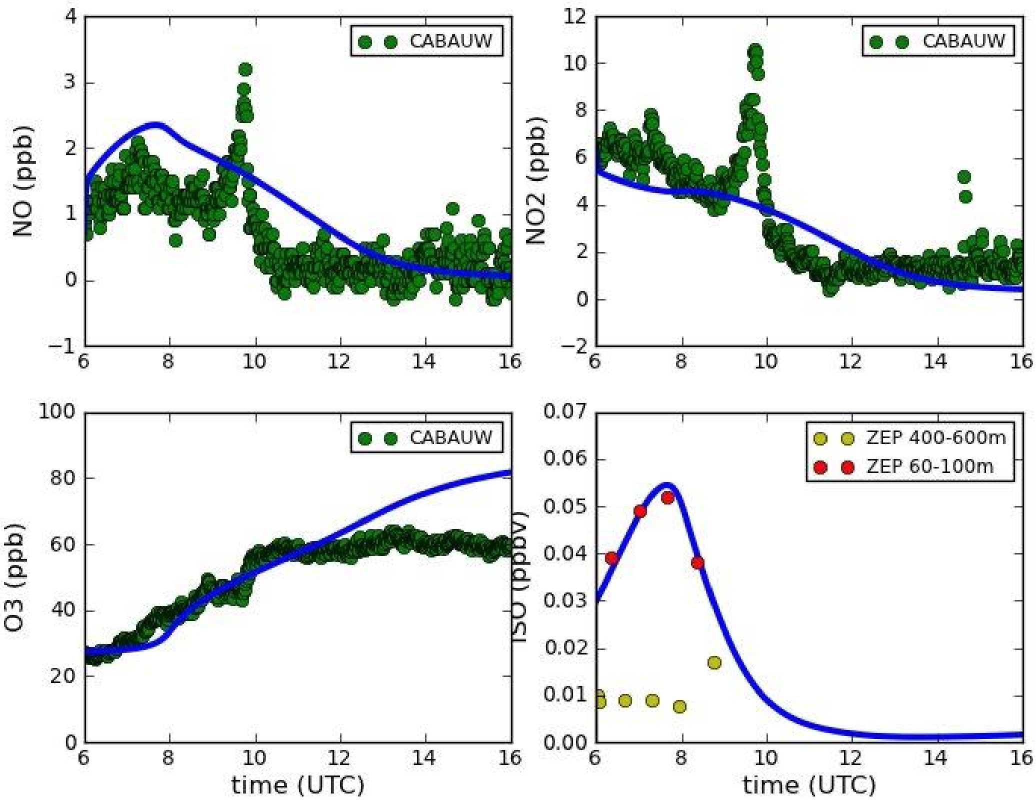


*Figure 11: Temporal evolution of the concentrations of (top left) NO,(top right) $NO_2$,(bottom left) $O_3$ and (bottom right) ISO. The solid lines are the model output, the green circular symbols are the RIVM data from Cabauw area and the yellow/red circular symbols are the Zeppelin data.*

Figure 11 shows the temporal evolution of chemical species compared with the MXL model. Generally, the diurnal trends of the chemicals are in agreement with the measurements, what indicates that the MXL model accurately captures processes such as entrainment, emission and chemistry and their impact on the temporal evolution of the chemicals. Due to the photo-stationary state, the NO and $NO_2$ decrease from 06-10 UTC and later they remain rather constant while the ozone increases rapidly at the same time and remains rather steady after 10UTC.

$NO_x$ results show an early-morning increase because the boundary layer is still shallow and after that the decrease until 10UTC where they remain approximately stabilized (de Arellano, van den Dries, & Pino, 2009). Comparison with initial $NO_x$ results from RIVM $NO_2$ sondes, showed the presence of advection from the east exactly before 10UTC. This is coincides with the visible high peak at the plots for $NO_X$. The advection from the east carries away higher concentrations of $NO_X$ from the industrial area of Nijmegen (Ankie, 2013).

From 06-10 UTC, ozone increases rapidly. This increase is related to entrainment of ozone from the free troposphere as it is indicated from the value of the mixing ratio difference between the mixed-layer and the free troposphere ($\Delta O_3$) and the fast early-morning growth of the boundary layer. After 10UTC the ozone concentration tends to stabilize around 60 ppb and the influence from chemical reactions is probably more dominant than entrainment (van Stratum et al., 2012). However, at the end of the day the model with the observations closes with a ~20 ppb difference between each other. The model keeps increasing whereas the observations show stabilization.

Last, for isoprene the PEGASOS data were split into two parts. First, when the Zeppelin was near the surface and second when it was at around 400-600 m. The model output agrees adequately with the observations for the surface. From 06-08 UTC, there is deviation between the two parts of the observations. After that the data seem to converge, indicating that the boundary layer becomes well mixed. The early-morning increase of the mixing ratio is followed by almost constant values after 10 UTC.

### 5.1.3 Vertical profiles

We combine the vertical profiles of potential temperature (θ), specific humidity (q) and ozone ($O_3$) with the Cabauw meteorological tower data in order to compare the airborne and surface measurements within a range of heights from the lowest part of the CBL until ~700 m. Figure 12 shows the vertical evolution of potential temperature and specific humidity. It includes the MXL model output, the Cabauw data from 2-200 m and the Zeppelin data ranging from 20-600 m. The black solid line for the model output represents the hourly average value for 08 UTC and the black solid line the hourly average value for 09 UTC. The Cabauw data are instantaneous values for the different measuring heights of the tower from 2 to 200 m. Last, the Zeppelin data cover two descendants of the aircraft that happened between 08 and 09 UTC. Specifically, the red dashed line includes observations between 08-0820 UTC and the yellow dashed line between 0829-0859 UTC.

