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Impact of HARMONIE high-resolution meteorological forecasts on the air quality simulations of LOTOS-EUROS

Jieying Ding

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Impact of HARMONIE high-resolution meteorological forecasts on the air quality simulations of LOTOS-EUROS

Author: Jieying Ding

Supervisors:

Dr. Henk Eskes (KNMI)

Dr. Ujjwal Kumar (KNMI)

Prof. Maarten Krol (WUR)

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Koninklijk Nederlands Meteorologisch Instituut
Ministerie van Infrastructuur en Milieu

Abstract

In this study, we couple the LOTOS-EUROS model with high resolution meteorological forecasts from the Harmonie model to study the impact on air quality in comparison to simulations driven by ECMWF meteorological forecasts. The main purpose of this study is to compare the differences in meteorological variables between the ECMWF and Harmonie model, and analyse the impact of these differences on the air quality simulations of LOTOS-EUROS. A comparison of the meteorological variables shows that the capability of these two models to simulate the surface temperature, relative humidity and wind speed is similar. Because of the high resolution of Harmonie, there are clear differences in meteorological variables between ECMWF and Harmonie, especially at the coast, over forest and urban areas. We performed a comparison of model meteorological forecasts with observations in the Netherlands. Apart from the boundary layer height (BLH), it is hard to conclude which meteorological model gives better scores. The boundary layer height simulated by Harmonie is significantly lower than for the ECMWF model. Also independent ceilometer observations show that the Harmonie BLH is too low during clear days in Summer 2012. The differences in the meteorological fields lead to significant changes in the local air pollutant concentrations. The root mean square difference (RMSD) between two simulations is 4ug/m3 for PM10; 20ug/m3 for O3; 6ug/m3 for NO2; 5ug/m3 for NO. However, the daily mean difference of PM10 concentration is small and the mean differences of other air pollutant concentration are not significant. We performed a sensitivity analysis of air quality on surface temperature, relative humidity, surface wind speed and boundary layer height. The effect of a 2K difference of surface temperature is very small. With a 10% increase of relative humidity at all layers, the concentration of PM10 increases and the concentration of secondary inorganic aerosols (SIA) gas precursors decreases. The impact on ozone is not significant. An increase of surface wind speed makes the PM10 concentration decrease over the land and increase over sea and near the coastline. The PM10 concentration decreases when the boundary layer height is increased. In contrast, the O3 concentration sometimes increases when the boundary layer height is increased, especially at night.

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1. Introduction

Air pollutants and their precursors play an important role in the climate system and also for human health. The presence of atmospheric aerosols will alter the solar and terrestrial radiation budget, which in turn affects the climate system (Ramanathan et al., 2001). For shorter time scales, the emission of air pollutants will reduce the visibility and may cause photochemical smog. Many studies show that inhaling fine particulate matter is harmful for human health, and an increased cardiopulmonary and cardiovascular morbidity and mortality could be related to it (Chow et al., 2006; REVIHAAP, 2013). The increasing air pollutant concentrations also affect global climate. For example, aerosols have a direct and indirect radiative forcing because they not only scatter and absorb infrared radiation in the atmosphere, but also alter the formation and precipitation efficiency of clouds (IPCC, 2001).

Atmospheric aerosols (particulate matter) are one of the main pollutants affecting air quality. The sources of aerosols can be divided into two groups: anthropogenic emissions and natural sources. The anthropogenic part is mainly from the combustion of fossil fuel. The improvements in the understanding of the processes of the aerosol formation, and depletion have led to the development of atmospheric chemistry transport models, which are used for air quality prediction (Kukkonen et al., 2012; Manders et al., 2009; Schaap et al., 2004; Stern et al., 2008). The air pollution levels in a specific region are not only determined by the emissions, but also by the meteorological conditions, because these control the natural emissions, transport, chemistry and deposition. In particular the variability on timescales from hours to months is determined for a large part by changes in the weather.

De Meij et al. (2009) studied the meteorological impact on the CHIMERE model over North Italy for two months January and June of 2005 by using input data from two different meteorological models: MM5 and WRF. The difference in PM10 concentration is correlated to the difference in boundary layer heights calculated. The boundary layer height is affected by the latent heat flux, which in turn depends on the shortwave incoming radiation at the surface. The difference in peak value for PM10 is mainly influenced by the cloud liquid water. Appel et al. (2010) also use the MM5 and WRF as meteorological drivers for the community Multiscale Air Quality model V4.7 to simulate air pollution levels in January and August 2006 over the eastern United States. They found that the sulfate aerosol concentration mainly depends on the cloud cover. More cloud cover increases the amount of aqueous-phase (in-cloud) sulfate aerosol and decreases wet deposition, which leads to higher concentration of sulfate aerosol. The nitrate aerosols are influenced by the wind speed and the boundary layer heights. In summer, the difference of surface solar radiation influences the concentration of O_3 in the two simulations. Higher surface solar radiation leads to higher surface temperature and higher concentrations of surface biogenic VOCs.

The LOTOS-EUROS model is used for air quality forecasts in the Netherlands, and contributes to the MACC-II ensemble forecast of air pollution over Europe (www.copernicus-atmosphere.eu). By default, the meteorological driver for LOTOS-EUROS is the ECMWF forecast model. The resolution of the ECMWF forecasts in 2012 was about 16 km, which is relatively low compared to the default LOTOS-EUROS resolution of 7 km. Since the end of 2011, the numerical weather prediction model Harmonie has been providing daily forecasts with a high resolution of 2.5 km at KNMI (Royal Dutch Meteorological Institute). The LOTOS-EUROS has been coupled to this high resolution model. The air pollutant concentration are expected to be different compared to the simulations driven by ECMWF model due to the difference in meteorological parameters between the ECWMF and Harmonie. In this study, we performed simulations with the chemistry transport model LOTOS-EUROS with the two meteorological inputs to simulate air quality from June to August in 2012 (summer) over Europe. Our main objectives are summarized in the following research questions:

- What are the differences in the meteorological fields of ECMWF and Harmonie?
- What are the differences in air pollutant concentrations using the two meteorological drivers?
- How sensitive are air pollutant concentrations to the individual meteorological parameters?

The report is structured as follows. In the next section, we introduce the model LOTOS-EUROS and some detail of the simulation settings. In section 3, we describe the influence of meteorological variables on air quality simulations of LOTOS-EUROS. In section 4 and 5, we introduce the two meteorological models ECMWF and Harmonie, and the observation data used to compare with the model results. The statistical methods used for the comparison will be introduced in section 6. Section 7 and 8 shows the comparisons of the meteorological fields and air pollution concentrations. In section 9, we show the results of the sensitivity analysis of air quality on individual meteorological variables. The report finishes with conclusions and recommendations.

2. Air quality model LOTOS-EUROS

The LOTOS-EUROS model (Sauter et al., 2012) is an integration of two models, LOTOS (Long Term Ozone Simulation) and EUROS (European Operational Smog), which were developed by Netherlands Organization for Applied Scientific Research (TNO) and National Institute for Public Health and the Environment (RIVM) respectively. The LOTOS-EUROS model can simulate the following air quality components: oxidants, primary aerosols, secondary inorganic aerosols (SIA), secondary organic aerosols (SOA) from terpenes, heavy metals and POPs. The LOTOS-EUROS model is in use to forecast air quality in the Netherlands and in Europe as part of the MACC project (www.copernicus-atmosphere.eu). The LOTOS-EUROS model is used for the assessment of air quality (Curier et al., 2012; Manders et al., 2009) and intercomparison with other chemistry transport models (Stern et al., 2008; Vautard et al., 2012). Furthermore, Manders et al. (2012) analyze the impact of different large-scale circulation on ozone and PM using the LOTOS-EUROS. Hendriks et al. (2013) applied LOTOS-EUROS to analyze the origin of ambient particulate matter (PM) concentration in the Netherlands.

However, there are still large uncertainties in simulating air pollutant concentration using LOTOS-EUROS. The strong local forcing due to emissions and the model uncertainties are the largest source of uncertainty when it comes to air quality forecasting(Curier et al., 2012). It has a systematic underestimation of PM10 concentrations, especially in the regions with high observed concentrations (Manders et al., 2009; Mues et al., 2012). The underestimation part of PM10 is largely associated with the non-modelled components and the components for which no observations are available. Resuspension of crustal material by wind or traffic, secondary organic aerosols are not yet incorporated in the operational model version of LOTOS-EUROS. Due to the lack of measurement data, some important source areas for wind-blown dust are usually not included in the model domain. Furthermore, the LOTOS-EUROS model also underestimates the secondary inorganic aerosols about 30% (R.J. Wichink Kruit et al., 2012). The uncertainties of tropospheric ozone concentrations depend on its own initial concentrations and the concentrations of the precursor species which leads to its production or destruction, such as NO_X (Curier et al., 2012).

In our study, we use version 1.8 of the LOTOS-EUROS model. One default domain of LOTOS-EUROS, used in the MACC project, is over Europe with the border of latitude 35° N and 70°N, and longitude 15°W and 35°E (Figure 1). Its standard resolution is 0.5°longitude * 0.25°latitude, which is about 30 by 30 km, and it can be increased up to a factor 8 respectively. The vertical extent of LOTOS-EUROS model is 3.5 km over the sea level divided into a surface layer and 3 dynamic layers. The surface layer has a fixed height of 25m. The lowest dynamic layer is the mixed layer and there are two reservoir layers on top. The height of mixed layer is equal to the boundary layer height derived from the input meteorological data. The difference between the top and mixed layer height is separated equally into two reservoir layers with a minimum thickness of 50m. The output data is provided on the model levels, with a separate output file for the surface concentration of trace gases and aerosols at measuring height of 3.6m. The time resolution of the output is hourly. The details of chemistry mechanism and emissions sources can be found in the LOTOS-EUROS v1.8 Reference guide (Sauter et al., 2012).

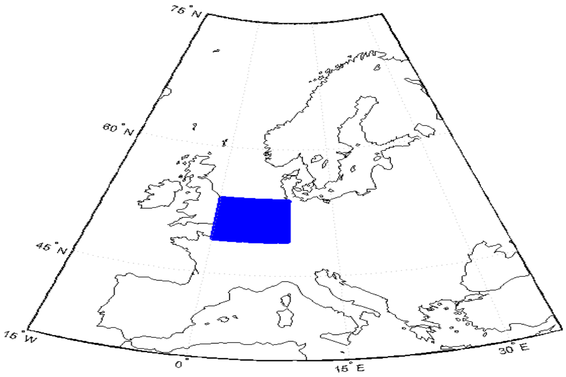


Figure 1 The domain of LOTOS-EUROS used to generate forecasts for the MACC project (black), and the target domain over the Netherlands used in our study (blue inset). The blue area is the target domain for our study.

The present study is based on two LOTOS-EUROS v1.8 runs, both on the target domain (0°E- 11°E, 49°N- 55°N, see Fig. 1) and both at a resolution of 3.5 km, with ECMWF and HARMONIE meteorological input respectively. The time period is from June to August, 2012. The boundary conditions at the outer boundaries of the target domain (see Fig. 1) are taken from the operational LOTOS-EUROS MACC forecasts over a large European domain [(15^0W, 35^0N) – (35^0E, 70^0N)] at a coarse resolution of 15 km (0.25°longitude*0.125°latitude). These MACC domain forecasts are produced using ECMWF meteorology fields (Figure 1). The only difference between these two runs is the meteorological driver.

3. The impact of meteorology on air quality

Air pollutant concentrations are determined by the emissions, chemical and aerosol processes in the atmosphere, and by the meteorology (Manders et al., 2009; Mues et al., 2012; Pielke and Uliasz, 1998; Seaman, 2000). The meteorological variables influence the following processes: chemical reactions, transport, emissions and deposition. In the LOTOS-EUROS model (Sauter et al., 2012), cloud cover, wind speed, boundary layer height, temperature, relative humidity, friction velocity and precipitation play important roles in determining the concentration of the air quality components. The basic relations between the air quality processes and meteorological variables are shown in Figure 2.