In general, a well-mixed BL has higher moisture content and lower potential temperature from the free troposphere (Tolk et al., 2009). From the two plots, we see that the surface measurements of Cabauw are in agreement with the model output. As it was see previously with the temporal evolution of potential temperature and

specific humidity, the model is able to reproduce adequately the measurements from 08 UTC since is it indicated as the time that boundary layer is becoming well-mixed. Therefore, figure 12 is another plot proving the capability of the MXL model.
Concerning the PEGASOS data, the red dashed line shows better results for the potential temperature. The data show the vertical structure of the mixed layer and provide a quantification of the potential temperature lapse rate. The model and the Zeppelin measurements are in good agreement concerning the lapse rate evolution which indicates that the chosen initial value for the model was good.

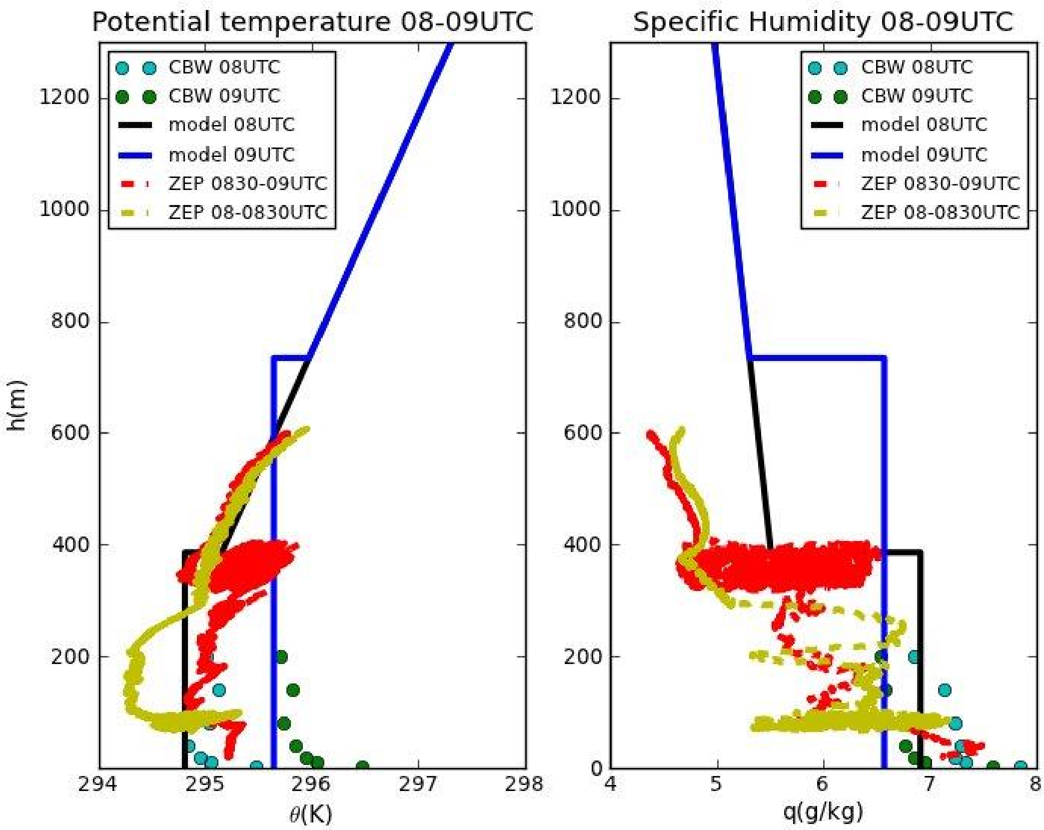


*Figure 12: Comparison between the (blue/black solid lines) model output, (circles) Cabauw tower data and (yellow/red dashed lines) Zeppelin data for the vertical evolution of (right) potential temperature and (left) specific humidity between 08-09UTC.*

For the specific humidity, comparing the Cabauw tower measurements with the model we found that they are in good agreement. The Zeppelin measurements seem to follow the general tendency even though there are differences. They do not present a well-mixed layer as the potential temperature data do. However, the inversion jump Δq is in agreement with model and decreases from ~6.5 to ~4.5 $gkg^{-1}$. Although biased, the lapse rate seems also to be relatively well reproduced.
Just like the vertical profiles of potential temperature and specific humidity, the vertical evolution of ozone shows good agreement between the model output and Cabauw tower data. This version of the MXL model does not provide the inversion jump and the lapse rate for the chemical species; therefore they are not plotted in figure13. However, comparing the Zeppelin data with the model output there is a strong deviation, around 30 ppb, between them. So, Zeppelin overestimates the

ozone concentrations. This may be caused due to the specific choice of the measuring instrument or the measuring method.

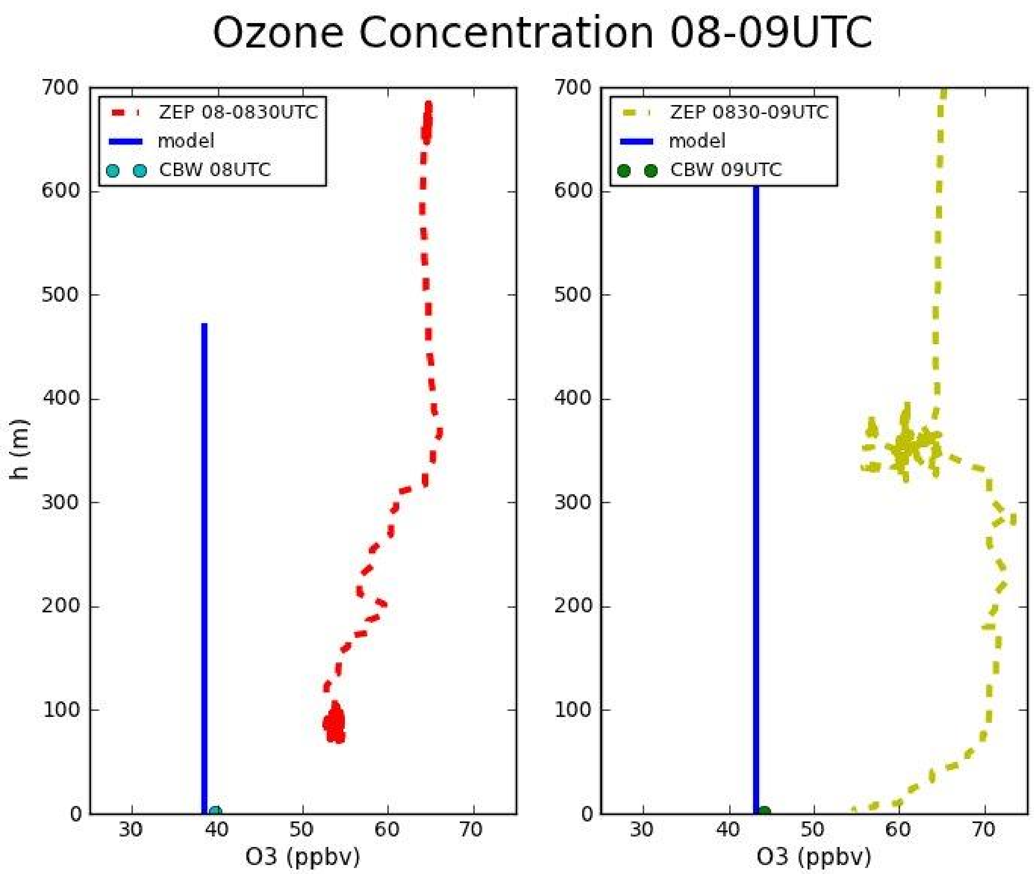


*Figure 13: Comparison between the (blue solid line) model output, (green/cyan circles) Cabauw tower data and (red/yellow dashed lines) Zeppelin data for the vertical evolution of ozone (left) between 08-0830UTC and (right) between 0830-09UTC.*

## 5.2 Sensitivity analysis

### 5.2.1 Shear effect – the role of wind

According to previous studies investigating the morning transition, shear is the most important mechanism in governing the time of the onset of the CBL (Angevine et al., 2002; Beare, 2008).

As have been explained in Chapter 2, within this lowest part of the atmosphere the wind speed is affected by friction drag near the surface and it becomes zero close to the ground (Stull, 1988). The important variable is the friction velocity, associated with the mechanical turbulence which originated or controlled from wind shear near the ground. The height that the wind speed becomes zero is defined as roughness length $z_{om}$. This variable is determined only by the nature of the surface. Surfaces with higher vegetation and in general more complex terrain, have larger values of roughness length (Stull, 1988).

Wind and shear are also linked with the presence of Low-Level Jet (LLJ). As the day proceeds, around sunset, the friction drag decreases resulting to an acceleration of

the air. The wind decouples from the surface as the nocturnal stable layer is forming and the wind above the stable layer quickens along the pressure gradient (Wang et al., 2007). It is influenced by roughness elements as it flows over a surface. Thus, surfaces with larger roughness values enhance the LLJ (Wang et al., 2007).
So, the four experimental cases were designed to study the role of shear and wind in the morning transition and how shear affects the dynamics and the chemicals to evolve, by varying the initial values of the roughness length. The experiments are planned to investigate four distinct surface types. The control case refers to the area around Cabauw, therefore is grassland. Apart from this, there is a forest, bush and bare soil case. The values for the variables for the four experimental cases are mentioned below in Table 6.

*Table 6: Initial conditions for the roughness length during the sensitivity analysis.*

| Experimental case | Property-Value |
|---|---|
| **Control case** | $z_{om}$=0.1m |
| **Forest case** | $z_{om}$=0.8m |
| **Bush case** | $z_{om}$=0.2m |
| **Bare soil case** | $z_{om}$=0.005m |

The initial values for the roughness length were based on literature for the vertical and horizontal extrapolation of the wind climate statistics (Stull & Ahrens, 2000) & (Technical University of Denmark, 2012). The roughness length value for the control case is 0.1m and it is determined from previous studies for the specific terrain surface around Cabauw (VanUlden & Wieringa, 1996).