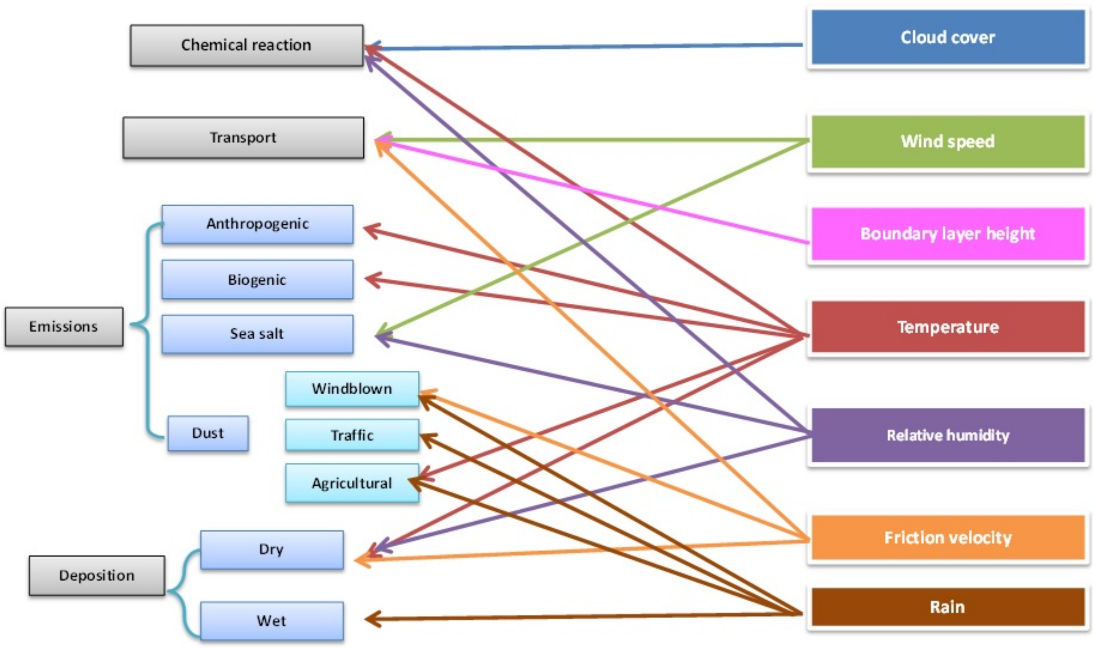


Figure 2. The relations between air quality processes with meteorological variables in the LOTOS-EUROS model.

The chemical reactions are directly influenced by temperature, humidity and cloud cover. The chemical reaction rates are in general temperature dependent, which holds for the gas-phase chemical reaction and the volatile species gas-particle equilibrium. Relative humidity affects the photochemical reactions between water vapor and oxygen radical, which in turn influences the concentration of the hydroxyl radical. Under the condition with proper concentration of nitrogen oxides, water vapor also affects the ozone concentration. Cloud cover directly influences the photolysis by decreasing the light intensity.

The transport has three parts in the LOTO-EUROS: advection, entrainment and vertical diffusion. The advection is driven by the wind field. For example, the NO concentration near an emission source could be underestimated due to strong advection transporting the NO outside of this region. The entrainment depends on the boundary layer development. The vertical diffusion is related to the friction velocity.

The meteorological variables also have an impact on the emissions. In LOTOS-EUROS, besides forest fire emissions, the residual emissions are divided into 4 parts: anthropogenic sources, biogenic emissions, sea salt, and dust sources. Among the anthropogenic emissions, a temperature-dependent factor is applied to the emissions of volatile organic compounds (VOC) and CO in road transport categories. The biogenic emissions are highly related to temperature and radiation. Surface temperature and rainfall gives a positive feedback to the soil nitrogen oxides (NO_x). Temperature strongly regulates vegetation VOC, especially when rising above 30 C. The sea salt generation is determined by the wind speed and relative humidity. The sea salt emissions depend on the surface friction covered with whitecap, which is modelled as a function of the wind speed at 10 m. The wet particle flux of sea salt is associated with the radius of particles influenced by relative humidity. The dust emissions include windblown dust, traffic generated dust and dust from agricultural land-management activities. The windblown dust is affected by the friction velocity and precipitation. The traffic-generated fugitive dust can be washed out by precipitation. The agricultural emissions are influenced by temperature and precipitation.

The deposition is the removal process of air pollutants in the air, which is defined as the process by which aerosol particles collect or deposit themselves on solid surfaces, decreasing the concentration of the particles in the air. The deposition includes dry and wet deposition. Dry deposition refers to acidic gases and particles, which is affected by temperature, relative humidity and friction velocity. Dry deposition depends on the aerodynamic properties and the deposition velocity (Zhang et al., 2001), and is normally modelled in terms of resistances. The deposition velocity contains components like the gravitational settling velocity, aerodynamic resistance and surface resistance. The aerodynamic resistance is computed from the friction velocity. The gravitation settling velocity depends on the diameter of the particles, in particular the particle diameter larger than 1um. The particle size is influenced by relative humidity, because particles can grow in high humidity conditions. The surface resistance is driven by the friction velocity and kinematic viscosity of air. The friction velocity is influenced by the wind speed. The kinematic viscosity of air is the dynamic viscosity divided by air density which is related to temperature and pressure. Thus, the meteorological variables determining the friction velocity, relative humidity and temperature have an impact on the dry deposition.

Wet deposition refers to acidic rain, fog and snow, which is related to the scavenging coefficient describing the rate of mass transfer of a contaminant from air into rain droplets. The scavenging coefficient depends on the rain rate, the size of particles, raindrop fall speed and a size-dependent collecting efficiency of aerosols by the raindrops with fine and coarse modes in LOTOS-EUROS. The approach used in LOTOS-EUROS v1.8 indicates that when the rain rate exceeds 2.5 mm/h, the coarse aerosols can be washed out from air within one hour. For the fine aerosol particles, the rain rate threshold is 10mm/hour.

Because the meteorological input data is one of the main causes of uncertainty in air quality transport model simulations (Stern et al., 2008), a lot of research focuses on the impact of meteorological variables on air quality in chemical transport models (Vautard et al., 2012). Manders et al. (2009) point out that the PM10 concentration decreases with the increasing wind speed and that easterly winds increase the concentrations in The Netherlands. Mues et al. (2012) indicated that the different meteorological variables and their impact on air quality is highly correlated. The daily precipitation sum, wind speed and daily maximum mixing layer height are correlated with the daily maximum temperature for the whole year. The maximum mixed layer height increases with daily maximum temperature linearly. The wind speed and precipitation are low at both very low and high daily maximum temperature. Both situations are characterized by high concentration of pollutants due to their influence on mixing, transport and deposition. For low temperatures, the concentration of secondary inorganic aerosols is inversely proportionally with temperature until the temperature arrives at 10-15 degree. As the temperature increases, the relations between concentrations of secondary organic aerosols (SOA) and temperature are different in different type of SOA. In the Netherlands, the concentration of sulfate increases with temperature, while the ammonium and nitrate concentration decrease.

Vautard et al. (2012) focused on the uncertainties of meteorological input used by air quality models, including the LOTOS-EUROS. They report the results from five meteorological models over North America and 11 models over Europe to simulate the weather in 2006. Temperature, shortwave radiation, wind speed, boundary layer height, relative humidity and precipitation were compared. They found the temperature was simulated well, while for the other variables there are still significant uncertainties. The shortwave radiation at noon varies with a factor up to two. The surface wind speed is generally overestimated over the Europe. The boundary layer height is poorly simulated at night and during the night-day transition time by models. The relative humidity and precipitation also have large uncertainties in the meteorological models. The occurrence of non-precipitation and extreme precipitation is underestimated while the overestimation of light to moderate precipitation could lead to an overestimate of the wet deposition in the air quality models. Thus, the uncertainties of the meteorological forecasts have a large impact on the air quality situations as the air quality processes are strongly influenced by the meteorology.

4. Meteorology models

4.1. ECMWF

The ECMWF (European Centre for Medium-Range Weather Forecasts) is providing global scale weather forecasts which are widely used at the European National Meteorological Services. Many regional atmospheric models and chemistry transport models use the ECMWF forecasts as boundary conditions or as driver (Awan et al., 2011; Del Genio and Wu, 2010; Flemming et al., 2009; Navascues et al., 2013). The ECMWF data is the default meteorology input for LOTOS-EUROS. In our study, we use the operational data from ECMWF with an interval of 3 hours. The meteorological fields are extracted from the ECMWF archive with a horizontal resolution of 0.5° longitude by 0.25° latitude (approximately 30 km). The fields are interpolated horizontally onto the LOTOS-EUROS grid, interpolated vertically onto the LOTOS-EUROS layers from pressure levels, and interpolated linearly in time to each hour.

4.2. Harmonie

The mesoscale Harmonie model is a non-hydrostatic spectral model with a high resolution of 0.037°longitude*0.023°latitude (2.5 km), which is used within the Hirlam/ALADIN community. Since 7th December 2012, Harmonie produces operational forecasts at KNMI. Harmonie has been observed to be very efficient in resolving deep convective processes. However, it also has weaknesses such as an over-prediction of fog and low clouds over the sea. Basically, Harmonie uses the same parametrizations as the AROME model (http://www.cnrm.meteo.fr/arome/). Either HIRLAM or ECMWF forecasts are used as boundary conditions for Harmonie. More information about the Harmonie model can be found on this website: http://hirlam.org/index.php?option=com_content&view=category&layout=blog&id=49&Itemid=102. The time interval of Harmonie is one hour. Compared to ECMWF, Harmonie has high temporal and spatial resolutions and shows more details of meteorological variables due to a partially explicit description of convection and turbulence, and due to a better resolved landuse.

5. Observations

5.1. Air quality observations

The National Air Quality Monitoring Network (LML) of RIVM measures several air pollution components including ozone, PM10, black carbon, NO, and NO_2 (http://www.lml.rivm.nl/data/gevalideerd/index.html). The measurements of ozone, NO, NO_2 are provided hourly, while for PM10 and black carbon, the stations provide daily average concentrations. For the period June to August 2012, there are 12 stations in the Netherlands reporting PM10 data, and 10 stations measuring black smoke, while ozone was measured at 37 locations. The stations are grouped in three types: regional, urban and traffic. Normally, the regional stations are used to evaluate air quality models, because they are far away from urban areas and highways and not influenced by local emissions which cannot be resolved by the model.

For this research, we choose 8 stations spread over the Netherlands (Figure 3): Vredepeel, Braakman, De Zilk, Wieringerwerf, Wekerom, Valthermond and Kollumerwaard, Cabauw. All these stations have observations of ozone and PM10. Except for Braakman and Kullumerwaard, these stations also provide measurements of black smoke.

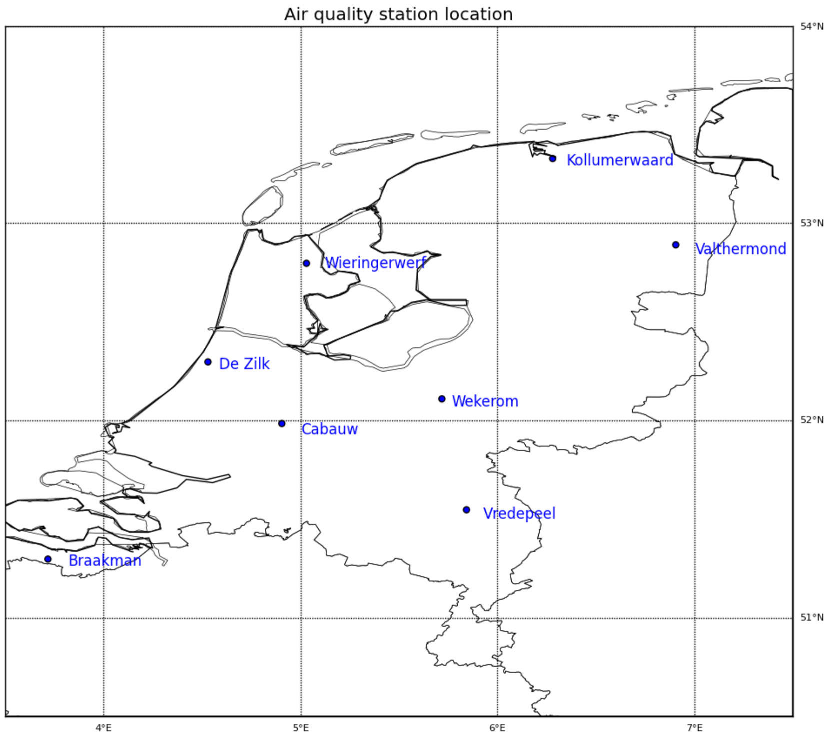


Figure 3. The air quality monitoring stations in the Netherlands selected for this research. All these stations have observations of ozone and PM10. Except for Braakman and Kollumerwaard, these stations also provide measurements of black smoke.