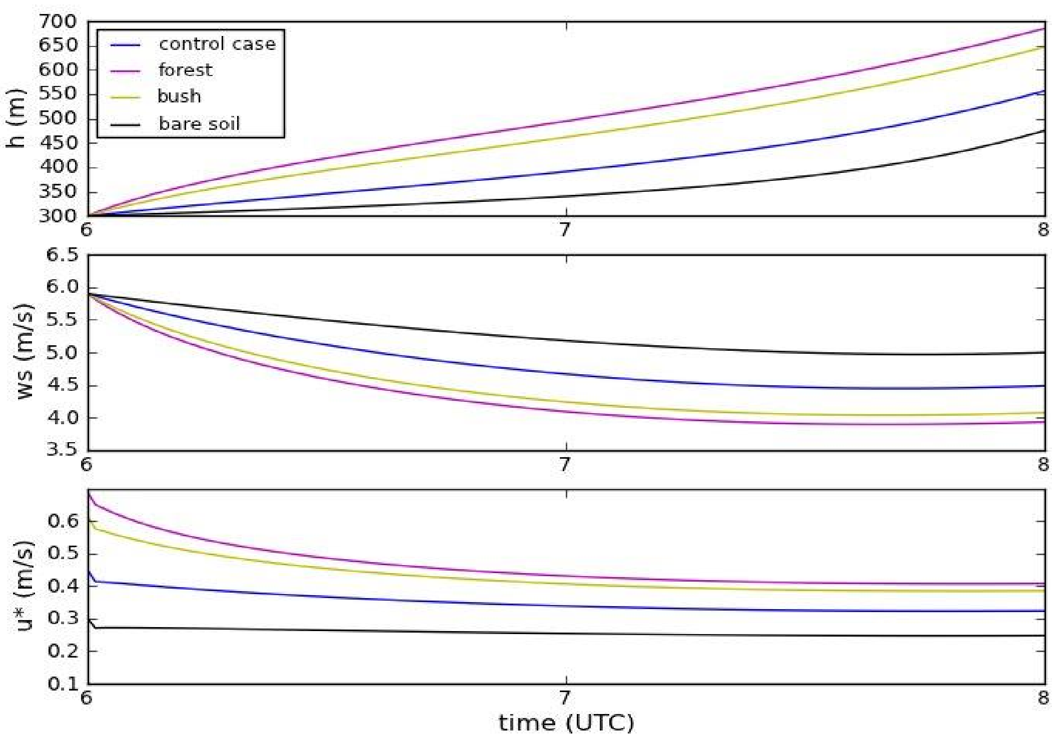


*Figure 14: Temporal evolution of (a) the boundary layer height, (b) wind speed and (c) friction velocity during the morning transition. The colours are indicated in the legend.*

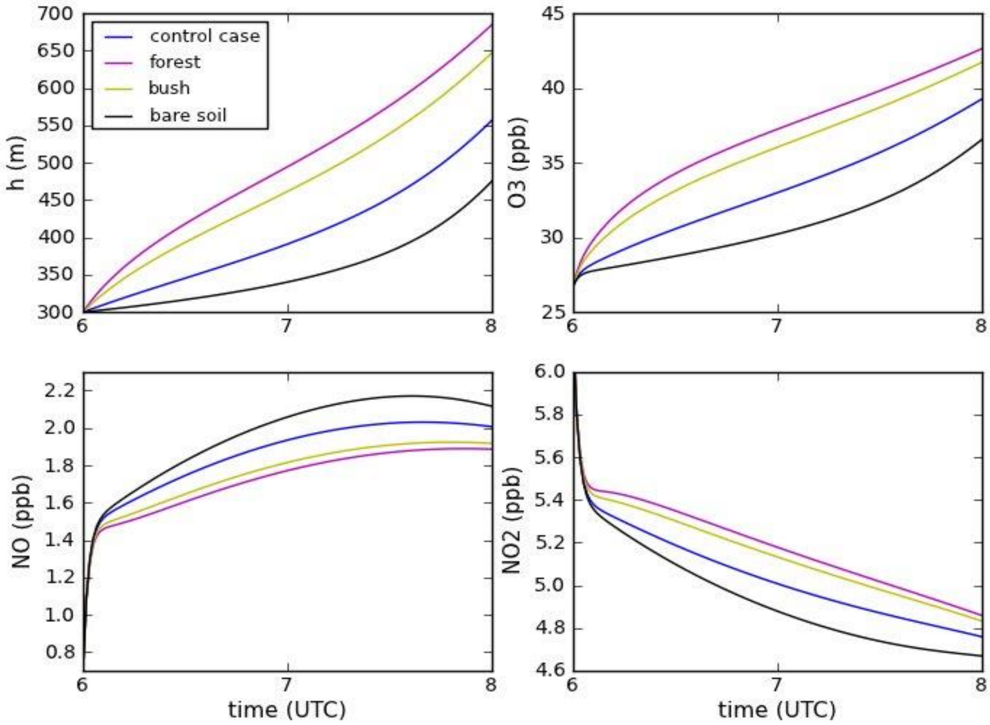


*Figure 15: Temporal evolution of (a) boundary layer height, (b) ozone, (c) NO and (d) $NO_2$ during the morning transition. The colours are defined in the legend.*

In this part we focus on the effect of shear only near the surface. As mentioned before, wind shear can either enhance the boundary layer growth or delay it. The relation between shear and boundary-layer height is given by equation $(ii)$. We see that higher values for roughness length (forest case) results in higher values of friction velocity and consequently higher values for boundary layer height. At around 08 UTC we observe a CBL height of around, 700 m. Thus, there is an important difference in CBL depth of around 200m between the control and the forest cases.

In figure 15 we show the influence of the boundary layer height on NO, $NO_2$ and ozone for the four experimental cases. First result is that the higher $O_3$ concentrations are linked with the higher boundary layer. During the morning transition, there are rapid changes to the chemicals since the boundary layer increases rapidly (Lee et al., 2003). Also, ozone-richer air that is engulfed from the free troposphere into the boundary layer, enhancing the pre-existing concentrations and the high values of roughness length are related with the high $O_3$ concentrations (van Stratum et al., 2012). Generally, determining the boundary layer height it is important for the chemical species since it indicates the mixing-depth of any input or loss of the chemicals either at the surface or at the top (van Stratum et al., 2012).

NO and $NO_2$ follow the general tendencies. Before NO decreases, it slightly increases during the morning transition. We observe the lowest concentrations for the forest case. Moreover, $NO_2$ keeps decreasing since the early morning and the forest case give the highest values. $NO_2$ values for the free troposphere are very low, therefore entrainment has an opposite effect comparing to $O_3$, which leads to a decrease of $NO_2$ (van Stratum et al., 2012).