5.2. Meteorological observations

The Royal Netherlands Meteorological Institute (KNMI) has 36 stations providing hourly weather data (http://www.knmi.nl/klimatologie/uurgegevens). For this study we chose the 9 meteorological stations shown in **Fout! Verwijzingsbron niet gevonden.**. The criterion was that they should be close to the selected air quality stations. As we mentioned in section 3, the important meteorological variables affecting air quality processes in LOTOS-EUROS model are: temperature, relative humidity, precipitation, wind, boundary layer height, cloud cover and friction velocity. The friction velocity is not a measured variable and only Cabauw has observations of boundary layer height among these selected stations. We compared the forecast results of the ECMWF and Harmonie models with the observations at these 9 stations. To evaluate the boundary layer height of the two models, we use the data from another station Cabauw located in the central-western part of the Netherlands (51.971° N, 4.927° E).

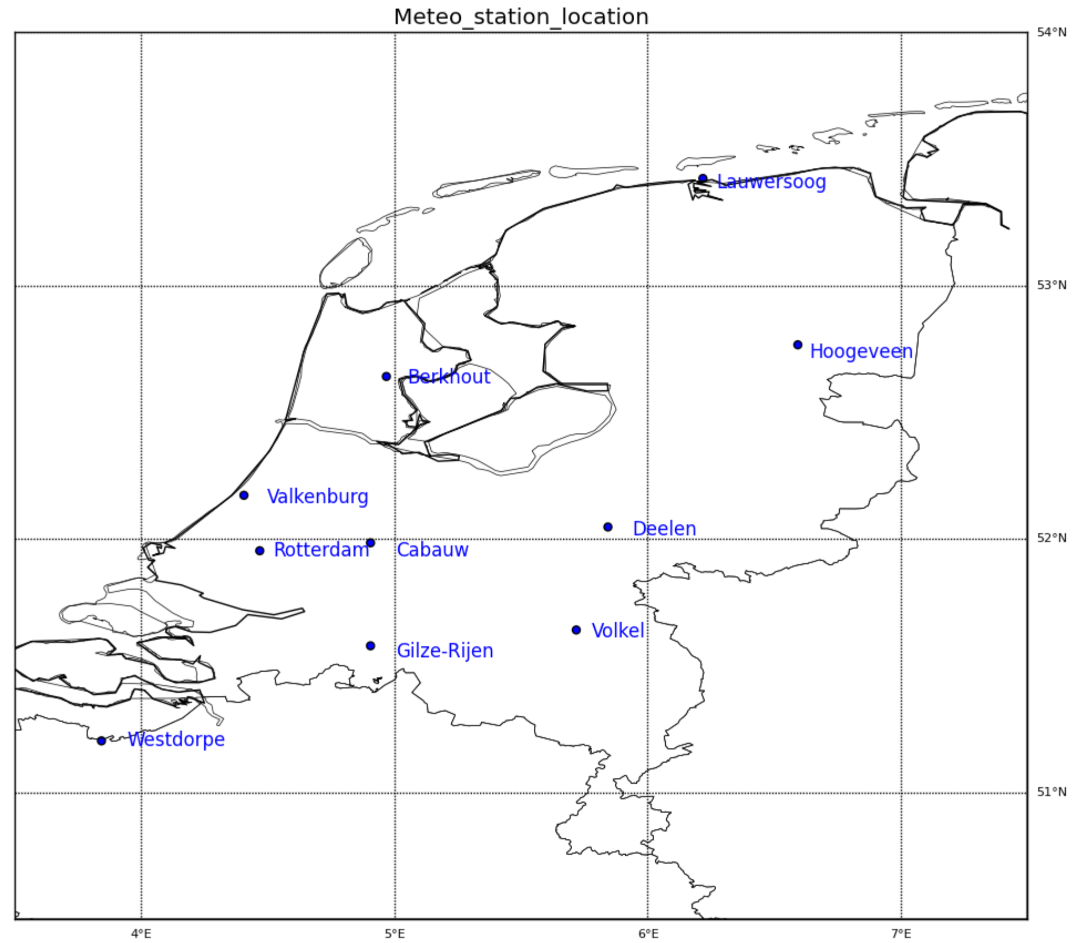


Figure 4 The selected meteorological observation stations in the Netherlands. Observations of the boundary layer height are available at the station Cabauw.

6. Methodology of statistical analysis

To assess results of models by comparing with the observation, we use statistical analysis by calculating the standard deviation (S), bias (B), root mean square difference (σ_{RMSD}), and correlation coefficient (r) averaged over the three month period(Wilks, 2006).

The standard deviation is calculated as:

$$S = \frac{\sqrt{\sum_{i=1}^{N}(x_i - \bar{x})^2}}{N}, \quad S = \sqrt{\frac{\sum_{i=1}^{N}(x_i - \bar{x})^2}{N-1}}$$

where *x* is the observation or the model result, $\bar{x}$ is the average of *x*.

The Bias is calculated as:

$$B = \frac{\sum_{i=1}^{N}(x_i - y_i)}{N}$$

where *x* and *y* represent the modelled and observed values respectively.

The root mean square difference is a measure of the differences between values simulated by a model and observation:

$$\sigma_{RMSD} = \sqrt{\frac{\sum_{i=1}^{N}(y_i - x_i)^2}{N}},$$

The correlation coefficient represents the linear relationship between a model and observation:

$$r = \frac{\frac{1}{N}\sum_{i=1}^{n}(x_i - \bar{x})(y_i - \bar{y})}{S_x S_y}$$

The correlation coefficient ranges between −1 and 1, where a value 1 or -1 indicate that x and y have perfect linear relation. A positive value of R means the two variables are proportional related while the negative value means the two variables are inversely proportionally related.

To analyze the difference of wind direction between observation and simulations, we use the same method as in De Meij et al. (2009). To evaluate wind direction, the mean absolute error (MAE) is used:

$$MAE = \frac{\sum DWD}{N},$$

where DWD is the difference of the wind speed and the DWD is calculated as:

$$DWD = \begin{cases} \min(Mod - OBS, OBS - Mod + 360), if\ Mod > OBS \\ \min\ (OBS - Mod, Mod - OBS + 360, if\ OBS > Mod \end{cases}$$

To assess the rainfall, rain specific hit rate statistics are used (De Meij et al., 2009):

		Observation	
		Yes	No
Model	Yes	A	B
	No	C	D

The probability of detection of the rain event is defined as:

$$\mathrm{POD} = \frac{A}{A + C}$$

The false alarm rate indicates the probability of false detection of the rain event:

$$\mathrm{FA} = \frac{B}{B + D}$$

The frequency bias represents the over- or underestimate of the events:

$$\mathrm{FBI} = \frac{A + C}{A + B}$$

If FBI =1, that means the event is simulated by the model exactly as often as it is observed. If FBI<1, it indicates the model is underestimate the frequency of the event and vice versa.

The Hansen Kuipers score is used to show the ability of the model to correctly simulate the events and to avoid false alarms.

$$\mathrm{HKS} = \frac{AD - BC}{(A+C)(B+D)},$$

When HKS is equal one, it means the model give the exact simulation as the observation. If HKS is close to zero or become negative, it means that the model simulates the event in a random way (De Meij et al., 2009; Wilks, 2006)

7. Comparison of the meteorological fields

7.1. Surface Temperature

We calculate the statistics of surface temperature at each meteorological station during June to August 2012. The differences of the average surface temperature between observation and both meteorological models are small, less than 1 K. The average surface temperature is underestimated 0.3 K in ECMWF while overestimated 0.2 K in Harmonie.

The Taylor diagram (Taylor, 2001) shows the capability of models to simulate the surface temperature (see Figure 5). The correlation coefficient of both two models is larger than 0.9 and the pattern RMSD of two models are both around 2K, which indicates that they can simulate the temperature very well. However, the correlation coefficient of Harmonie is a bit lower for ECMWF and the pattern RMSD of Harmonie for these 9 stations is larger than that of ECMWF. The ECMWF's result is a little bit better than Harmonie.

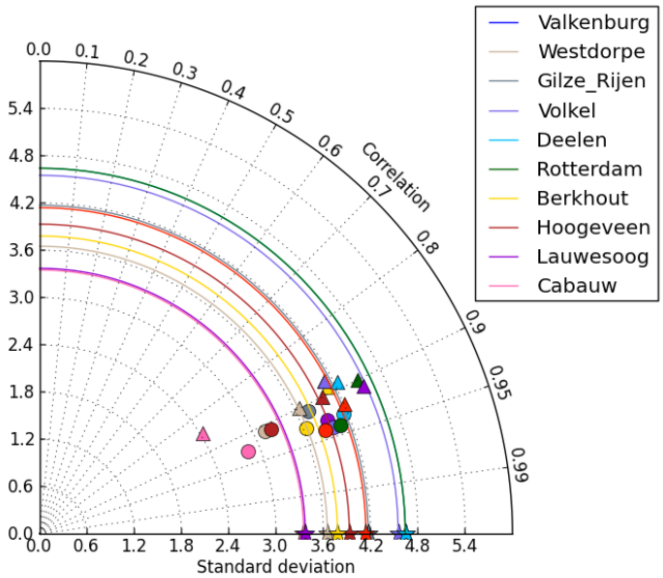
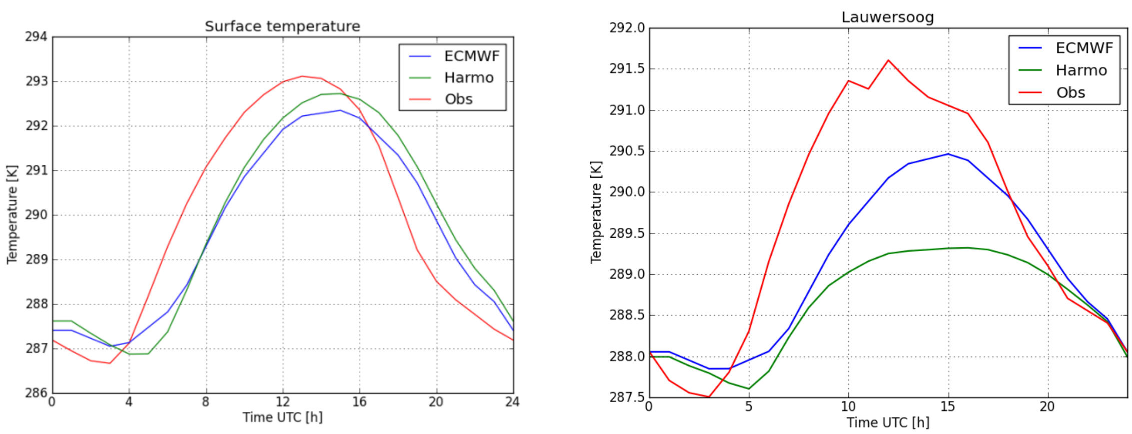


Figure 5. Taylor diagram of surface temperature [K] during period June to August, 2012. The stars are the observations; the circles correspond to ECMWF, and the triangles to Harmonie. The standard deviation is the radial distance to the origin. The distance between observation and model represents the pattern RMSD (in the same units as standard deviation which is K). The correlation coefficient between model and observation is given by the azimuthal position.

As the surface temperature has a diurnal cycle, we also calculate the average for each hour. Figure 6 (a) shows the average diurnal cycle of surface temperature for the 10 selected stations. The observations and the two simulations show a similar pattern. However, the morning increase of surface temperature in both models is two hours later than the observation, and the surface temperature of the two models is lower than the observation before the temperature arrives at the maximum. Most of the times, the surface temperature predicted by Harmonie is higher than ECMWF. The station Lauwersoog, located at the coastline of the northern part of Netherlands, is different from other nine stations. In Lauwersoog, both models underestimate surface temperature during the daytime (Figure 6 b). The underestimation can be as large as 1K by the ECMWF and 2K by the Harmonie.

Figure 7 shows the average model difference (left) and RMSD (right) of surface temperature at 12 UTC. The main differences in surface temperature between the ECMWF and Harmonie model occur near the coast and in urban areas. The RMSD of these two models is about 1.5 to 2 K. The difference in surface temperature between the two models is due to the high resolution in the Harmonie model, which has a more detailed description of land-use and of the land-sea transition.



(a) (b)

Figure 6. The average diurnal cycle of surface temperature during the period June to August 2012. (a) Avereage for 10 stations ; (b) Lauwersoog. The red line is observation data. The blue and green lines present the ECMWF and Harmonie respectively.

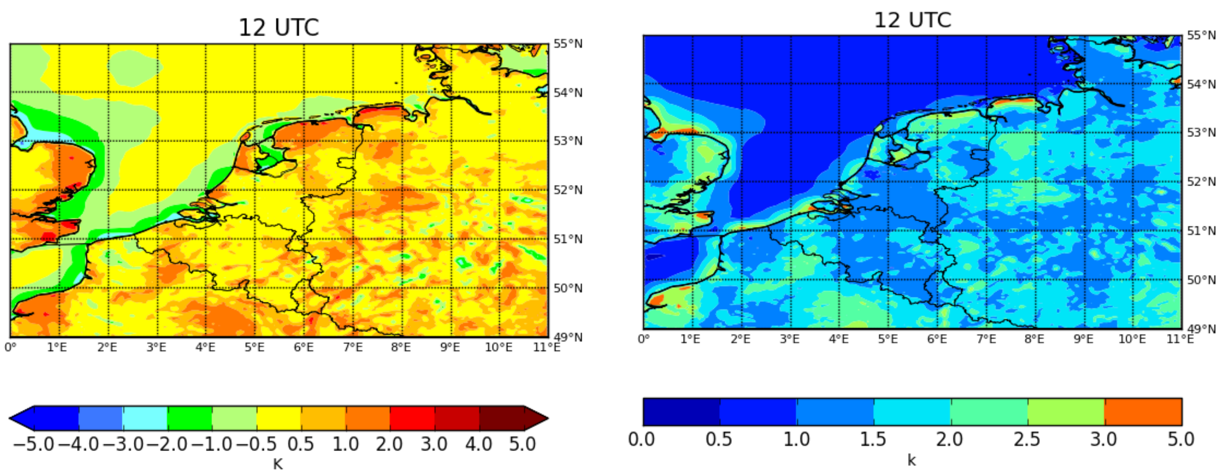


Figure 7. The average difference (Harmonie-ECWMF, a) and RMSD (b) in surface temperature (K) between the two models over the whole domain at 12 UTC from June to August 2012.

7.2. Relative humidity

Figure 8 shows the Taylor diagram of surface relative humidity at the selected stations. Compared to the ECMWF, the correlation coefficient in Harmonie is lower and the pattern RSME is larger. The pattern RSME of ECMWF is around 9%, while the pattern RSME of Harmonie is around 12%. The pattern of surface relative humidity simulated by ECMWF is closer to the observation. . In comparing high and low resolution models, the high resolution model may score worse simply because of more spatial variability. Thus, the ECMWF model has a better simulation of surface relative humidity.

Figure 9 shows the average daily evolution of relative humidity at the selected stations. After sunrise, as the temperature increases, the relative humidity decreases. Here the decrease in the model simulations happens also two hours later than the observation. During daytime, both models underestimate the relative humidity and during the night they overestimate it. As the relative humidity is related to temperature, the difference in relative humidity between the observation and model simulations is partly inversely related to the difference in surface temperature. The RMSD of these two models is around 10% over the land and 5% over the sea.

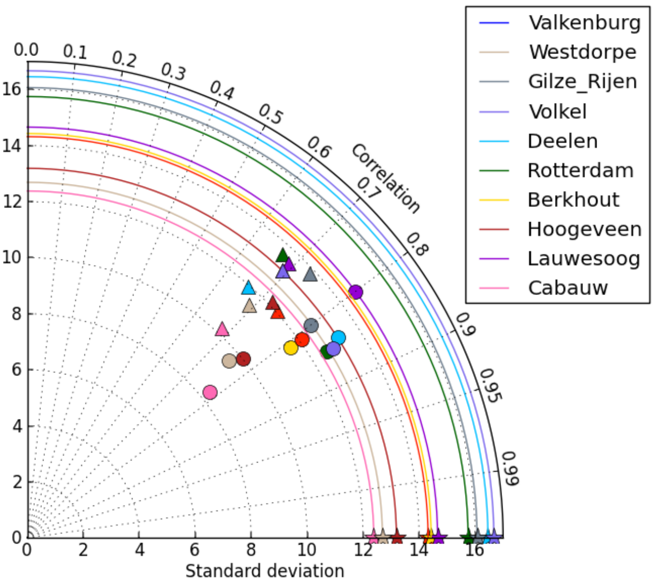


Figure 8. Same as figure 3, but now for relative humidity(%)

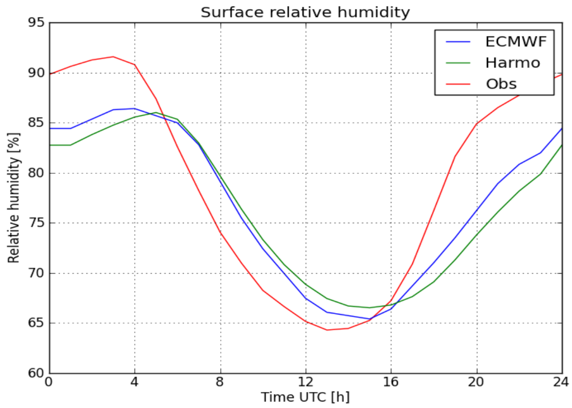


Figure 9. The average diurnal cycle of Relative humidity for the selected stations from June to August, 2012. The red line is observation data. The blue and green lines present the ECMWF and Harmonie respectively.

7.3. Wind speed and direction

In the Taylor diagram (not shown), the cloud of points of ECMWF and Harmonie largely overlap, which shows the similar capability to simulate the surface wind speed. The average of surface wind speed in ECMWF is 0.3m/s higher than the observation while it is 0.3m/s lower in Harmonie. Figure 10 shows the diurnal evolution of surface wind speed. The ECMWF analysis overestimates the surface wind speed during the night and the Harmonie underestimates it during the daytime.

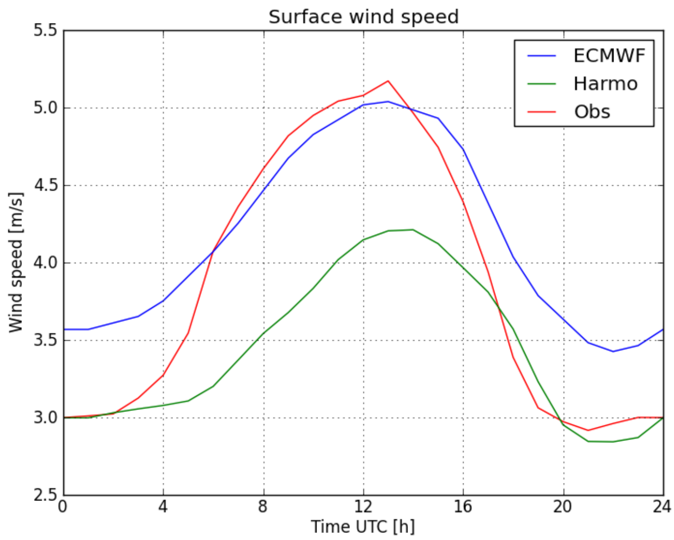


Figure 10. The average diurnal evolution of surface wind speed during the period from June to August 2012. The red line is observation data. The blue and green lines present the ECMWF and Harmonie respectively.

Figure 11 (a) shows the average difference of surface wind speed at midday. In the forest area, the surface wind speed modeled by Harmonie is lower as the forest can reduce the wind speed. The surface wind speed difference near the coast is also significant. Figure 11 (b) shows that the RMSD between the two models is about 1.5 to 2 m/s, which is about 20% of the mean surface wind speed.

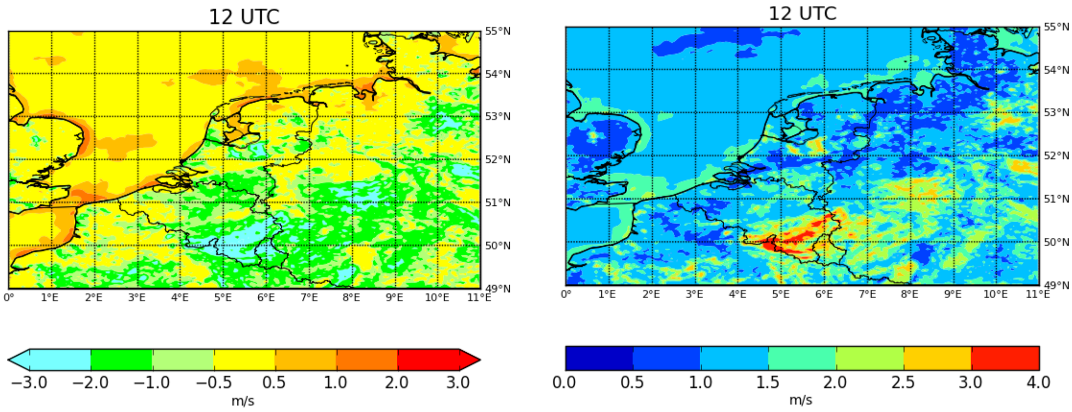


Figure 11. The mean bias (Harmonie-ECMWF, left) and RMSD (right) difference of surface wind speed (m/s) between the two models over the whole domain at 12 UTC from June to August, 2012.

Friction velocity (u*) is a measure of the turbulent momentum transport. In LOTOS-EUROS, the U* is computed from the Monin-Obukhov length and wind speed at 10 meters. And Monin-Obukhov length is also related to the cloud cover and wind speed. The friction velocity in the ECMWF model is higher than in Harmonie, which is consistent with the wind speed comparison. Thus, the turbulent transport of the simulation driven by the Hamonie model is weaker than for the ECMWF model.

The wind direction is as important as the wind speed for air pollution levels. Table 1 shows the mean absolute error (MAE) of the wind direction of ECMWF and Harmonie at surface layer. The MAEs of these two models are almost the same, which is around 20 degree. The mean absolute difference (MAD) between Harmonie and ECMWF is also around 20 degree (Figure 12). The capacity in forecasting the wind direction of the two models is similar.

Table 1. The Mean absolute error (MAE) of wind direction for each station during the period June to August 2012.

	Mean Absolute Error (MAE)	
Station	ECMWF	Harmonie
Valkenburg	27	26
Westdorpe	23	23
Gilze-Rijen	19	22
Volkel	17	20
Deelen	22	23
Rotterdam	27	27
Berkhout	22	23
Hoogeveen	21	24

Lauwersoog	21	22
Cabauw	19	21

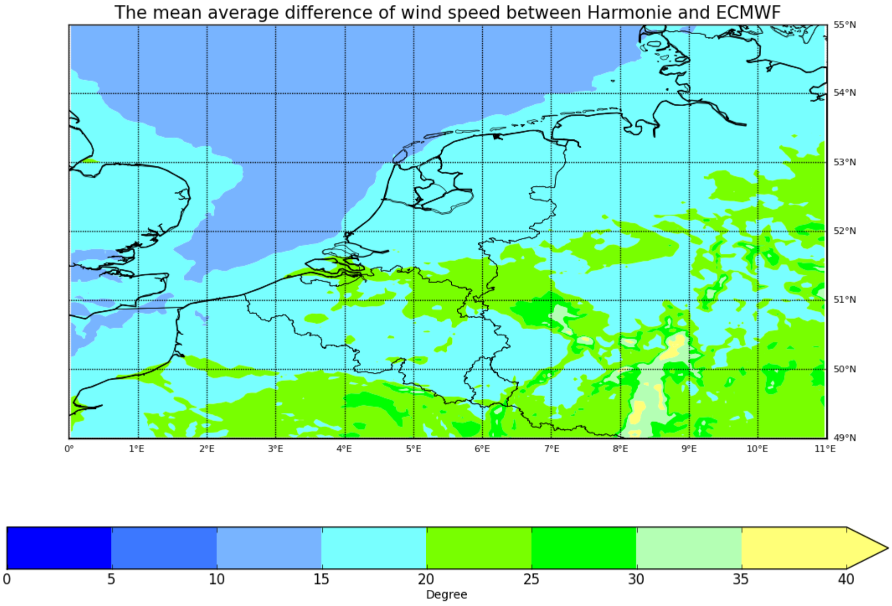


Figure 12. The mean absolute difference in wind direction between the ECMWF and Harmonie from June to August, 2012.