An additional investigation for the relation between the roughness length and the chemical concentrations is shown in Figure 16. $O_3$, NO and $NO_2$ average concentrations between 06-07UTC and 07-08UTC were calculated and plotted compared with the roughness length values for each experimental case

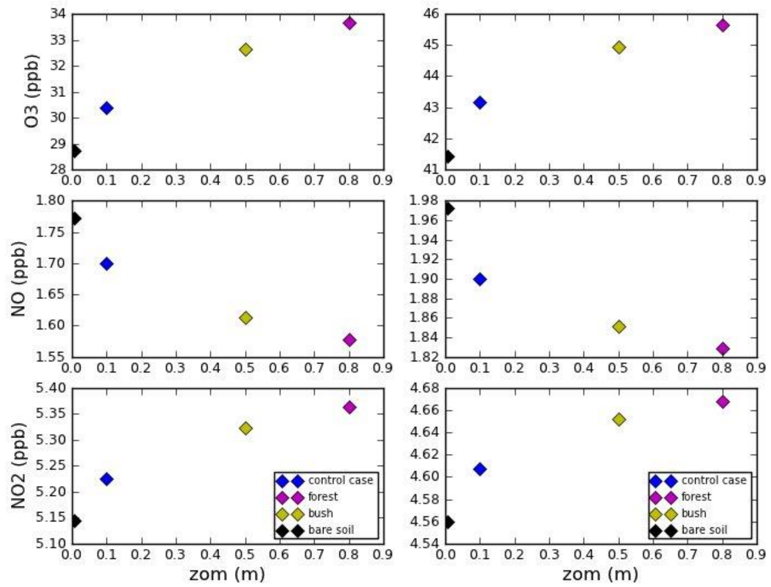


*Figure 16: Chemical concentrations in comparison with roughness length for the four experimental cases.*

Again, we observe that the forest case presents the largest roughness length for both time periods. So, the largest value of roughness length is related to the largest value of $O_3$ and $NO_2$ concentrations and with the lowest NO concentration.
As the time pass, the boundary layer will increase monotonically with time for all the experimental cases, leading to higher height for the case of forest. In the afternoon both the boundary layer height and ozone concentrations will be maximum. NO and $NO_2$ concentrations, except the early morning peak of NO between 06-09 UTC, will decrease throughout the day without big deviation between the experimental and the control cases. This temporal evolution is considered logic due mainly to the photochemical reactions between them and partly due to the homogeneity of the mixed layer later (Lee et al., 2003).

### 5.2.2 Surface fluxes partitioning – the role of surface fluxes

During night, the boundary layer is shallow and stable. However, after sunrise, the surface heats leading to a convective boundary layer. Therefore, the surface fluxes affect the early stage of the morning transition period. So, different surface conditions, e.g. dry or wet, or whether precipitation occurred during the previous night, affect differently the morning transition development. For example, in case of a surface with large water content , such as the area around Cabauw, or if precipitation occurred during the previous night, there will be the need for more sensible heat flux or more time, to overpass the negative values of the night time. Therefore, there will be a time delay or a height limitation to the boundary layer growth during the early morning hours (Stull, 1988).

To investigate the role of surface fluxes on the morning transition and how it affects the evolution of the CBL and the chemicals within it three experimental cases were designed changing the initial values of the sensible and latent heat flux (Table 7).

*Table 7: Initial conditions for the sensible and latent heat flux during the sensitivity analysis.*

| Experimental case | Property-Value | |
|---|---|---|
| **Control case (SH<<LE)** | SH=54 W m$^{-2}$ | LE=362 W m$^{-2}$ |
| **High SH flux (SH>>LE)** | SH=379.2 W m$^{-2}$ | LE=331.43 W m$^{-2}$ |
| **Equal fluxes (SH≈LE)** | SH=204 W m$^{-2}$ | LE=208.49 W m$^{-2}$ |

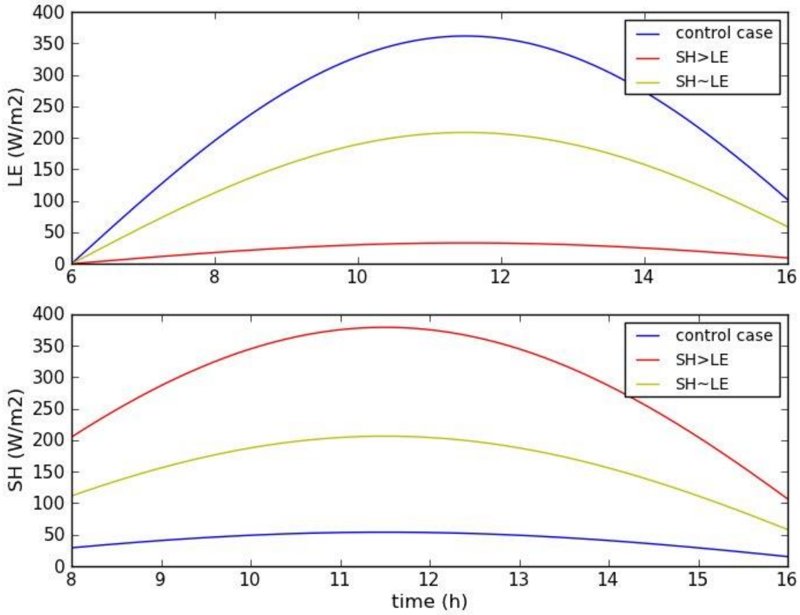


*Figure 17: Temporal evolution of sensible and latent heat flux for the three experimental cases.*

Figure 17 above shows the temporal evolution of sensible and latent heat flux for the three experimental cases. At noon the control case has the highest values for latent heat flux, reaching at approximately ~362 W m$^{-2}$. Sensible heat flux reaches also at noon high values at ~54 W m$^{-2}$.

Figure 18 represents the temporal evolution of the boundary layer height, $O_3$, NO and $NO_2$. The experimental case with larger sensible heat flux results in higher boundary layers, approximately 1000m. The way that the available energy is partitioned between sensible and latent heat flux controls the boundary layer growth. Consequently, the ozone reaches higher concentrations if the sensible heat flux increases due to higher boundary layer height. Different land conditions affect differently the surface fluxes and the chemicals. NO is increasing during the morning transition and the $NO_2$ decreasing. Dry soil leads to larger amount of sensible heat fluxes, resulting to a faster boundary-layer growth and enhancement of the chemicals On the other hand, wet soil results to a weak boundary-layer growth and favours the chemical reactions, which play a more important role for the temporal evolution of them (Janssen et al., 2012).