7.4. Rainfall.

Table 2 shows the average hit statistics of the rainfall from June to August 2012 at the 10 selected stations. The probability of detection (POD) of a rainfall event for ECMWF is higher than that of Harmonie with a factor of two, and the false alarm (FA) rate of ECMWF is also higher. For the frequency of rainfall occurrence (FBI), the ECMWF overestimates the rainfall event while the Harmonie underestimates it. These differences are partly due to the different time intervals in the output data files. In the ECMWF meteorological fields, the rainfall is accumulated in every three hours, while in the Harmonie data, it is available every hour. In the ECMWF simulations, if there is a rainfall event simulated, we assume that in the three hours, it has been raining, which makes the POD, FA and the rainfall frequency higher for an amount large than 0mm. The Hensen Kuiper Score (HKS) of ECMWF is closer to 1 which indicates that the ability of this model to give the correct rainfall occurrence is better than for Harmonie.

Table 2. The average hit statistics of rainfall during the period June to August 2012 at 10 meteorological stations.

	POD		FA		FBI		HKS	
	ECMWF	**Harmonie**	**ECMWF**	**Harmonie**	**ECMWF**	**Harmonie**	**ECMWF**	**Harmonie**
Occurrence	0.70	0.39	0.15	0.07	0.75	1.51	0.55	0.32
≥2.5mm/h	0.02	0.11	0.00	0.02	8.10	0.80	0.02	0.09

Both the ECMWF and Harmonie cannot simulate well a rainfall rate larger than 2.5 mm/h, especially the ECMWF. The values of POD and HKS are almost zeros, which mean that the prediction of the rainfall larger than 2.5 mm/h is random. The frequency of this rainfall rate, the ECMWF highly underestimates it and the frequency simulated by Harmonie is close to the observation. However, the timing of this high rainfall rate in Harmonie is not consistent with the observation. These differences in rainfall could affect the wet deposition, but if we have the long-lasting rainfall, the influence of different rain amount is not significant. For example, if it starts to rain, the aerosol in the air is washed out or rained out.

7.5. Boundary layer height.

We used observations of the boundary layer height at the station Cabauw. The boundary layer height in Cabauw is measured by a lidar cloud ceilometer (Haij et al., 2009) The technique, which measures the backscatter of light by aerosols is highly affected by the weather conditions. For example, precipitation, fog and clouds could complicate the measurement of boundary layer height. Thus, good weather conditions without rainfall or clouds and the presence of aerosols necessary for an unambiguous detection of the boundary layer height. The boundary layer height can be well detected at the day with the clouds above the boundary layer. During the period from June to August 2012, we found 5 days with good observations: 22 to 24 July, 10 August and 12 August.

The boundary layer height simulated by ECMWF during these days is much higher than that of Harmonie. Compared to the observation, ECMWF performs better (Figure 13). Figure 13a shows the diurnal boundary layer height for June 22, which is overestimated by ECMWF and underestimated by Harmonie. The transition from night to day for ECMWF happens at the same time as the observation. Figure 13b shows that ECMWF forecast almost matches the observation, but the transition time for this day is about two hours earlier than the observation. The Harmonie model still underestimates the boundary layer height but the transition time matches. For the five days with good weather, the boundary layer height simulated by ECMWF is closer to the observation. Harmonie underestimates the boundary layer height for all five days.

The comparison of the boundary layer heights of the two models and its average diurnal evolution shows that for ECMWF it is always higher than that for Harmonie during daytime. The map plots (Figure 14) shows the difference of average boundary layer height at four different hours. During the day Harmonie gives a lower value, especially over forest regions. During night, the boundary layer height simulated by Harmonie is higher than that simulated by ECMWF.

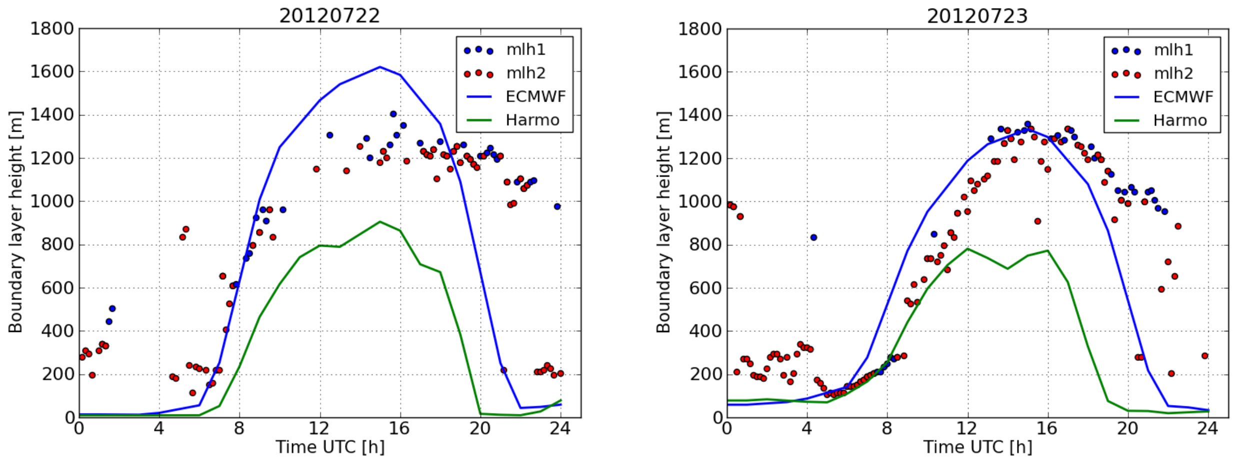


Figure 13 The diurnal boundary layer height. (a) 22 July, 2012; (b) 23 July,2012. (There are two ceilometers to detect the boundary layer height shown by dots. There are four levels of observation data quality. Only the observation data with highest quality level are used.)

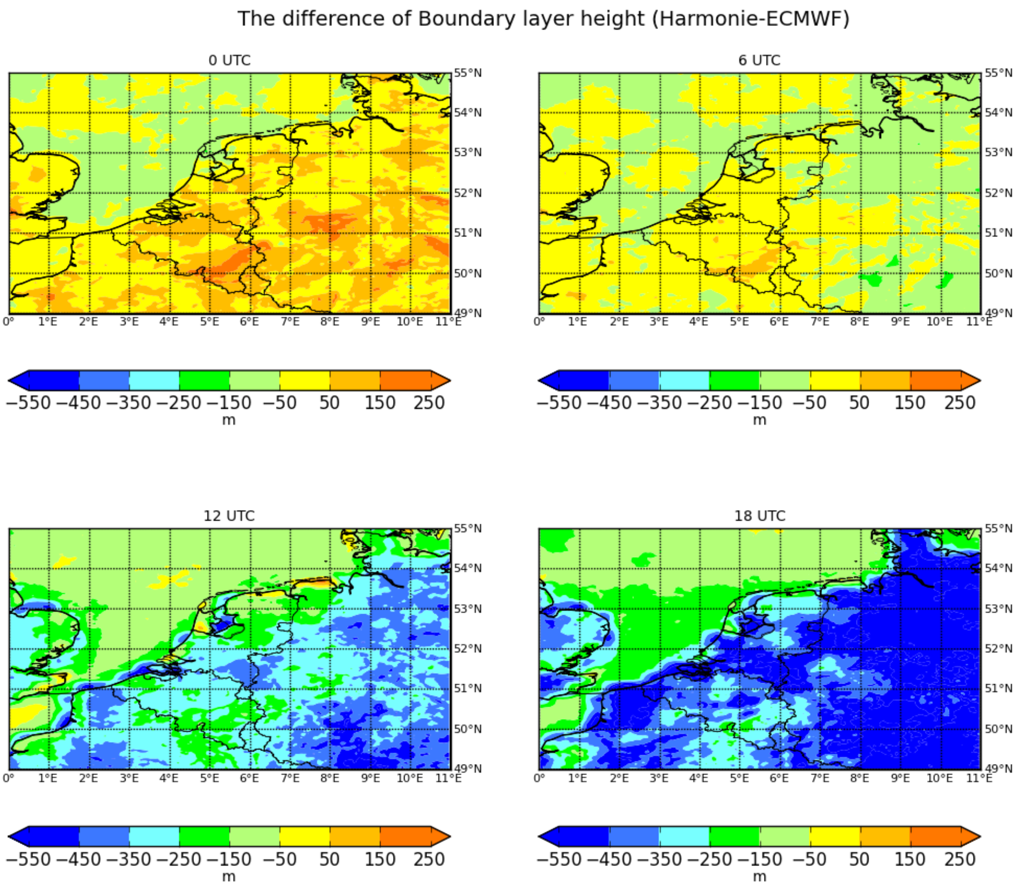


Figure 14 The difference of average boundary layer height (m) between ECMWF and Harmonie over the whole domain at 4 times (0, 6, 12, 18 UTC).

7.6. Total Cloud Cover.

The difference of cloud cover between the observations and models is large. The correlation coefficient between observations and the two models is around 0.6 and the RMSE is 30%. The average cloud cover for the three months simulated by the models is 5% higher than the observation. Compare to Harmonie, the ECMWF shows a little higher correlation coefficient and lower RMSE. Harmonie has a higher resolution, which gives more detail in the total cloud cover. The total cloud cover of Harmonie is about 10% higher than that of ECMWF, especially in the coastal areas.

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8. The impact of meteorological fields on air pollutant concentrations

In his section, a simulation of air pollutant concentrations with LOTOS-EUROS using Harmonie meteorological fields is compared with a simulation using ECMWF fields. All other settings of LOTOS-EUROS are kept the same. Concentrations simulated for the major pollutants are compared with observations in the Netherlands.

8.1. PM10

We compare the daily mean PM10 concentration of the two simulations with the daily-mean observations from June to August at 8 selected stations (Figure 15). Both simulations underestimate PM10 concentrations as we described in section 2. The difference between these two simulations of PM10 is small, and it is not obvious which simulation performs better. At most stations, the correlation coefficient is somewhat lower for Harmonie meteorology compared to ECMWF (Table 3), but at the station Kollumerwaard near the coastline and at the station Wekerom near forests, the correlation coefficient is higher for Harmonie. This may indicate a benefit of high resolution of Harmonie and the more detailed description of landuse.

Figure 16 shows that the root mean square difference (RMSD) of hourly concentration between the two simulations is large, around 4 ug/m3. Because the average PM10 concentration simulated by LOTOS-EUROS is about 10 ug/m3, the mean relative hourly difference is almost 50%.

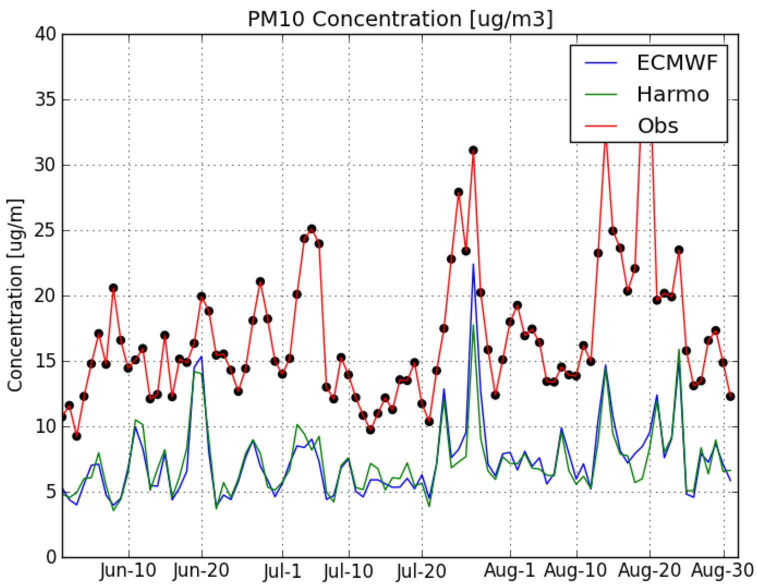


Figure 15. The time series of PM10 concentration from June to August 2012 at 8 selected stations. The red line is observation data. The blue and green lines present the simulations driven by ECMWF and Harmonie respectively.

Table 3. The correlation coefficient of PM10 between model simulations and observations at 8 stations.