In the afternoon, the boundary layer will increase monotonically with time for all the experimental cases, reaching very high values for the case of high sensible heat flux. Therefore, ozone concentrations will be also quite high. NO and $NO_2$ concentrations will decrease throughout the. However, altering the surface fluxes values leads to visible deviation between the control and the experimental cases. It is considered logic due to the much larger amount of heat that is introduced from the ground in case of bigger sensible heat flux. Additionally, bigger values of sensible heat flux are linked with weak inversion and promotion of dry-air entrainment from the free troposphere (van Heerwaarden et al., 2009).

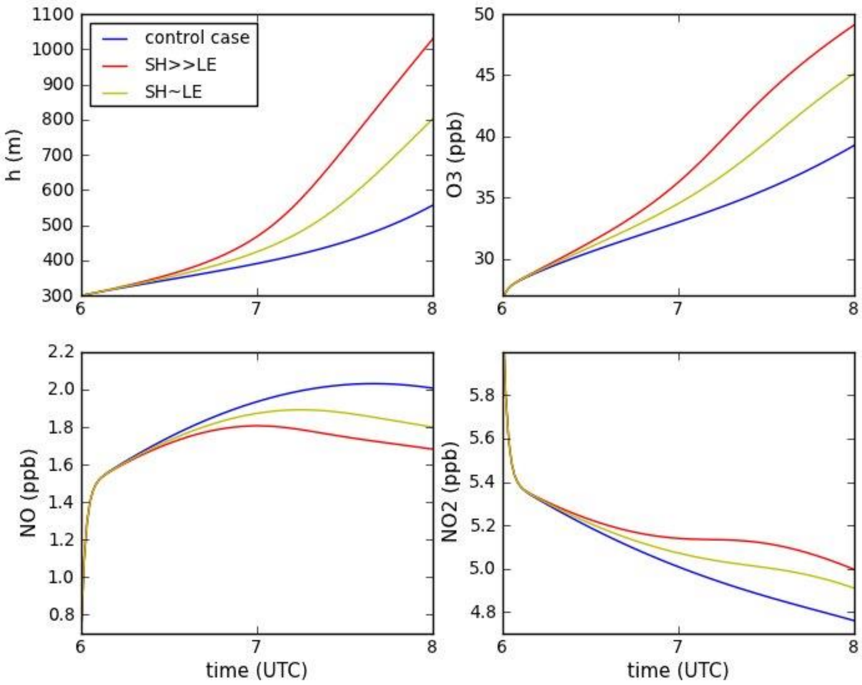


*Figure 18: Temporal evolution of the (top left) boundary layer height, (top right) ozone, (bottom left) NO and (bottom right) $NO_2$ for the (blue) control case, (red) high sensible heat flux, (yellow) equal sensible and latent heat flux.*

### 5.2.3 Strength of inversion – the role of upper air conditions

Another factor involved to morning transition is entrainment. An early start of the transition has been associated with strong nocturnal inversion (White et al., 2002). During the night, due to longwave radiation emission at the surface, we observe lower temperatures in the near-surface atmosphere. So, surface cooling can affect the strength of the nocturnal inversion through its impact on temperature (White et al., 2002). Moreover, during nights with clear skies and little or no cloud cover, stronger nocturnal inversion can be observed (Stull, 1988).

We, therefore, investigate the role of upper air conditions during the previous night and how these affect the evolution of dynamics and chemistry. To do so, three experiments were designed by changing the initial values of the potential temperature jump (Table 8).

*Table 8: Initial conditions for the potential temperature jump during the sensitivity analysis.*

| Experimental case | Property-Value |
|---|---|
| **Control case** | Δθ=1.45 K |
| **Bigger jump (3Δθ)** | Δθ=4.35 K |
| **Smaller jump (Δθ/3)** | Δθ=0.48 K |

Figure 19 shows the temporal evolution of the boundary layer height, $O_3$, NO and $NO_2$ during the three experimental cases. The boundary layer growth presents a fast and drastic increase for the case with weak inversion in the top. Before 07 UTC the CBL is already over 400 m reaching at 08 UTC approximately 750 m. This shows that morning transition happens earlier in case of a weak inversion. As a consequence, ozone concentrations reach also fast higher concentrations at approximately 45 ppb at 08 UTC.

In case of strong inversion (blue line), the boundary layer delays its growth and ozone concentration delays its increase. As the inversion becomes stronger, the effect of buoyancy at the inversion area, restrains the turbulence production, therefore the boundary layer growth (Kosovic & Curry, 2000).

During the afternoon, in case of strong inversion, there is strong deviation between this and the other two. Even though, the control case and the weak inversion will have close values for the boundary layer height, the strong inversion results to significantly lower heights and lower ozone concentrations, indicating the influence of inversion on dynamics and chemistry.

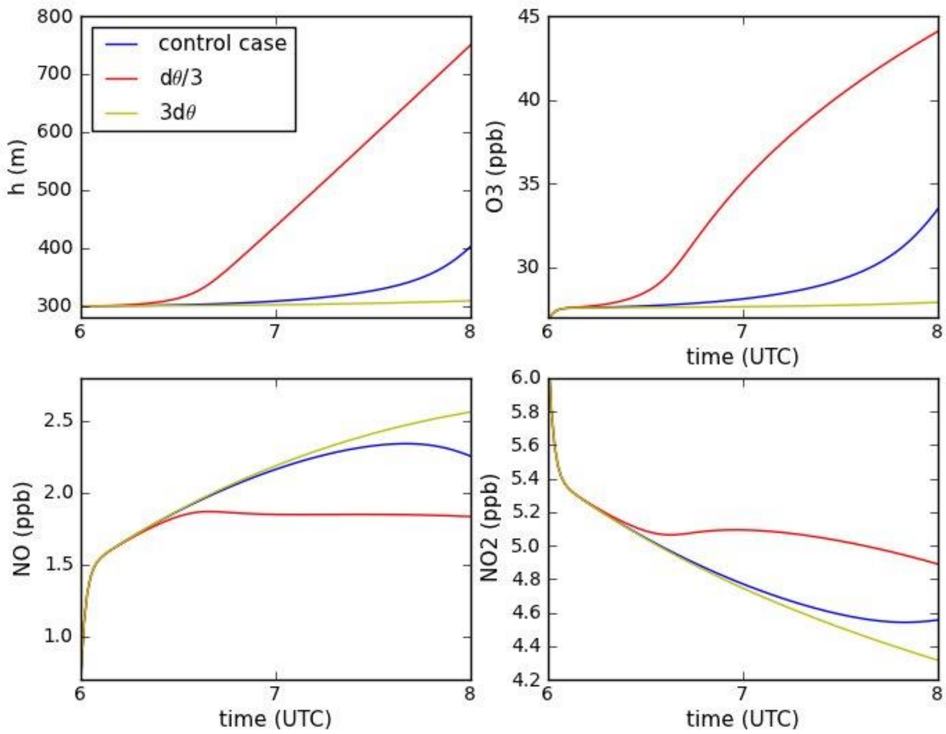


*Figure 19: Temporal evolution of the (top left) boundary layer height, (top right) ozone, (bottom left) NO and (bottom right) $NO_2$ for the (blue) control case, (red) weak inversion, (yellow) strong inversion.*

# 6. Discussion

In the preceding sections, we have shown the temporal evolution of the CBL dynamics and chemistry during the morning transition in Cabauw, the Netherlands on 27th of May 2012. The characterization of the case study has been done by examining the temporal and vertical evolution of the various meteorological variables and chemical species. Three mechanisms are found to be closely related to morning transition. The first mechanism is shear which is associated with the impact of wind on our simulations. The second mechanism is the partitioning of surface fluxes which is due to different land conditions. The third mechanism is the strength of inversion layer at the top of the CBL and how the upper air conditions affect the temporal evolution of the dynamics and chemistry within the CBL. Considering these three aspects we have separated our research in two parts (1) model validation and (2) sensitivity analysis. By doing so, we answer two questions, respectively: (1) was the model able to reproduce the case study, and (2) which of these three mechanisms has the largest impact?

The first part of this research was to demonstrate the ability of the MiXed Layer (MXL) model to properly simulate the surface fluxes, thermodynamics and chemistry of the boundary layer for a selected day (27th May 2012). By using the MXL model coupled to the chemistry module we were able to reproduce the $O_3$, NO and $NO_2$ concentrations throughout the day. Moreover, the model was able to represent the temporal evolution of potential temperature, surface fluxes, specific humidity and boundary layer height for the convective period. The diurnal cycle of $O_3$-NO-$NO_2$-ISO is adequately represented despite the lack of data on the emission/deposition of NO, $NO_2$ and $O_3$. To circumvent that, the fluxes were prescribed based on literature studies for similar conditions at grasslands. Therefore, since chemistry depends on the location of the observations, the discrepancies can be explained by the uncertainty in the fluxes. The isoprene fluxes were prescribed to mimic the Cabauw data and not on literature, since the concentrations were exceptionally small.
The chemistry dataset was provided by RIVM surface observations. For most of the chemicals, e.g. NO and $NO_2$, we observed some discrepancies for the early morning, until the formation of the well-mixed boundary layer. However, the diurnal evolution of $O_3$-NO-$NO_2$-ISO has been represented adequately, which is more important for this study, than exact concentrations values, since the study tries to show the integration of chemistry and dynamics.
The novelty of this research lies also on the Zeppelin data originated from the PEGASOS campaign. The observations provided us with information of the atmospheric vertical structure (temperature, humidity, $O_3$). The MXL model results showed a good agreement with the surface observations from Cabauw. The main discrepancies happened for the zeppelin. Whereas the potential temperature profile showed a well-mixed structure the vertical profile of the specific humidity was not clear (Figure 12). Both variables represented the inversion at the top of the boundary layer and potential temperature lapse rate was in agreement with the model output, corroborating our choice of initial condition. The ozone vertical profile was plotted showing a strong deviation from the model output and surface data (Figure 13). The

special way that the Zeppelin performed the measurements, sampling the air and measuring meteorological variables within a range of heights during the morning transition, leaded to a sensitivity analysis that will examine the mechanisms that affect the transition. Moreover, previous studies focusing on the phenomenon identified shear, partitioning of surface fluxes and strength of inversion among the most influential factors, therefore the sensitivity analysis investigated them.