Station	Correlation coefficient	
	ECMWF	Harmonie
Vredepeel	0.64	058
Braakman	0.62	0.53
De Zilk	0.39	0.36
Wieringerwerf	0.42	0.36
Wekerom	0.37	0.4
Valthermond	0.34	0.22
Kollumerwaard	0.41	0.49
Cabauw	0.51	0.48

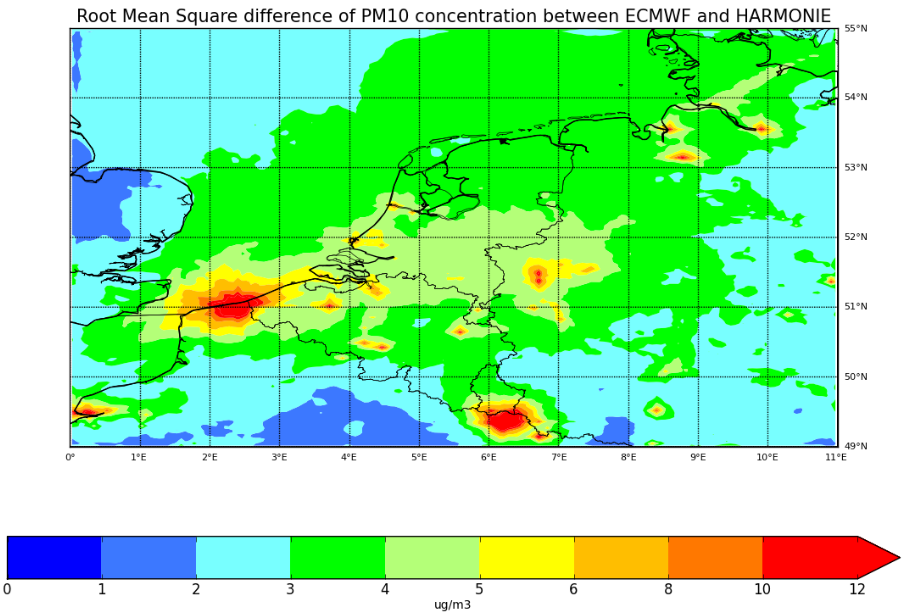


Figure 16. PM10 RMS difference between the two LOTOS-EUROS simulations driven by ECMWF and Harmonie.

8.2. Ozone(O_3)

Figure 17 shows that both simulations overestimate the O_3 concentration, in particular at night. The O_3 concentration in the runs driven by Harmonie is about 5 ug/m3 higher than the runs driven by ECMWF fields during daytime. The underestimation of boundary layer height in Harmonie could be one of the reasons. The RMSD of the hourly O_3 concentration between the ECMWF and Hamonie runs can be as much as 20 ug/m3 in the Netherlands (Figure 18).

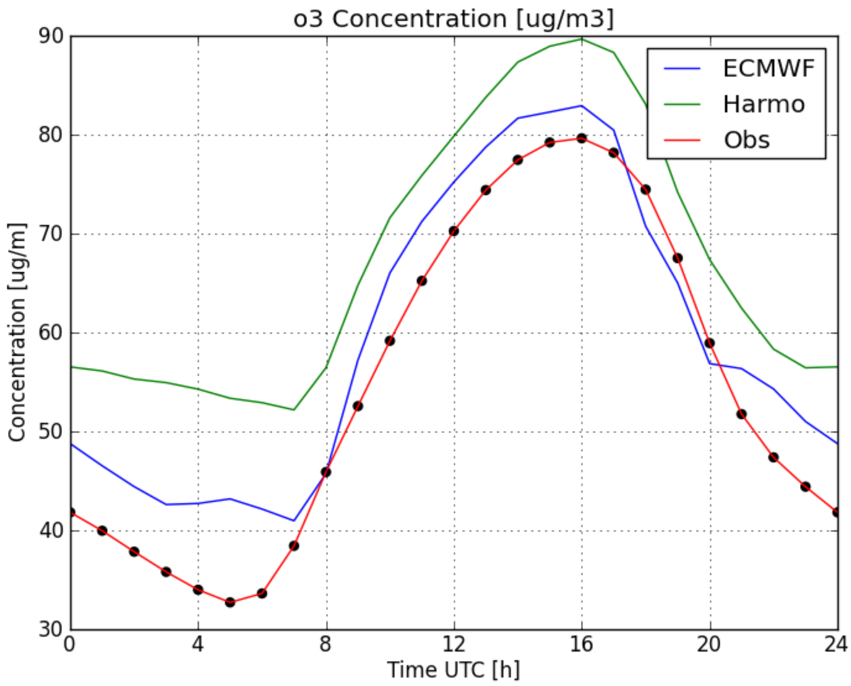


Figure 17. The average daily evolution of ozone at the 8 Dutch stations from June to August, 2012. The red line is observation data. The blue and green lines present the simulations driven by ECMWF and Harmonie respectively.

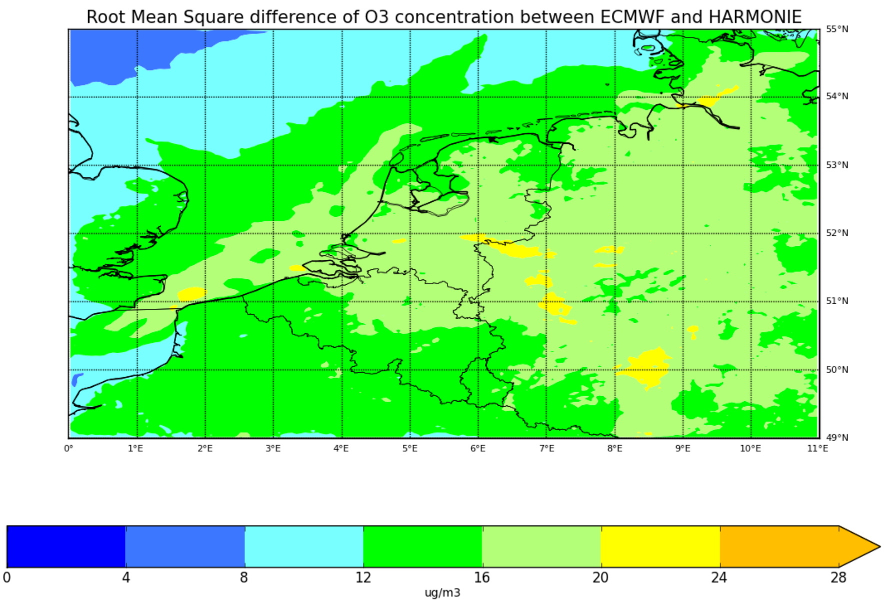
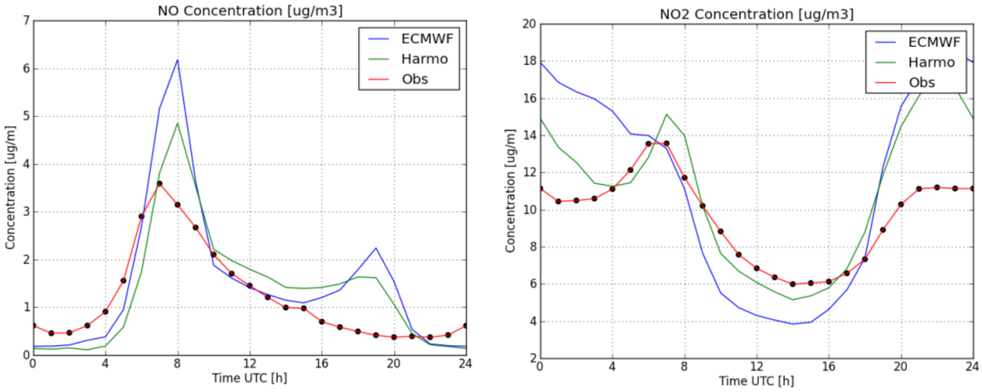


Figure 18. O_3 RMS difference between the two LOTOS-EUROS simulations driven by ECMWF and Harmonie.

Surface ozone is a secondary species, which is highly influenced by NOx (NO_X=NO+NO2). The higher O3 concentration of Harmonie can also be resulted in lower NO2 concentration during the night (Figure 19a). De Meij et al. (2009) also analyzed the daily evolution of O3 and NO_X concentration. The pattern of NO_X concentrations is similar to our model simulations. Due to the large uncertainties of NO_X measurement (Sluis et al., 2010), we cannot determine which model shows a better performance based on Figure 19 only. The RMSDs of NO and NO_2 are 6 ug/m^3 and 5 ug/m^3 between two simulations, respectively.



(a) (b)

Figure 19. The average daily evolution of NO (a) and NO_2 (b) concentration at the 8 Dutch stations for the period June to August, 2012. The red line is observation data. The blue and green lines present the simulations driven by ECMWF and Harmonie respectively.

9. Sensitivity analysis of air quality variables on meteorological variables

In the previous sections, we have compared meteorological fields of the ECMWF and Harmonie models as well as air pollutant concentrations from LOTOS-EUROS driven by the two meteorological models. To understand the individual contribution of each meteorological variable to the concentration differences observed, we have performed a sensitivity analysis for four meteorological variables, namely surface wind speed, boundary layer height, surface temperature and relative humidity. Table 4 shows adjustment factor in the sensitivity runs, and one of the meteorological input fields was increased and decreased by this amount. For the boundary layer height, we use the average difference as the adjustment factor. For the other three meteorological parameters, we use the regional mean RMS differences because their average differences between ECMWF and Harmonie are small. We use the Harmonie result as the control case. In total there 4x2=8 sensitivity runs performed.

Stern et al. (2008) pointed out that the special weather conditions lead to high concentrations of air pollutants. We chose the one week period of July 25 to 31 with relative high PM10 and O_3 concentrations, so that the influence of the different meteorological parameters during polluted conditions could be studied. For the following comparison of PM10, we show the relative changes of PM10 concentration at 12 UTC. During the night, the PM10 concentration is relative low compared to the daytime concentration. Thus, even small absolute changes could lead very high relative impact on the PM10 concentration at night.

Table 4. The adjustment of each variable used in the sensitivity experiments.

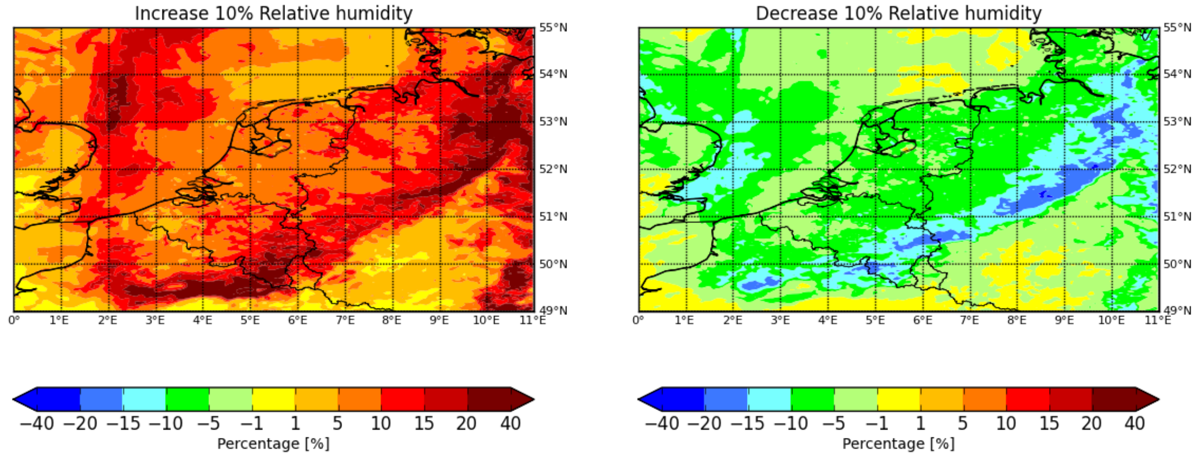
Meteorological variables	Surface Temperature	Surface Relative humidity	Wind speed	Boundary layer height
The change value	2K	10%	20%	30%

9.1. Sensitivity to surface temperature and relative humidity.

Based on the RMS difference values given in Table 4, we increase and decrease the surface temperature of the Harmonie output by 2 K and perform two runs with LOTOS-EUROS. We find that the relative change of PM10 and O_3 concentrations over the land are less than 1%, so that the influence of the difference in surface temperature between the two meteorological models is not significant. The temperature at upper levels, such as boundary layer may have an important influence on air pollutants concentrations.