According to previous studies, shear is the most important mechanism in governing the time of the onset of the CBL (Angevine et al., 2002; Beare, 2008). The control case (grassland around Cabauw) have been compared with three experimental cases, covering three types of surface, forest, bush and bare soil. Altering values for the roughness length ($z_{om}$) we alter the friction velocity, affecting the boundary layer growth. The friction velocity and the boundary layer height are connected by equation $(ii)$ which shows that high values for friction velocity result to enhancement of the boundary layer growth. So, by increased shear, we noticed an enhancement of the growth of the boundary layer and an increase/decrease on the evolution of the concentration of the chemicals. The importance of shear as an influential factor to the evolution of dynamics and chemical concentrations during the early morning is in agreement with the results of Beare (2008).
The partitioning between the surface fluxes, which means different surface conditions, e.g. dry or wet, or whether precipitation occurred during the previous night, affects differently the morning transition development. The results of the sensitivity analysis show that the process with the most positive overall impact on the boundary layer growth and chemicals evolution is the increasing of the sensible heat flux (SH). The physical process is the following: by increasing the sensible heat flux more energy is available to enhance the boundary layer growth and the ozone concentrations (since higher O3 concentrations are observed on the free troposphere).
The third sensitivity analysis discussed entrainment. The weak inversion at the top of the boundary layer allows the CBL to grow faster. Also, for chemistry, this means that the vertical mixing of $O_3$-NO-$NO_2$ occurred earlier. The strength of the nocturnal inversion has a visible effect on the dynamics and chemistry as seen from the results (Figure 19). However, the strongest impact occurred from altering the surface fluxes and not from the strength of the inversion (Figure 18).
These interactions between dynamics, chemistry and the three processes give important insights on the morning transition issue. The transition processes are rapid and complex and set the initial conditions for the CBL depth and the timing of the chemicals start to mix with the air masses.

More research needs to be done on how each parameter will develop and what is the impact on dynamics and chemistry. Further improvement could include an extension of this analysis to more days or other areas with different land conditions. Also, vertical profiles of ozone and potential temperature would give a better representation of the morning transition effect on dynamics and chemistry. Lastly, a quantification of the interdependence of the mechanisms would give a more complete view of the phenomenon, since it provides insight on how the phenomena feedback on each other.

# 7. Conclusions

In this study we used data collected from radiosondes, wind profiles, surface and airborne observations during the 27th of May 2012, at Cabauw, the Netherlands aiming to simulate and describe the morning transition effect on dynamics and chemistry during the PEGASOS campaign. The sensitivity analysis focus on the impact of the three mechanisms involved to the transition, shear, surface fluxes partitioning and strength of inversion, aiming to understanding and describing their influence.

The results show that the model was able to simulate rather adequately the temporal evolution of the dynamics and chemistry, showing their complex relation. The vertical profiles including the Zeppelin data showed deviations from the model output especially in the case of ozone.

The impact of shear and as a consequence of wind showed that influence the evolution of ozone and boundary layer the most for the case of high values of roughness length. However, the strongest impact appeared from altering the surface fluxes, promoting the sensible heat flux, therefore enhancing the boundary layer growth and ozone. The results from the impact of upper air conditions showed that the strength of inversion has a direct and drastic effect on the temporal evolution of dynamics and chemistry, setting the timing of the CBL formation and the chemical mixing, earlier. However, morning transition is affected by a combination of the above mentioned processes, therefore to fully understand the phenomenon, further investigation is needed.

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# APPENDIX I: Reactions in the chemical scheme of the MXLCH model

*Table 1: Reactions in the chemical scheme of the MXLCH model. Adopted from Vilà-Guerau de Arellano & Van Heerwaarden (2012).*

| Number | Reaction | | |
|---|---|---|---|
| R1 | $O_3 + h\nu$ | $\rightarrow$ | $O(1D) + (O_2)$ |
| R2 | $O(1D) + H_2O$ | $\rightarrow$ | $OH + OH$ |
| R3 | $O(1D) + N_2$ | $\rightarrow$ | $O_3$ |
| R4 | $O(1D) + O_2$ | $\rightarrow$ | $O_3$ |
| R5 | $NO_2 + h\nu$ | $\rightarrow$ | $NO + O_3$ |
| R6 | $CH_2O + h\nu$ | $\rightarrow$ | $HO_2$ |
| R7 | $OH + CO$ | $\rightarrow$ | $HO_2 + CO_2$ |
| R8 | $OH + CH_4$ | $\rightarrow$ | $CH_3O_2$ |
| R9 | $OH + ISO$ | $\rightarrow$ | $RO_2$ |
| R10 | $OH + MVK$ | $\rightarrow$ | $HO_2 + CH_2O$ |
| R11 | $OH + HO_2$ | $\rightarrow$ | $(H_2O) + (O_2)$ |
| R12 | $OH + H_2O_2$ | $\rightarrow$ | $HO_2 + (H_2O)$ |
| R13 | $HO_2 + NO$ | $\rightarrow$ | $OH + NO_2$ |
| R14 | $CH_3O_2 + NO$ | $\rightarrow$ | $HO_2 + NO_2 + CH_2O$ |
| R15 | $RO_2 + NO$ | $\rightarrow$ | $HO_2 + NO_2 + CH_2O + MVK$ |
| R16 | $OH + CH_2O$ | $\rightarrow$ | $HO_2 + CO + (H_2O)$ |
| R17 | $HO_2 + HO_2$ | $\rightarrow$ | $H_2O_2$ |
| R18 | $CH_3O_2 + HO_2$ | $\rightarrow$ | PRODUCT |
| R19 | $RO_2 + HO_2$ | $\rightarrow$ | nOH + PRODUCT |
| R20 | $OH + NO_2$ | $\rightarrow$ | $HNO_3$ |
| R21 | $NO + O_3$ | $\rightarrow$ | $NO_2 + (O_2)$ |
| R22 | $NO + NO_3$ | $\rightarrow$ | $2NO_2$ |
| R23 | $NO_2 + O_3$ | $\rightarrow$ | $NO_3 + O_2$ |
| R24 | $NO_2 + NO_3 + M$ | $\rightarrow$ | $N_2O_5 + M$ |
| R25 | $N_2O_5 + M$ | $\rightarrow$ | $NO_3 + NO_2 + M$ |
| R26 | $N_2O_5 + H_2O$ | $\rightarrow$ | $2HNO_3$ |
| R27 | $N_2O_5 + 2H_2O$ | $\rightarrow$ | $2HNO_3 + H_2O$ |