In two additional sensitivity runs, we increase and decrease relativity humidity by 10% at all levels. The relative humidity is not changed when it is 100%. The result shows that when we modify the relative humidity at all levels in LOTOS-EUROS, the change in ozone concentration is not significant, but we can see a large change in PM10 concentration (Figure 20) as well as in the concentration of the gaseous precursors of secondary inorganic aerosols. If the relative humidity rises 10%, the PM10 concentration increases about 10%

over the domain. For some regions the increase is more than 40%. At the same time, the gas phase precursors decrease, in particular NH_3 (Figure 21). The increase of the PM10 concentration is due to the increase of secondary inorganic aerosols (SIA). As the relative humidity increases, some chemical reactions with H_2O are enhanced. For example, the NO and NO_2 will more rapidly react with H_2O to produce HNO_3 (Sauter et al., 2012). The oxidation of SO_2 will increase in high relative humidity conditions, which leads to more sulphate acid (De Meij et al., 2009; IPCC, 2001). Adding NH_3 neutralizes the nitric and sulphuric acid and leads to the production of additional ammonium, the decrease of NH_3 concentration and increase of ammonium. Thus, the relative humidity has a negative impact on gaseous precursors and a positive impact on SIA concentration, which is a major composition of PM10 over the Netherlands.



(a) (b)

Figure 20. The difference in surface PM10 concentration between the sensitivity experiments and the control result at 12 UTC for the period 25 to 31 July (Unit: %). (a) Increasing relative humidity by 10%. (b) Decreasing relative humidity by 10%.

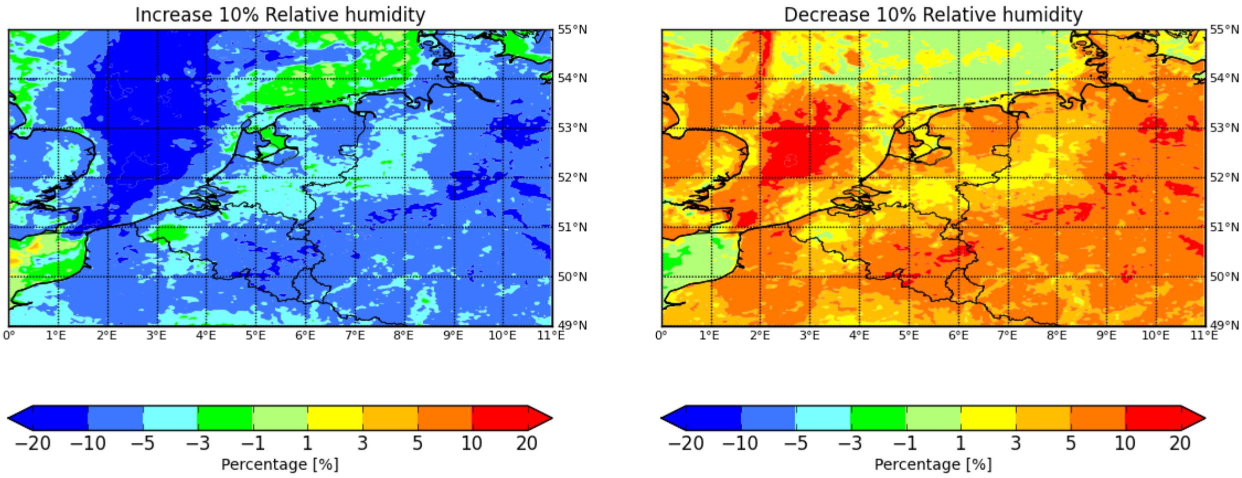


Figure 21. The difference in surface NH_3 concentration between the sensitivity experiments and the control result at 12 UTC for the period 25 to 31 July (Unit: %). (a) Increasing relative humidity by 10%. (b) Decreasing relative humidity by 10%.

9.2. The wind speed

Figure 22 shows the average change of PM10 concentration at 12 UTC during July 25 to 31 by modifying the wind speed by 20% in Harmonie. We can see that when the wind speed increases, the PM10 concentration will decrease by 5% over the land and increase by 40% over the sea and vice versa. The wind speed affects the dry deposition of PM10 (Figure 23). As the wind speed increases, the dry deposition also increases. The 20% change in wind speed could cause the domain about 20% difference in PM10 dry deposition, which in turn cause the decrease of PM10 concentrations. And the wind speed will also affect the emissions and other processes of air quality in the LOTOS-EUROS, so that the combined change of PM10 is only about 5%. Over the sea, the PM10 concentration is more sensitive to the wind speed, because a wind speed increase gives rise to additional sea salt emissions (Figure 22, Figure 24). Near the coastline, the sea salt concentration goes up 40% when the wind speed is increased by 20%. Over ocean, the increase in sea salt concentrations is even 100%. Actually, over the sea, the difference of wind speed between Harmonie and ECMWF is smaller than 20%. Therefore, the influence of wind speed uncertainties on sea salt is overestimated by our sensitive analysis. In summary, over the land, a wind increase has a net negative impact on PM10 concentration, while over the sea and near the coastline, it has a positive impact.

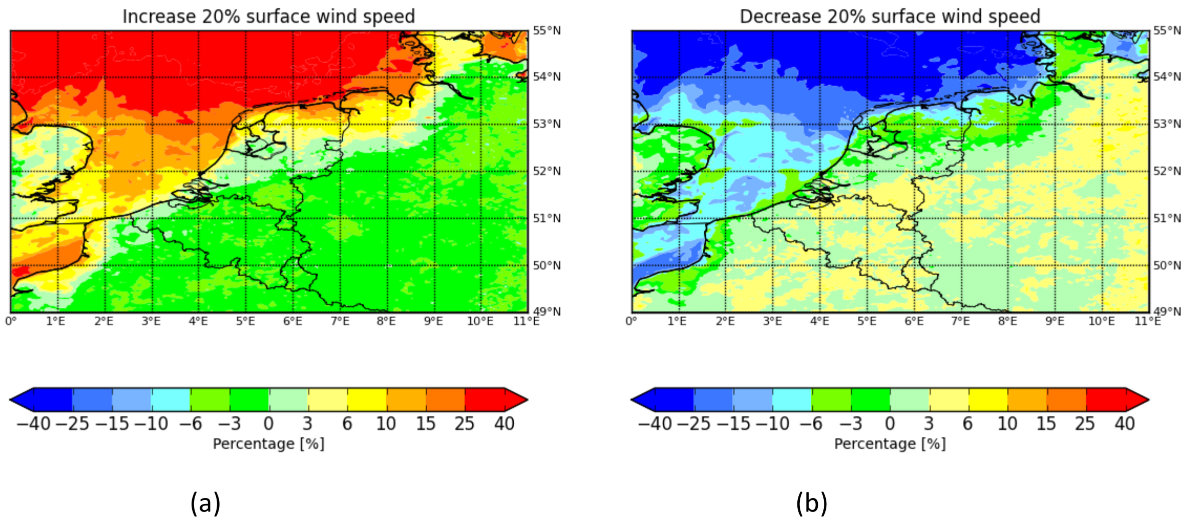
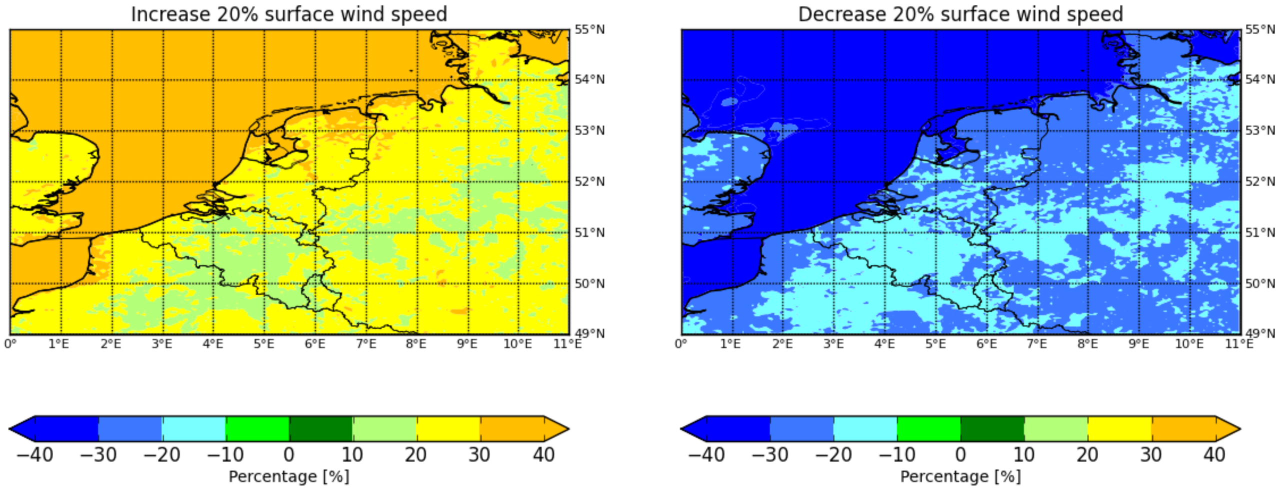


Figure 22. The difference in surface PM10 concentration between the sensitivity experiments and the control result at 12 UTC for the period 25 to 31 July (Unit: %). (a) Increasing surface wind speed by 20%. (b) Decreasing surface wind speed by 20%.



(a) (b)

Figure 23. The difference in PM10 dry deposition between the sensitivity experiments and the control result at 12 UTC for the period 25 to 31 July (Unit: %). (a) Increasing surface wind speed by 20%. (b) Decreasing surface wind speed by 20%.

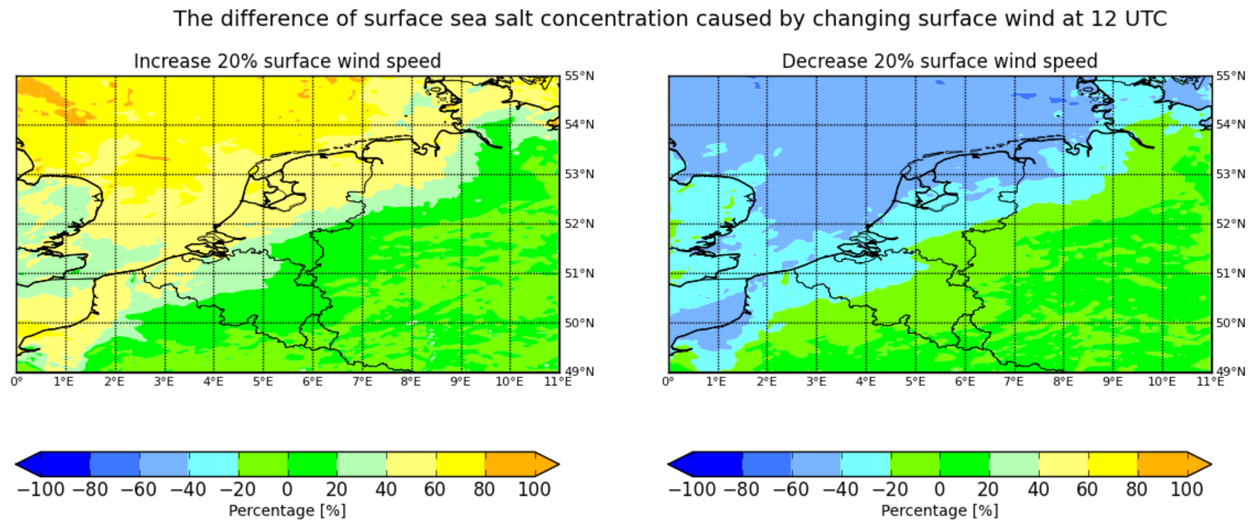


Figure 24. The difference in surface sea salt concentration between the sensitivity experiments and the control result at 12 UTC for the period 25 to 31 July (Unit: %). (a) Increasing surface wind speed by 20%. (b) Decreasing surface wind speed by 20%.

9.3. Sensitivity to boundary layer height

Due to the large systematic difference in boundary layer height between ECMWF and Harmonie forecasts (c.f. section 7.5), we changed the boundary layer height of Harmonie by +30%. Figure 25 shows that this leads to a 15% decrease of the PM10 concentration. With a higher boundary layer top, the PM10 released at the surface will be diluted over a larger vertical volume, leading to lower concentrations.

Figure 26 shows that O_3 increases when we increase the boundary layer height during the transition from stable to unstable in the early morning. At some regions, the increase of the O_3 concentration is even about 10%. When we modify the boundary layer height, we increase the boundary layer not only during the day but also during the night. If the boundary layer height is higher during the night, there could be more O_3 preserved in the boundary layer, which leads to higher concentrations in the morning. For example, near the coast, the processes determining the boundary layer development are complex near, and the exchange of air between land and sea is changing rapidly during the transition time.

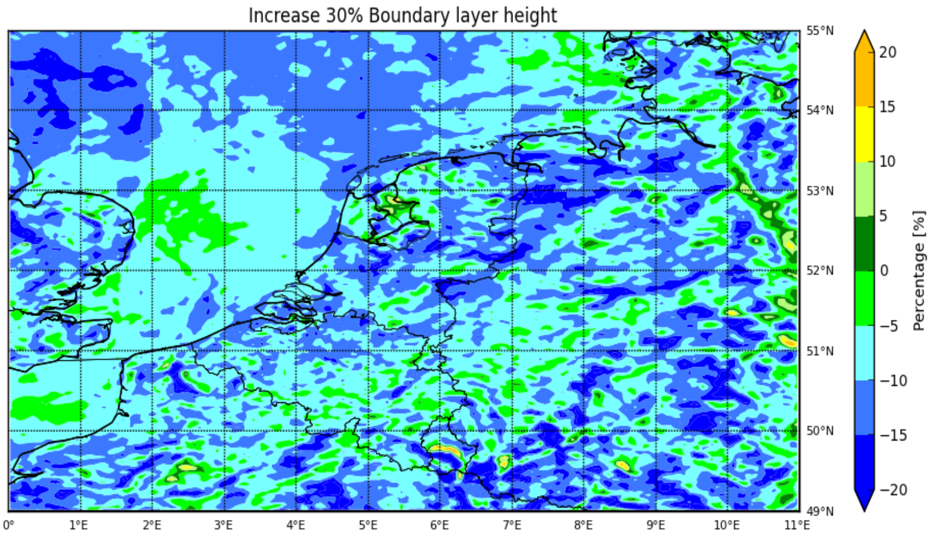


Figure 25. The difference in surface PM10 concentration between the sensitivity experiments and the control result at 12 UTC for the period 25 to 31 July (Unit: %). The sensitivity run is increasing the boundary layer height by 30%.

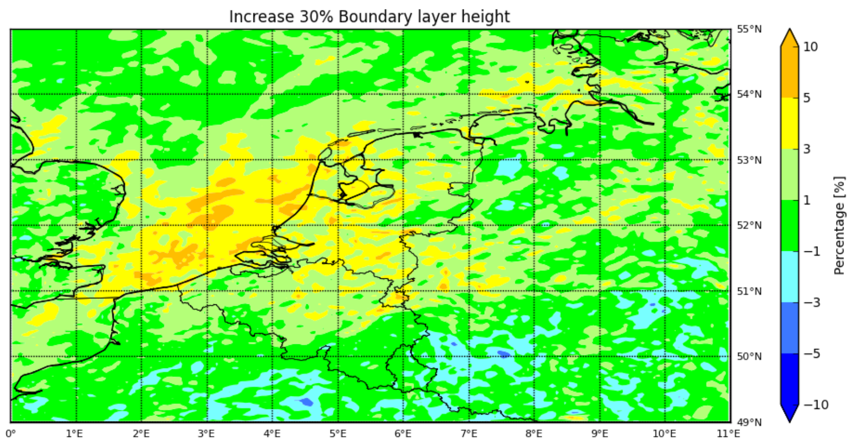


Figure 26. The difference in surface O_3 concentration between the sensitivity experiments and the control result at 12 UTC for the period 25 to 31 July (Unit: %). The sensitivity run is increasing the boundary layer height by 30%.

10. Discussion and Conclusions

In this study, we compared the air pollution concentrations over the Netherlands simulated by the LOTOS-EUROS model from June to August. 2012. The model is driven by ECMWF and Harmonie meteorological models. One main difference of these two models is the resolution: ECMWF has a coarse resolution of 16 km and Harmonie has a high resolution of 2.5km. Furthermore, Harmonie is a non-hydrostatic model which in part resolves the convective transport and cloud formation processes. First, we compared the meteorological variables of ECMWF and Harmonie. Second, we compared the air quality variables of the two LOTOS-EUROS simulations driven by ECMWF and Harmonie. To study the impact of individual meteorological variables on air quality as simulated the LOTOS-EUROS model, we set up a sensitivity analysis.

Our first research question concerns the difference in meteorological variables between the ECMWF and Harmonie models. The capability of these two models to simulate the surface temperature, relative humidity and wind speed observed in the Netherlands is similar. Due to the high resolution of Harmonie, in particular near the coastline, over forest and urban areas, there are apparent differences as compared to the ECMWF forecasts. This may be partly attributed to a better resolved description of land use. The RMS difference of surface temperature between the ECMWF and Harmonie is small, about 1.5 to 2 K. The relative humidity, wind speed and cloud cover in Harmonie differ considerably from ECMWF, by more than 10 %. The ECMWF has a better ability to simulate the rainfall occurrence than the Harmonie, but neither ECMWF nor Harmonie can give a good prediction of the high rainfall rates observed, which are underestimated especially by the ECMWF related to averaged cloud properties and low resolution. The boundary layer height simulated by Harmonie is much lower than for either ECMWF or ceilometer observations. Apart from the boundary layer height, it is hard to conclude which meteorological model gives better scores. High-resolution model Harmonie has better simulation in local weather forecast(Navascues et al., 2013). In comparing high and low resolution models, the high resolution model may score worse simply because of more spatial variability.

Vautard et al. (2012) also found that the differences in temperature between different meteorological models are small, but the differences in wind speed are large. Our diurnal cycle plots of meteorological varables show that the morning increase of meteorological variables in both models is two hours later than the observation, which can also be seen in the diurnal cycle plots of surface temperature and wind speed in Vautard et al. (2012). The boundary layer height of Harmonie is much lower than observed, pointing at a difficulty in simulating the boundary layer development. Many other meteorological models also underestimate the boundary layer height due to uncertainties in atmospheric dynamic variables, such as surface fluxes (Appel et al., 2010; De Meij et al., 2009). Using different boundary layer schemes leads to differences in boundary layer height, which can be as large as 50% (Mao et al., 2006; Xie et al., 2012). In my master thesis, I found that even in runs with the same model for the same period, but using different methods to calculate the boundary layer height such as parcel method that the temperature of boundary layer is equal to the surface temperature, the boundary layer height is substantially different(Ding; et al., 2013; Hennemuth and Lammert, 2006). In the present study we did not analyze the reason why Harmonie highly underestimate the boundary layer height, but we would like to encourage the people involved in the Harmonie development to have a closer look at the development of the boundary layer.

The second research question addresses the difference in air pollutant concentrations of two simulations driven by ECMWF and Harmonie respectively. The average difference in air pollutant concentrations is small. However, due to the differences of these meteorological drivers, the air pollution concentrations in LOTOS-EUTOS also show large differences. We calculated the root mean square difference between the two simulations for four main air quality variables: the RMS difference is 4ug/m3 for PM10; 20ug/m3 for O_3; 6ug/m3 for NO_2; 5ug/m3 for NO. And the relative RMS differences of the four air pollutant concentrations are above 30%.

In our study, the bias, or average difference, in the concentrations of air pollutants between the two simulations driven by ECWMF and Harmonie are not significant. Therefore we mainly focus the RMS difference to compare the two meteorological models. Please note that the RMS difference can be large if there are a few cases showing more extreme differences, which may be the case given the high resolution of Harmonie. The RMS difference does not represent the average difference very well in such cases. Using mean absolute difference (MAD) instead of the RMS may improve the interpretation of the results.

The last research question addresses how a perturbation of one of the meteorological variables affects the pollutant concentrations in LOTOS-EUROS. From the sensitivity analysis, we find that the effect of the 2K difference in surface temperature on air quality variables is small that the relative change of PM10 and ozone concentration is about 1%. As a result of a 10% increase in relative humidity, the concentration of PM10 will increase and the concentration of SIA gas precursors will decrease. The PM10 concentration is sensitive to the surface wind speed. Over the land, PM10 concentration decrease with the increase of the wind speed, while over the sea and the coastline, it will decrease. In the sensitive analysis of boundary layer height, we changed the boundary layer height uniformly for the whole day. The PM10 concentration decreases with higher boundary layer height. However, O_3 increases as a result of the higher boundary layer.

In the sensitivity analysis, we use the RMS difference to determine the adjustment factor in the sensitivity runs. We did not use the bias, because the mean difference between the two simulations driven by ECMWF and Harmonie is small. The sensitivity runs present the influence of how individual meteorological parameters affect air quality. We find that a surface temperature perturbation has a small impact on the concentrations of air pollutants. However, this does not mean that the temperature is not important: the surface layer is thin, and temperature perturbations at higher levels, in particular for the boundary layer, may be substantial. For instance Manders et al. (2009) have pointed out that SIA concentrations increase with temperature. Ozone is temperature-dependent air pollutants in summer time (Meleux et al., 2007; Ordonez et al., 2005)

11. Recommendations

From the conclusion, we know that the meteorological fields affect air pollutant concentrations. The uncertainties of surface wind speed can influence the PM10 concentration. The boundary layer height simulated by Harmonie is much lower than the reality, which has impact on all air pollutants concentrations. To improve the accuracy of meteorological fields is important to develop air quality prediction in LOTOS-EUROS.

Harmonie model highly underestimate the boundary layer height by using a TKE scheme. As different boundary layer schemes could lead difference on boundary layer height (Hu et al., 2010; Mao et al., 2006), we recommend using another scheme to calculate the boundary layer height and see if there is any improvement in simulating the boundary layer height.

As we show in the chapter 3, chemistry reaction, emissions, transport and deposition are all related to temperature. In temperature sensitivity runs, we only change the surface temperature. To improve the sensitivity analysis, we suggest to modify temperature at each layer to see how the temperature affect the concentration of air quality variables and on which layer the air quality variables are sensitive to temperature.

Our study only focuses on the summer time in 2012. However, weather conditions are different in winter, which could lead the difference impact on air quality simulation (De Meij et al., 2009). To extend this research, we recommend repeat this research for winter period.

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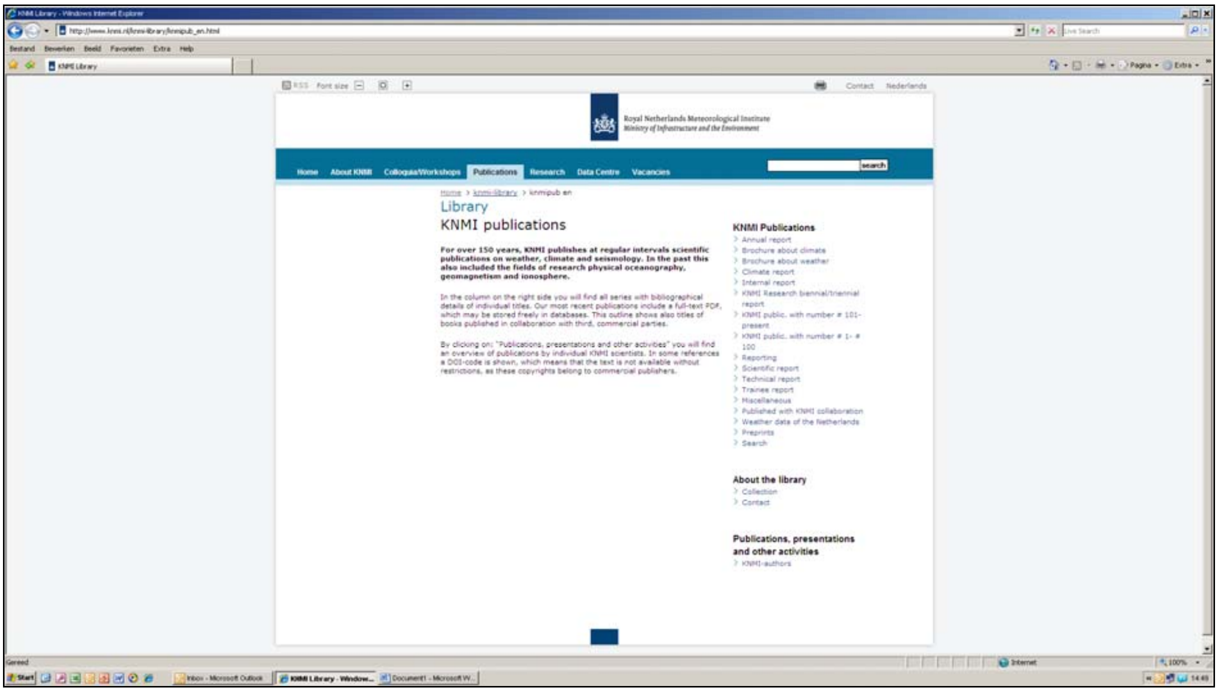

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